

UNIVERSITÉ DU QUÉBEC À MONTRÉAL

DISTRIBUTION OF COCCOLITHS AND ALKENONES IN RECENT AND LATE
QUATERNARY SEDIMENTS OF THE NORTHWEST NORTH ATLANTIC AS
PALEOCEANOGRAPHIC PROXIES AND INDICATORS OF PAST PRODUCTIVITY

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DISTRIBUTION DES COCCOLITHES ET ALCÉNONES DANS LES SÉDIMENTS
RÉCENTS ET QUATERNAIRES SUPÉRIEUR DU NORD-OUEST DE L'ATLANTIQUE
NORD – PROXIES PALÉOCÉANOGRAPHIQUES ET INDICATEURS DE
PALÉOPRODUCTIVITÉ

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DEDICATION

*My PhD is dedicated to my husband,
Aerion Snyder.
Thank you for always being my rock.
I love you.*

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TABLE OF CONTENTS

DEDICATION.....	iii
ACKNOWLEDGEMENTS.....	iv
TABLE OF CONTENTS.....	v
LIST OF FIGURES	viii
LIST OF TABLES.....	x
RÉSUMÉ	xi
ABSTRACT.....	xiii

INTRODUCTION	1
1. General objectives of this thesis	1
2. Geographic and hydrographic setting of the study areas	2
3. Previous research	4
4. Materials	6
5. Methodology	7
5.1 Coccoliths	7
5.1.1 Genetic variations of coccoliths.....	8
5.2 Alkenones	9
5.3 Foraminifers.....	12
5.4 Calcium carbonate preservation and dissolution indicators.....	13
6. Thesis structure	16
References	18
Tables.....	34
Figure	37

CHAPTER ONE	38
Abstract	39
1. Introduction.....	40
2. Regional context	42
2.1 Hydrography	42
2.2 Previous coccolith studies from the study area.....	43
2.3 Previous alkenone studies from the study area	44
3. Materials and Methods.....	45
4. Results.....	49
4.1 Coccolith distribution.....	49
4.2 Alkenones and organic carbon.....	50
4.3 Alkenone-coccolith relationships.....	52
4.4 Sea-surface properties versus alkenone and coccolith concentrations	53
4.5 Alkenone unsaturation and temperature	54
5. Discussion	54
5.1 Coccoliths and surface water mass properties	54

5.2 Coccoliths versus alkenones	57
5.2.1 Calcite dissolution.....	57
5.2.2 Lateral transport and allochthonous materials	59
5.2.3 Preferential degradation	60
5.2.4 Non-calcifying haptophytes	61
6. Summary and Conclusions	62
Acknowledgements.....	64
References.....	65
Figures.....	81
Tables.....	94
Appendices.....	101
Appendix A: Coccolith Taxonomy	101
Appendix B: Scanning electron micrographs of core-top sediments.....	102
Appendix C: Oceanographic parameters	111
 CHAPTER TWO	 112
Abstract.....	113
1. Introduction.....	114
2. Oceanographic setting.....	116
3. Materials and Methods.....	117
3.1 Coccoliths	119
3.2 Alkenones	119
3.3 Foraminifers.....	120
3.4 Calcium carbonate dissolution indices.....	120
4. The stratigraphical context.....	123
5. Results.....	124
5.1 Coccoliths	124
5.2 Alkenones	125
5.3 Foraminifers and dissolution indices	126
6. Discussion	128
6.1 Calcite preservation and IRD.....	128
6.2 Productivity.....	129
6.3 Decoupling of coccolith and alkenone abundances	130
7. Conclusions.....	133
Acknowledgements.....	135
References.....	136
Figures.....	147
Table	157
Supplemental figure and tables	158
 CHAPTER THREE	 165
Abstract	166
1. Introduction.....	167

2. Regional setting	168
2.1 Oceanographic context.....	168
3. Materials and Methods.....	170
3.1 Core location and stratigraphy	170
3.2 Coccolith analyses	173
3.3 Alkenones	175
4. Results.....	176
4.1 Coccolith concentrations and fluxes	176
4.2 Alkenone concentrations and fluxes	177
4.3 Correlations of <i>Emiliana huxleyi</i> and alkenones	178
5. Discussion	179
5.1 Coccoliths and alkenones in Baffin Bay	179
5.2 Coccolith and alkenone production across the northwest North Atlantic.....	181
5.3 Contrasting environmental conditions	183
6. Conclusions.....	184
Acknowledgements.....	186
References.....	187
Figures.....	199
Tables.....	205
Supplemental figures and tables	208
 SUMMARY & CONCLUSIONS.....	 218
References.....	223

LIST OF FIGURES

INTRODUCTION

Figure 1. Map of core site locations for all sediment samples used in this thesis.	37
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CHAPTER ONE

Figure 1. Map of the core site locations in the northwest North Atlantic.....	81
Figure 2. Coccoliths concentrations vs. distance to coast and CaCO_3 and in space.....	82
Figure 3. Coccolith concentrations and percentages of the main coccolith taxa	83
Figure 4. Results from the redundancy analysis (RDA)	84
Figure 5. Organic carbon, inorganic carbon, $\delta^{13}\text{C}_{\text{org}}$, alkenone concentrations, and $\%C_{37:4}$	85
Figure 6. Distribution maps of alkenone concentrations and $\%C_{37:4}$	86
Figure 7. Alkenone concentrations vs. $\%C_{\text{org}}$ and $\delta^{13}\text{C}_{\text{org}}$, and $\delta^{13}\text{C}_{\text{org}}$ vs. $\%C_{\text{org}}$	87
Figure 8. Concentrations of alkenones vs. coccoliths, and high alkenone concentrations map ...	88
Figure 9. Coccolith and alkenone concentrations vs. sea-surface parameters	89
Figure 10. Concentrations of alkenones and coccoliths, and sea-surface parameters vs. $\%C_{37:4}$.	90
Figure 11. Temperature differences	91
Figure 12. The residuals in sea-surface temperatures vs. $\%C_{37:4}$	92
Figure 13. Map of sites with calcite dissolution and calcite preservation	93
Figure B1. The core locations of the samples in the scanning electron micrographs.....	102
Figure B2. 2008 029 070 TWC 0-1 cm (#7 on map; Baffin Bay).....	103
Figure B3. 2008 029 008 TWC 0-1 cm (#11 on map; Davis Strait).....	104
Figure B4. 2013 029 0018 VVG 0-1 cm (#13 on map; northwest Labrador Sea).....	105
Figure B5. 2008 029 004 TWC 0-1 cm (#14 on map; northwest Labrador Sea)	106
Figure B6. 91045 028 BC 0-1 cm (#22 on map; central Labrador Sea).....	107
Figure B7. 91045 005 BC surface (#27 on map; Labrador Shelf).....	108
Figure B8. 91045 080 TWC 0-1 cm (#30 on map; central North Atlantic).....	109
Figure B9. 2009 061 0119 PSH 0-2 cm (#39 on map; southeast Labrador Sea).....	110
Figure C1. Oceanographic parameters.....	111

CHAPTER TWO

Figure 1. Map of the study area showing the core site, bathymetry, and surface currents	147
Figure 2. Age-depth relationship for core 004 based on the Bacon software.....	148
Figure 3. Percentages of CaCO_3 and IRD, and isotope stratigraphy ($\delta^{13}\text{C-Npl}$ and $\delta^{18}\text{O-Npl}$)..	149
Figure 4. Total coccolith concentrations and percentages of coccolith species	150
Figure 5. Concentrations and fluxes of total coccoliths, <i>Emiliania huxleyi</i> , and alkenones.....	151
Figure 6. Alkenone versus <i>Emiliania huxleyi</i> concentrations in samples from core 004	152
Figure 7. Concentrations and fluxes of organic linings and planktonic foraminifers.....	153
Figure 8. Planktonic foraminifer fragmentation index (%), B/P index, and log(OL/B)	154
Figure 9. Foraminifers, coccoliths, productivity, sea-ice cover, and $\delta^{13}\text{C-Npl}$	155
Figure 10. U^{K}_{37} -SST and MAT-SST	156

Figure S1. Scanning Electron Microscope (SEM) images of coccoliths from core 004	158
--	-----

CHAPTER THREE

Figure 1. Map of the five core site locations	199
Figure 2. Concentrations of alkenones and coccoliths from cores 174 and 170.....	200
Figure 3. Concentrations of alkenones and coccoliths from cores 004, 094, and 080.....	201
Figure 4. Fluxes of all coccoliths and <i>Emiliania huxleyi</i> in cores 004, 094, and 080	202
Figure 5. Alkenone fluxes for cores 004, 094, and 080.....	203
Figure 6. <i>Emiliania huxleyi</i> versus alkenone concentrations for all five cores	204
Figure S1. Age-depth relationship for core 004 based on the Bacon software.....	208
Figure S2. Age-depth relationship for core 094 based on the Bacon software.....	209
Figure S3. Age-depth relationship for core 080 based on the Bacon software.....	210

LIST OF TABLES

INTRODUCTION

Table 1. Examples of approaches used to study calcite dissolution	34
Table 2. List of surface sediment samples used in this thesis.....	35
Table 3. The long cores analyzed in this study, ordered by longitude (west to east)	36

CHAPTER ONE

Table 1. List of surface sediment samples used in this study	94
Table 2. Coccolith concentrations in the surface sediment samples.....	95
Table 3. Percentages of coccolith taxa.....	96
Table 4. Environmental data	97
Table 5. RDA results for 10 taxa, 6 environmental parameters, and 49 sites.....	98
Table 6. Distance to coast, alkenones, U^K , and organic and inorganic carbon content data	99
Table 7. Core-top sediment samples with at least one foraminiferal dissolution index	100
Table A1. Coccolith taxonomy of all species identified in our study.....	101

CHAPTER TWO

Table 1. Comparison and interpretation of the Bølling-Allerød and the deglacial interval	157
Table S1. Depth intervals, ages, sediment densities, and the sedimentation rates	159
Table S2. Concentrations of coccoliths, alkenones, foraminifers, and organic linings	160
Table S3. Fluxes of coccoliths, alkenones, foraminifers, and organic linings.....	161
Table S4. Percentages of coccolith species in the samples.....	162
Table S5. The calcite dissolution indices: fragmentation index, B/P, and $\log(OL/B)$	163
Table S6. The U^{K}_{37} , U^{K}_{37} -SST (Filippova, 2016), and the MAT-SST (Gibb et al., 2014).....	164

CHAPTER THREE

Table 1. List of cores analyzed in this study, ordered by longitude (west to east)	205
Table 2. Modern sea-surface conditions at/near the location of the study sites	206
Table 3. LGM sea-surface conditions at the study sites (core 004, 094, and 080 only)	207
Table S1. AMS- ^{14}C ages for cores 004, 094, and 080	211
Table S2. Core 174 data.....	212
Table S3. Core 170 data.....	213
Table S4. Core 004 data.....	214
Table S5. Core 094 data.....	215
Table S6. Core 080 data.....	216
Table S7. Age-depth windows for core 094	217

RÉSUMÉ

Cette thèse aborde le thème de la productivité primaire et de la préservation de la calcite dans le nord-ouest de l'Atlantique Nord et les bassins subpolaires adjacents, y compris la mer du Labrador et la baie de Baffin. Elle contribue à la compréhension des effets de changements climatiques extrêmes depuis la dernière glaciation sur le cycle du carbone à une échelle régionale. Pour ce travail de recherche, des analyses de coccolithes et d'alcénones ont été réalisées dans soixante-six échantillons de sédiments de surface et dans cinq carottes de sédiment couvrant les 25 000 dernières années. Les coccolithes, qui sont les restes calcitiques de représentants des haptophytes (organismes unicellulaires phototrophes), et les alcénones, qui sont des biomarqueurs organiques produits par certaines espèces d'haptophytes (notamment *Emiliania huxleyi*), fournissent des informations complémentaires sur la productivité des coccolithophores et la préservation du carbonate de calcium.

Une comparaison du contenu en coccolithes et alcénones des échantillons de surface avec des paramètres océanographiques de surface, incluant la température (SST), la salinité (SSS), la couverture de glace de mer et la productivité primaire, révèle des relations étroites entre la distribution des espèces de coccolithes et les propriétés des eaux de surface. Par exemple, *Coccolithus pelagicus* est étroitement associé à des faibles températures et des salinités relativement élevées. Dans certaines régions du nord-ouest de l'Atlantique Nord, la dissolution de la calcite a un impact sur la distribution des coccolithes dans les sédiments. Tel est le cas, dans la baie de Baffin, où la dissolution de la calcite s'initie dans la colonne d'eau, ainsi que sur le plateau continental du Labrador où la dissolution est probablement issue de la diagénèse oxydante du sédiment riche en matière organique. Par ailleurs, dans certains environnements néritiques, des haptophytes non-calcifiants produisent sans doute des alcénones, ce qui expliquerait les concentrations anormalement élevées obtenues dans le Chenal Laurentien ainsi que l'incohérence des températures U^K enregistrées le long la pente continentale de la Nouvelle Écosse et du bassin de Orphan Knoll dans la mer du Labrador.

Afin de documenter les changements de la production et de préservation des carbonates biogéniques, on a porté une attention particulière à la carotte sédimentaire HU2008-029-004 PC prélevée dans le nord de la mer du Labrador. Les résultats d'analyses de la séquence sédimentaire couvrant les 25 000 dernières années indiquent des changements importants de l'abondance des coccolithes, avec des concentrations plus faibles pendant le dernier stade glaciaire (moyenne de $9,5 \pm 3,9 \times 10^5$ coccolithes g^{-1}) que le postglaciaire (maximum de $3,1 \times 10^8$ coccolithes g^{-1}). Trois indices de dissolution de la calcite ont été utilisés afin d'évaluer la préservation des carbonates biogéniques : l'indice de fragmentation des foraminifères planctoniques, le rapport entre les foraminifères benthiques et planctoniques (B/P), et le rapport entre les réseaux organiques et les tests calcitiques des foraminifères benthiques (OL/B). L'indice de fragmentation est faible avec $7,1 \pm 0,8$ % pour la période glaciaire et $3,9 \pm 0,2$ % pour la période postglaciaire. Les valeurs B/P ($< 0,2$) et OL/B sont également faibles tout au long de la séquence étudiée. Ces indices indiquent une bonne préservation des carbonates biogéniques, en générale. Ainsi, la faible teneur en carbonate biogénique dans les sédiments du dernier stade glaciaire serait principalement liée à la faible productivité des organismes calcifiants. Néanmoins, ces indices de dissolution suggèrent une préservation de la calcite légèrement inférieure lors du Bølling-Allerød (14,75 à 12,7 ka) et de

12,2 à 8,3 ka. La fragmentation un peu élevée pendant le Bølling-Allerød pourrait être due à l'abrasion mécanique des débris transportés par la glace abondante dans cet intervalle, tandis que celle enregistrée de 12,2 à 8,3 ka, correspond à une productivité élevée et à de faibles apports détritiques suggérant une dissolution de la calcite biogène provoquée par la reminéralisation oxydante de la matière organique dans les sédiments. Dans les sédiments glaciaires, la présence d'alcénones malgré l'absence de coccolithes et sans indice de dissolution apparente, soulève des questions sur la source des alcénones lors de la dernière glaciation.

À plus grande échelle, un transect de cinq carottes sédimentaires de la zone de fracture Charlie-Gibbs (ZFCG), dans le nord-ouest de l'Atlantique Nord, la mer du Labrador et la Baie de Baffin, permet de dresser un portrait des changements de flux de carbonates biogéniques depuis le dernier épisode glaciaire. Dans les enregistrements de la ZFCG et de la mer du Labrador, la dernière période glaciaire est caractérisée par de faibles flux de coccolithes qui augmentent à l'Holocène. Les flux d'alcénones varient également. Dans la ZFCG et le sud-est de la mer du Labrador, le passage de la période glaciaire à interglaciaire s'accompagne d'une forte augmentation concomitante des concentrations de coccolithes et d'alcénones. Dans le nord de la mer du Labrador, il n'y a pas de relation entre les variations de concentrations de coccolithes et d'alcénones, ce qui suggère un découplage entre les flux de coccolithes et ceux des alcénones. Dans les deux carottes de la baie de Baffin, très peu de spécimens de coccolithes ont été dénombrés dans les sédiments du dernier épisode glaciaire et ils sont absents des sédiments holocènes en raison de la dissolution de ces derniers. En revanche, les deux séquences renferment des alcénones dans les sédiments d'âge glaciaire et interglaciaire. Les données illustrent une forte déconnexion entre la préservation des coccolithes dans la baie de Baffin versus la mer du Labrador. Elles suggèrent également que des haptophytes non-calcifiants peuvent avoir contribué aux flux d'alcénones dans le nord-ouest de la mer du Labrador et dans la baie de Baffin.

Cette thèse documente de grands changements de productivité à l'échelle du nord-ouest de l'Atlantique Nord, de la dernière glaciation au présent interglaciaire. Au sud du détroit de Davis, dans la mer du Labrador, la productivité des coccolithophores calcifiants aurait été minimale lors de la dernière glaciation et aurait augmenté par plus de deux ordres de grandeur de la déglaciation au début de l'Holocène. La baie de Baffin est restée un bassin épicontinental où la transition glaciaire-interglaciaire aurait été accompagnée d'une dissolution accrue du carbonate de calcium.

Mots-clés : Coccolithes, alcénones, foraminifères, carbonate biogénique, dissolution, préservation, productivité, baie de Baffin, mer du Labrador, pente Scotian, zone de fracture Charlie-Gibbs

ABSTRACT

This thesis focuses on primary productivity and calcite preservation in the northwest North Atlantic and adjacent subpolar basins, including the Labrador Sea and Baffin Bay, to document modifications of the marine carbon cycle over the extreme climatic changes that occurred there since the last glaciation. To this end, coccolith and alkenone analyses were undertaken on sixty-six surface sediment samples and on five sediment cores encompassing the past 25,000 years. Coccoliths, the calcitic remains of some haptophytes (unicellular phototrophic taxa), and alkenones, organic biomarkers produced by certain species of haptophytes (notably *Emiliania huxleyi*), provide complementary information on coccolithophore productivity and calcium carbonate preservation.

A comparison of the coccolith and alkenone contents of surface sediment samples with environmental ocean conditions, including sea-surface temperature (SST) and salinity (SSS), sea-ice cover, and primary productivity, reveals close relationships between the distribution of coccolith species and surface water properties. For example, the presence and high abundance of *Coccolithus pelagicus* is closely related to low sea-surface temperatures and relatively high sea-surface salinities. In the northwest North Atlantic, calcite dissolution impacts the coccolith distribution in sediments, particularly in Baffin Bay, an area of active calcite dissolution in the water column, as well as along the Labrador Shelf where dissolution appears to be triggered in the sediment by oxic diagenesis. In neritic environments, non-calcifying haptophytes probably produce alkenones, which may explain the abnormally high alkenone concentrations in the Laurentian Channel and the $U^{K_{37}}$ -temperature offsets recorded on the Scotian Slope and the Orphan Basin region of the Labrador Sea.

To document the glacial to interglacial changes in biogenic carbonate production and preservation in the northwest Labrador Sea, core HU2008-029-004 PC was analyzed. The results, spanning the last 25,000 years, show large changes in coccolith abundance with concentrations much lower in glacial sediments (mean of $9.5 \pm 3.9 \times 10^5$ coccoliths g^{-1}) than postglacial sediments (up to a maximum of 3.1×10^8 coccoliths g^{-1}). Three calcite dissolution indices were also used to evaluate biogenic carbonate preservation: planktonic foraminifer fragmentation, the ratio of benthic-to-planktonic foraminifers (B/P), and the ratio of organic linings to benthic foraminifers (OL/B). The fragmentation index remains low throughout the entire sequence with 7.1 ± 0.8 % in the glacial unit, and 3.9 ± 0.2 % in the postglacial sediments. Likewise, the B/P (< 0.2) and OL/B values are low throughout the studied sequence. These indices indicate good biogenic carbonate preservation throughout most of the studied interval, which implies that the low biogenic carbonate content in the sediments of the last glacial stage is primarily related to the low productivity of calcifying organisms. In contrast, the dissolution indices suggest slightly lower calcite preservation during the Bølling-Allerød (14.75–12.7 ka) and from 12.2 to 8.3 ka in the northwest Labrador Sea. During the Bølling-Allerød, sediments recorded elevated fragmentation that may be due to mechanical abrasion from ice-rafted debris, whereas the slightly elevated fragmentation index from 12.2 to 8.3 ka, a time of elevated productivity and little ice-rafted debris, may reflect dissolution in the sediments during oxic diagenesis. The presence of alkenones, despite the absence of coccoliths, in the sediments of glacial age also raises questions about the source of the alkenones during the last glaciation.

At a larger scale, a transect of five marine sediment cores from the Charlie-Gibbs Fracture Zone (CGFZ) in the northwest North Atlantic, to the Labrador Sea and the Baffin Bay, document changes in biogenic carbonate production since the last glacial interval. In the CGFZ and Labrador Sea sediment records, coccolith fluxes are low during the last glaciation but rise at the onset of the Holocene. The alkenone fluxes also show large variations. In the cores from the CGFZ and the southeast Labrador Sea, the coccolith and alkenone concentrations record a large concomitant increase from the glacial to interglacial. In the northwest Labrador Sea core, there is no correlation between the coccolith and alkenone concentrations, suggesting a decoupling between their respective fluxes. In the Baffin Bay cores, coccoliths are not preserved in Holocene sediments due to the presence of undersaturated bottom waters and consequent calcium carbonate dissolution, but a few specimens were recovered in the glacial sediments, whereas alkenones occur throughout the sequences. The data illustrate a clear disconnect between coccolith preservation in Baffin Bay and the Labrador Sea. They also suggest that non-calcifying haptophytes may have contributed to alkenone fluxes in the northwest Labrador Sea and Baffin Bay.

This thesis documents large variations in coccolithophore productivity from the last glaciation to the present interglacial at the scale of the northwest North Atlantic. South of Davis Strait, in the Labrador Sea, calcifying coccolithophore productivity was minimal during the last glaciation and rose into the deglaciation and early Holocene by at least two orders of magnitude. Baffin Bay behaved as an epicontinental basin, where the transition from glacial to interglacial was accompanied by enhanced calcium carbonate dissolution.

Keywords: Coccoliths, alkenones, foraminifers, biogenic carbonate, dissolution, preservation, productivity, Baffin Bay, Labrador Sea, Scotian Slope, Charlie-Gibbs Fracture Zone

INTRODUCTION

1. General objectives of this thesis

There are inherent complexities involved with paleoceanographic studies of marine sediments in subpolar environments, especially those subject to calcite dissolution (e.g., Jackson et al., 2017). Nevertheless, the northwest North Atlantic is an important pathway for Arctic freshwater export and global oceanic circulation (Wang et al., 2018), and obtaining records of the oceanographic and biologic trends in the study area since the last glaciation is critical to interpret the marine carbon cycles in the region and how they have changed since the peak of the last ice age. My research focuses on marine sediments from deep core sites of the northwest North Atlantic Ocean to generate large-scale basin-wide records, with an emphasis on coccolith and alkenone contents. Alkenones are not frequently reported with coccoliths, but I document corresponding alkenone concentrations with all coccolith records. I also assembled a dataset of core-top seafloor sediments as a dataset to correlate with modern oceanographic records of temperature, salinity, sea-ice cover, and annual primary productivity.

This thesis addresses coccolithophore productivity and calcium carbonate preservation in the subpolar northwest North Atlantic, based on the analyses of both coccoliths and alkenones, in addition to foraminifers, in marine sediment samples encompassing the Last Glacial Maximum (LGM) to the present interglacial. In the first phase of this thesis, the analyses of coccoliths and alkenones in seafloor sediments are used to document their recent spatial distribution and relationships with surface ocean parameters (temperature, salinity, sea-ice cover, and primary productivity). In the second phase, I explore the temporal variations of coccoliths, alkenones, and foraminifers in one core spanning the last 25,000 years from the northwest Labrador Sea. This core was studied previously to document the palynological assemblages and stable isotopes in

foraminifers in addition to the geochemical properties of the sediments (Gibb et al., 2014; Nuttin et al., 2015). By completing the data set with foraminiferal data to document preservation of biogenic carbonate remains, we provide a comprehensive picture of changes in productivity in the northwest Labrador Sea. Finally, the third phase of this thesis expands on the records of coccoliths and alkenones at a regional scale in the northwest North Atlantic, from the mid-latitude Charlie-Gibbs Fracture Zone to the subpolar Baffin Bay.

The overall objective of my thesis was to assess past primary productivity and preservation in marine sediments of the northwest North Atlantic based on coccoliths and alkenones, variables that are rarely investigated together, along with foraminifer-based dissolution indicators and other previously published micropaleontological and isotope indicators.

2. Geographic and hydrographic setting of the study areas

The Baffin Bay, the Labrador Sea, the Scotian Slope, and the Charlie-Gibbs Fracture Zone make up the study area for this thesis (thereafter collectively called the northwest North Atlantic; Fig. 1). The Labrador Sea, a central sub-basin in this study, is separated from the semi-enclosed Baffin Bay by a shallow ~ 700 m deep sill to the north (Davis Strait; Baker and Friedman, 1973; McLaughlin et al., 2006). Davis Strait impedes bottom water exchange (Rudels, 1986) and isolates the bottom ecological environments of Baffin Bay from the Labrador Sea (Schröder-Adams and Van Rooyen, 2011). Baffin Bay is also connected to the Arctic Ocean via the Canadian Arctic Archipelago. Baffin Bay Bottom Water (~ 1300 m to the bottom) is old (~ 100–500 years old; Top et al., 1980) and is characterized by high metabolic carbon dioxide concentrations because it is not readily ventilated. Bottom waters in Baffin Bay are presently

undersaturated with respect to both calcite and aragonite (Azetsu-Scott et al., 2010). Tan and Strain (1980) show that Baffin Bay Bottom Water is sourced from the Arctic Ocean and is either injected into Baffin Bay in irregular pulses or through a slow and continuous advection process. Along the Greenland margin, the West Greenland Current flows northwards through eastern Davis Strait, and contains a component of the cool and low saline ($-1.8\text{ }^{\circ}\text{C}$; practical salinity (S_p) ≤ 34.5) waters of the East Greenland Current over the shelf (Cuny et al., 2002). In the west, the Baffin Island Current flows through western Davis Strait and into the Labrador Sea, where it mixes with Hudson Strait outflow water to become the Labrador Current (Percy and Fife, 1993; Lazier and Wright, 1993).

The Labrador Sea has an average depth of $\sim 2\text{ km}$ (maximum depth is $\sim 4.3\text{ km}$). Deep-to-intermediate water formation in the Labrador Sea (Labrador Sea Water) is directly impacted by the Labrador Current and the North Atlantic Drift and it influences the formation of North Atlantic Deep Water (NADW) of the Atlantic Meridional Overturning Circulation (AMOC) (Yashayaev, 2007; Jones and Anderson, 2008; Yashayaev and Loder, 2009). Labrador Sea Water is formed by deep convection (overturning) following strong surface cooling during severe winters (Clarke and Gascard, 1983). It spreads throughout the entire deep North Atlantic (Yashayaev et al., 2007a, b) and plays an important role in the transfer of heat, water, and biogeochemical properties (e.g., oxygen and carbon) between the atmosphere and the deep ocean (Yashayaev et al., 2007a, b).

The Labrador Current flows along the Labrador coastline and continues southward, parallel to the coastline along eastern Newfoundland and then it takes a sharp westward turn along the edge of the Grand Banks to make it to the Scotian Shelf/Slope, offshore Nova Scotia. The steep topography along the Scotian Shelf break causes enhanced vertical mixing that results

in high primary productivity (Breeze et al., 2002; Greenan et al., 2004; Ji et al., 2006; Stortini, 2015). Oceanic fronts also regularly occur along the Scotian Slope as cold shelf waters meet the warmer slope waters and the locations of these frontal areas varies throughout the year (Horne, 1978; Fournier, 1978). Submarine canyons are found along the Scotian Slope. They host many different habitats within a relatively small area and, thus, support high species diversity (Hecker, 1990; Hargrave et al., 2004).

The study area extends eastwards to the far offshore Charlie-Gibbs Fracture Zone (CGFZ), a natural tectonic corridor that contains a flat seafloor surrounded by gentle slopes and rocky cliffs (Alt et al., 2019) that coincide with the northern edge of the surface subpolar front (Priede et al., 2013). The CGFZ is also where both warm and cold limbs of the AMOC cross the Mid-Atlantic Ridge (Furey et al., 2014). The surface flow of the North Atlantic Drift circulates above the Charlie-Gibbs Fracture Zone (Schott et al., 1999).

3. Previous research

Baffin Bay surface sediments collected below 900 m water depth were free of biogenic calcite, indicating 100 % loss of foraminifers (and coccoliths) to dissolution (Aksu, 1983). There is intense calcite dissolution between 600 to 900 m in modern Baffin Bay, based on core top and grab sediments (Aksu 1983). The calcite saturation state (Ω_{cal}) is the product of the concentrations of the carbonate ($[\text{CO}_3^{2-}]$) and the calcium ($[\text{Ca}^{2+}]$) ions divided by the stoichiometric solubility product of calcite (K_{cal}), where K_{cal} is a function of salinity, temperature (Mucci, 1983), and pressure (Millero, 1995). The calcite saturation horizon ($\Omega_{\text{cal}} = 1$) represents the depth in the water column below which calcite starts to dissolve (Peterson, 1966). In contrast to Baffin Bay, where the calcite saturation horizon ($\Omega_{\text{cal}} = 1$) is relatively shallow, this horizon is

not found within the modern Labrador Sea. In fact, Ω_{cal} is ~ 1.2 at the seafloor at 3,500 m water depth in the Labrador Sea (Azetsu-Scott et al., 2010). Thus, Baffin Bay and the Labrador Sea are chemically different water bodies. Likewise, the calcite saturation horizon in the central North Atlantic is found at 4.3 to 4.5 km water depth (Chung et al., 2003).

Dinocyst assemblages have been used to reconstruct regional changes in oceanographic circulation and sea-surface conditions from deep and central sites throughout Baffin Bay and the Labrador Sea (de Vernal et al., 2000, 2005, 2006; Radi and de Vernal, 2008a, b; Gibb et al., 2014, 2015). South of Davis Strait, the breakup of perennial sea ice began at ~ 11.9 ka, although the shift to seasonal sea-ice cover and warmer sea-surface temperatures in Baffin Bay did not occur until after ~ 7.4 ka (Gibb et al., 2015). The differential evolution of sea-surface conditions in the Labrador Sea and the Baffin Bay was likely a result of limited water exchange through Davis Strait and the strengthened North Atlantic component of the West Greenland Current in relation to the final retreat of the Laurentide and Greenland ice sheets (Gibb et al., 2015). Gibb et al. (2014) also documented the presence of a large amount of reworked materials in glacial sediments of the northwest Labrador Sea, and proposed, based on dinocyst abundances, that there was a crash in primary productivity during the last glacial interval. Primary productivity in the southeast Labrador Sea was low during the last glaciation and increased into the Holocene (Hillaire-Marcel et al., 1994a, b).

Calcite dissolution has been studied in many locations using a variety of methods (Table 1). A quantitative method to assess the degree of calcite shell preservation in Davis Strait was developed using the ratio of organic linings of benthic foraminifers and their calcite shells (de Vernal et al., 1992). This method was later applied to a core from the northeast Labrador Sea (Hillaire-Marcel et al., 1994b) and to northern Baffin Bay surface sediment samples (Hamel et

al., 2002). This dissolution indicator revealed that calcite dissolution has increased in Davis Strait since the last glaciation, and that there was a peak in calcite dissolution when interglacial conditions were fully established in the surface waters (de Vernal et al., 1992). According to this dissolution indicator, calcite dissolution currently occurs in the deep, modern northern Baffin Bay (Hamel et al., 2002) but there was preservation in glacial sediments based on other foraminifer dissolution indices (fragmentation, benthic-to-planktonic foraminifers, etc.; Aksu, 1983; Table 1). Likewise, Hillaire-Marcel et al. (1994b) found a slight but insignificant increase in the dissolution index in the early-to-mid Holocene sediments of a core recovered from the northeast Labrador Sea. Dissolution indicators discussed in this thesis are solely based on foraminifers and not on coccoliths. Subhas et al. (2019) found that foraminifers are preferentially dissolved compared to coccoliths in undersaturated seawater. Consequently, whereas foraminifer dissolution does not necessarily imply coccolith dissolution, intense foraminifer dissolution infers potential coccolith dissolution.

In the Gulf of St. Lawrence and on the Scotian Shelf, Mucci et al. (2000) and Nesbitt and Mucci (2020) found that while the bottom waters are supersaturated with respect to calcite, the amount of metabolic CO₂ generated during oxic diagenesis of organic matter was sufficient to induce calcite dissolution in most retrieved cores.

4. Materials

The sediments used to address my research objectives were retrieved during several research cruises over the last three decades in the northwest North Atlantic. The sediments include sixty-six core-top subsamples (black dots; Table 2; Fig. 1) and five long cores (Table 3).

5. Methodology

5.1 Coccoliths

Coccolithophores are unicellular phytoplankton of the Haptophyta division and the Prymnesiophyceae class (Jordan and Chamberlain, 1997). They are one of the dominant primary producers and one of the most important pelagic calcifying organisms in the modern ocean (Baumann et al., 2004; Hay, 2004; Monteiro et al., 2016). Coccolithophores take up inorganic carbon in the surface ocean. They are grazed upon by zooplankton, die, and sink, thus sequestering carbon to the deep ocean (Ducklow et al., 2001). Calcite export through coccolith production and sedimentation contributes to buffering ocean alkalinity and regulating pH (Schiebel, 2002; Henehan et al., 2016).

Coccolithophores thrive in the photic zone of stratified and oligotrophic water masses (Baumann et al., 2005). They are protected within an exoskeleton composed of numerous calcite plates, or coccoliths, that can be preserved in the sedimentary record (Baumann et al., 2005). Coccolith assemblages may be used to infer past ocean conditions as their distribution in sediments is strongly linked to the properties of surface water masses (e.g., McIntyre and Bé, 1967; Eide, 1990; Samtleben and Schröder, 1992; Baumann and Matthiessen, 1992; Levesque, 1995; Baumann et al., 1999; Boeckel and Baumann, 2004). Coccoliths can be a major component of the fine-grained sediments of the Mesozoic-Cenozoic geologic record, preserving valuable paleoceanographic information. However, several studies have shown that sinking coccoliths can be highly altered or dissolved by the time they reach the seafloor and thus

represent a fraction of the coccoliths that were originally produced in surface waters (Andruleit, 1997; Milliman et al., 1999; Kinkel et al., 2000).

Some coccolithophore species, including the cosmopolitan *Emiliania huxleyi* and the closely related *Gephyrocapsa oceanica*, synthesize long-chain unsaturated ketones (alkenones; C₃₇–C₃₉; de Leeuw et al., 1980; Volkman et al., 1980; 1995; Brassell et al., 1986; Conte et al., 1998; Theroux et al., 2010). Among the haptophytes of the marine realm, *Emiliania huxleyi* is the dominant alkenone producer (Volkman et al., 1980; Marlowe et al., 1984a, b, 1990; Conte et al., 1994; Rosell-Melé et al., 2000). This species is the most widespread coccolithophore in the modern ocean, ranging from the subpolar to the oligotrophic tropical ocean (Winter et al., 2014). Some non-calcifying haptophytes from coastal seas, estuaries, and lakes also produce alkenones (Marlowe et al., 1984a; 1990; Coolen et al., 2004; de Vargas and Probert, 2004; Malinverno et al., 2008; Bendif et al., 2013).

5.1.1 Genetic variations of coccoliths

It is questionable whether a universal response to changes in temperature can be expected from different haptophyte producers. Brand (1982) found that genetic strains from cold environments are more adapted to cold temperatures than those from warmer waters. Brand (1984) also documented genetic differences in salinity tolerances in oceanic and coastal genotypes. Likewise, seventy globally-distributed Holocene sediment samples contained coccolith assemblages that showed strong environmental control on the morphotypes (Bollmann, 1997). Conversely, Medlin et al. (1996) found little genetic variation in ribosomal ribonucleic acid (rRNA) from three geographically different types of coccoliths and Fujiwara et al. (2001)

reported a series of genetic similarities between *Emiliana*, *Isochrysis*, *Gephyrocapsa*, and *Chrysotila*.

The relationship between *Emiliana huxleyi* coccolith dissolution or calcification is non-linear and complicated by extensive morpho- or genotypic variability (Winter et al., 2014). In fact, some strains of *Emiliana huxleyi* may withstand changes in pH better than others (Langer et al., 2009). Boye et al. (2019) conclude that small coccolithophores (e.g., *Emiliana huxleyi* and *Gephyrocapsa oceanica*) likely increase the pH (reducing acidity) in their internal vesicle as pH decreases in the surrounding water column, in order to precipitate calcite, while the larger coccoliths (mainly *Calcidiscus leptoporus*) do not show evidence for this kind of response to acidification. Different genetic strains of the main alkenone producer in this thesis, *Emiliana huxleyi*, could be significant and contributing to the alkenones analyzed in this study. For example, Nam et al. (2018) identified several candidate genes associated with the coccolith production of *Emiliana huxleyi*. While genetic differences were not investigated in this thesis, I am aware that there is likely more than one genetic strain of *Emiliana huxleyi* within the analyzed samples and that their genetic differences could play a role in whether coccoliths are produced and in their alkenone production. This complication is beyond the scope of this thesis but should be examined in future research.

5.2 Alkenones

Alkenones are long-chain and unbranched organic molecules (unsaturated ketones) synthesized by a select group of unicellular haptophyte algae (Class: Prymnesiophyceae; Hibberd, 1976; Jordan and Kleijne, 1994), including the calcifying coccolithophore *Emiliana huxleyi* and *Gephyrocapsa* spp. (the genera that are the main producers of alkenones in the

marine environment; Volkman et al., 1980; Marlowe et al., 1984a,b, 1990; Conte et al., 1994; Rosell-Melé et al., 2000). Alkenones are commonly used to reconstruct sea-surface temperatures using U^{K}_{37} (Brassell et al., 1986) and $U^{K}_{37'}$ (Prah1 and Wakeham, 1987), a relative abundance ratio of $C_{37:2}$, $C_{37:3}$, and $C_{37:4}$ alkenones (e.g., $C_{37:2}$ stands for an alkenone with thirty-seven carbons and two double bonds) in sediments that change linearly with sea-surface temperature (Prah1 and Wakeham, 1987; Brassell et al., 1986; Müller et al., 1998). The U^{K}_{37} (or $U^{K}_{37'}$) index is larger in warmer waters where alkenones with a lower degree of unsaturation (i.e., fewer double bonds) are synthesized (Brassell et al., 1986). Since the tetra-unsaturated alkenone, $C_{37:4}$, is only common in sediments from high latitudes, the U^{K}_{37} equation was modified to exclude $C_{37:4}$ ($U^{K}_{37'}$; Prah1 and Wakeham, 1987). Nonetheless, U^{K}_{37} has been found to be more robust than $U^{K}_{37'}$ down to 6 °C (Rosell-Melé, 1998; Bendle and Rosell-Melé, 2004). Some studies have also raised the possible presence of allochthonous alkenones in the study area (e.g., Rosell-Melé et al., 2011; Filippova et al., 2016), which would directly impact the validity of alkenone-based temperature reconstructions in sediments.

Alkenones in sediments are not powerful tools to estimate past changes in surface ocean temperatures in regions where alkenone productivity and alkenone sediment content are very low, including areas in which temperatures are beyond the range of the temperature calibration (< 5 and > 26 °C; Conte et al., 2006) such as the polar oceans (Mix et al., 2000). Nevertheless, Moros et al. (2016) analyzed subpolar alkenones from Disko Bugt, offshore western Greenland, and derived U^{K}_{37} -based temperature estimates and % $C_{37:4}$ (relative proportion of $C_{37:4}$ to total alkenones). Given the large errors associated with alkenone-based estimates at cold temperatures, their sea-surface temperature reconstructions were qualitative rather than quantitative (Moros et al., 2016). Rosell-Melé et al. (2011) raised the possibility that coccoliths

and alkenones in the sediments of the North Atlantic may originate from different sources and that they may be subjected to different transport and preservation processes. Rosell-Melé et al. (2011) found that higher concentrations of coccoliths and alkenones occur in sediments of the North Atlantic during the Holocene ($9.2 \pm 7.2 \times 10^8$ coccoliths g^{-1} ; 169 ± 87 ng g^{-1}) than during the last glacial period ($1.9 \pm 1.6 \times 10^8$ coccoliths g^{-1} ; 34 ± 23 ng g^{-1}).

Alkenone concentrations in marine sediments can vary widely. The alkenone concentrations produced by *Emiliania huxleyi* (5 ml packed cell volume) grown in laboratory cultures varied between $1.43\text{--}2.65 \times 10^6$ ng g^{-1} and showed a decreasing trend with growth period (Yamamoto et al., 2000). Furthermore, alkenones produced in surface waters are likely largely respired and only an extremely small fraction of the alkenones exported from surface waters are preserved in the sediments. In fact, the export flux of alkenones to the sediments from the water column in pelagic regions is generally less than 1 % of the total production in the euphotic zone (Müller and Fischer, 2001). In addition, diagenetic alteration and differential preservation of alkenones in the sediments is possible, as greater undersaturation of more double bonds could mean higher reactivity. However, no study to date has demonstrated a diagenetic alteration bias in alkenones (Rosell-Melé and McClymont, 2007). For example, sediment trap studies have failed to reveal a change in the U^{K}_{37} index with depth in the water column that could be related to microbial degradation (Sawada et al., 1998; Müller and Fischer, 2001). Likewise, Prah et al. (1989) observed no shift in U^{K}_{37} of alkenones in sediments above and below an oxidation front, even though > 85 % of the alkenones degraded over the 8000 years of burial.

5.3 Foraminifers

Foraminifers are heterotrophic protists that surround their unicellular bodies with calcite shells (also called tests) that can be preserved in sediments (Kucera, 2007). Fossilized planktonic foraminifer shells form the backbone of Cenozoic biostratigraphy (Berggren et al., 1995) and foraminifers have been instrumental in the study of rates and patterns of evolution (Norris, 2000). The overwhelming majority of planktonic foraminifers in the northwest North Atlantic are *Neoglobobulimina pachyderma* left-coiled (also referred to as Npl; and a planktonic foraminifer indicator species for polar waters; Wu and Hillaire-Marcel, 1994). A reduction in sea-ice cover in high-latitude environments typically leads to an increase in surface productivity and uptake of atmospheric carbon dioxide in open water areas (Mucci et al., 2010; Butterworth and Miller, 2016; DeGrandpre et al., 2020; Mayot et al., 2020) available for primary productivity. Under these conditions, fluxes of planktonic foraminifers (and coccoliths) in marine sediments can serve as a measure of the carbon flux to the seafloor, and thus productivity, as has been interpreted for dinoflagellate cysts found in sediments (Rochon and de Vernal, 1994; Hillaire-Marcel et al., 1994b).

Benthic foraminifers occupy a wide range of marine environments at all latitudes, from brackish estuaries to deep ocean basins (Todd and Low, 1980; Mudie et al., 1983/84; Wollenburg and Mackensen, 1998). Many of them have well-defined salinity and temperature preferences (e.g., Polyak et al., 2002). The organic linings of benthic foraminifers are often found among the sediment palynomorphs and their abundance is related to benthic foraminifer productivity and organic fluxes to the seafloor (e.g., de Vernal et al., 1992; Wollenburg and Kuhnt, 2000; Wollenburg et al., 2004; Gooday et al., 2012). Organic linings are refractory and preserved in the sediments after the external and calcitic benthic foraminifer shells have

dissolved away (de Vernal et al., 1992). If dissolution does not occur, the shell remains, and the organic lining is not exposed.

5.4 Calcium carbonate preservation and dissolution indicators

Biogenic carbonate shells from nanno- and micro- organisms make up most of the calcite that accumulates on the seafloor (Honjo et al., 1982; Sun and Turchyn, 2014; Dutkiewicz et al., 2015). Calcite shells are produced by planktonic foraminifers and coccolithophores that inhabit ocean surface waters as well as by calcitic benthic foraminifers that live on the seafloor (Morse and Mackenzie, 1990). Upon the death or grazing of these organisms, their shells will either dissolve or accumulate at the seafloor (Jansen et al., 2002). Their preservation in marine sediments depends largely on the saturation state of the overlying waters and sediment porewaters (e.g., Morse and Mackenzie, 1990; Mucci et al., 2000). The saturation state of oceanic waters typically decreases with depth because calcium carbonate solubility increases with depth (pressure) while pH decreases because of the accumulation of metabolic carbon dioxide resulting from the microbial decay of settling organic matter. Increased acidity is detrimental to the formation of calcite shells in the water column and to the preservation of these shells in sediments over time (Pytkowicz and Connors, 1964).

Sediment and dynamic horizons define the changing saturation state of the ocean with respect to calcite (Pälike et al., 2012). The calcite lysocline, located below the calcite saturation horizon ($\Omega_{\text{cal}} = 1$), is the depth at which there is a rapid change in the rate of calcite dissolution. At a depth below the lysocline, the rate of calcite dissolution is equal to the rate of calcite input from the overlying water column so there is little or no net accumulation of calcite shells in the sediment. This horizon is empirically-defined as the calcite compensation depth (CCD;

Bramlette, 1961; Boudreau, 2013; Berger, 2016; Boudreau et al., 2020). Apart from the saturation state of the overlying waters, other factors impact the biogenic carbonate content of sediments such as surface water productivity, shell characteristics (size, wall thickness, and microstructure), rate of bottom water flow as it determines the replenishment of undersaturated bottom water, dilution by non-calcitic minerals, and the sedimentation rate that controls the time that shells are exposed to corrosive bottom waters (Li et al., 1969; Broecker and Takahashi, 1978; Berger et al., 1982). Calcite dissolution can also occur above the calcite saturation horizon through biologically-mediated dissolution inside zooplankton (Harris, 1994; Milliman et al., 1999) and through dissolution in oxic sediments in response to microbial organic matter degradation and metabolic carbon dioxide production, as commonly observed in organic-rich coastal sediments (Froelich et al., 1979; Emerson and Bender, 1981; Hales and Emerson, 1996; Martin and Sayles, 1996; Mucci et al., 2000; Nesbitt and Mucci, 2020).

A quantitative method to assess the degree of calcite preservation and dissolution was developed by de Vernal et al. (1992) using the ratio of the relative abundance of organic linings of benthic foraminifers and their calcite shells. This method was applied to Late Quaternary marine sediments in Davis Strait to reveal that calcite dissolution has increased in Davis Strait since the LGM, and that dissolution peaked when interglacial conditions were fully established in the surface waters (de Vernal et al. 1992). Planktonic foraminifer fragmentation (Berger, 1968; Thunell, 1976) and the benthic-to-planktonic foraminiferal shell ratio (B/P; Aksu et al., 1989) are two additional and commonly used indicators of calcite dissolution. The fragmentation index is based on counts of planktonic foraminifer fragments and whole shells (Williams et al., 1985; Malmgren, 1987; Le and Shackleton, 1992; Wang et al., 2016). To determine the extent of planktonic foraminifer fragmentation, whole and fragments (less than one half of the original

shell) (Malmgren, 1987) of planktonic foraminifer shells are counted in populations of > 300 specimens in the > 150 μm size fraction (Le and Shackleton, 1992; Kucera, 2007). The B/P ratio is based on counts of whole shells (Conan et al., 2002; Kucera, 2007). Deep-sea benthic foraminifers generally have thick, smooth shells that are more resistant to dissolution than those of planktonic foraminifers. Consequently, B/P typically increases with increasing calcite dissolution (Thunell, 1976; Kucera, 2007) so high values approaching infinity imply strong dissolution. The B/P ratio provides more accurate estimates of changes at a single location than between locations due to regional variations in productivity and water depth, but B/P must be interpreted with caution since it has also been used as a non-independent indicator of productivity and water depth (Berger and Diester-Hass, 1988; van der Zwaan et al., 1990; Mleneck-Vautravers, 2018). The B/P ratio can also record very pronounced changes during large scale climatic changes such as between glacial and interglacial times, because of changes in ecological conditions (Henrich, 1986). The B/P ratio can also be impacted by freshwater inputs since they can significantly alter surface productivity.

Aksu (1983) used semi-quantitative methods to study the degree of fossil biogenic carbonate dissolution in Baffin Bay and Davis Strait sediments. Through the use of independent calcite dissolution proxies (e.g., the ratio of arenaceous to calcareous benthic foraminifers, the B/P ratio, the relationships between the dissolution-susceptible and dissolution-resistant foraminifers, the degree of foraminifer fragmentation, and the relationships between the abundance of calcite shells in water-column plankton tows and in the surface sediments), Aksu (1981, 1983) found that the modern calcite compensation depth (CCD) in Baffin Bay is located between 600 to 900 m water depth and demonstrated that the ratio of living planktonic foraminifers in the water column to the number of foraminifer shells in sediment core tops is an

effective tool to estimate the modern preservation of biogenic carbonate on the seafloor. The dissolution of foraminifers (from core top samples) was estimated to be $< 30\%$ in Davis Strait, but was $> 90\%$ throughout Baffin Bay (Aksu 1983). In addition, based on the analysis of long cores, Aksu (1983) concluded that there was intense dissolution of biogenic carbonate shells in Baffin Bay during interglacial or interstadial isotope stages 1, 3, 5, 7, and 9, likely as a result of the input of cold and CO_2 -rich waters from the Arctic into this poorly-ventilated basin.

6. Thesis structure

This thesis is divided into three research chapters:

Chapter One consists of a manuscript entitled *Carbonate dissolution and environmental parameters govern coccolith vs. alkenone abundances in surface sediments from the northwest North Atlantic*. It has been submitted for publication to *Marine Micropaleontology*. This chapter focuses on spatial variations of coccolith assemblages and concentrations as well as alkenone concentrations in surface sediments in relation to sea-surface characteristics (temperature, salinity, sea-ice cover, and productivity). In this chapter, we evaluate assumptions and complications that are inherent with the study of biogenic carbonate productivity and preservation and consider the possibility of using coccolith assemblages in combination with the modern analogue technique to reconstruct past ocean conditions.

Chapter Two reports on a reconstruction of biogenic carbonate productivity and preservation based on the analysis of sediments from one core retrieved in the northwest Labrador Sea. Coccoliths, alkenones, and foraminifers are used to infer primary productivity and calcite shell preservation over the past 25,000 years. The decoupling of coccoliths and alkenones

and the possibility of non-calcifying haptophytes producing alkenones during the last glacial interval are also discussed in this chapter. *Chapter Two* is entitled *Biogenic carbonate fluxes and preservation in the northwest Labrador Sea since the Last Glacial Maximum* and has been submitted for publication to *Palaeogeography, Palaeoclimatology, Palaeoecology*.

Chapter Three is a manuscript entitled *Coccolith and alkenone records from the northwest North Atlantic illustrate contrasted productivity from glacial to interglacial* and this manuscript will be submitted for publication to *Marine Geology*. This manuscript contains a synthesis of the analyses of coccoliths and alkenones from five sediment cores recovered along a transect across the northwest North Atlantic. It includes the coccolith and alkenone records presented in *Chapter Two* from the northwest Labrador Sea with an additional two records from eastern and western Baffin Bay, one record from the southeast Labrador Sea and one record from the Charlie-Gibbs Fracture Zone of the open North Atlantic. With these records, I interpret regional variations in coccolithophore productivity for the past 25,000 years from across the northwest North Atlantic.

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Tables

Reference	Region	Time Frame	Purpose	Method
Aksu (1983)	Baffin Bay and Davis Strait	Pleistocene	Intensity of calcite dissolution	Arenaceous/calcareous benthic forams, benthic/planktonic forams, dissolution-susceptible vs. resistant forams; test fragmentation, plankton tows and surface sediment fauna
Morris and Clark (1986)	Central Arctic Ocean	Pleistocene	Lysocline curve and paleocurrents	Planktonic foram abundance and fragmentation, $\delta^{18}\text{O}$, and grain size
de Vernal et al. (1992)	Davis Strait	Late Quaternary	Evaluate quantitatively the degree of calcite preservation/dissolution	Dissolution index with benthic forams: $\log(\text{organic linings/carbonate shells})$
Boeckel and Baumann (2004)	South Atlantic Ocean	Modern	Modern position of the coccolith lysocline	Coccolith content, coccolith CaCO_3 contribution, Coccolith abundances, CEX' dissolution index
Pälike et al. (2006)	Equatorial Pacific	Oligocene	Lysocline curve	Bulk sediment stable isotopes, CaCO_3 data, modeling
Chiu and Broecker (2008)	Ceara Rise and Ontong-Java Plateau	Late Quaternary	New insights into the calcite size index	$\%\text{CaCO}_3$ content, size separation, SEM observation, ratio of dissolution-susceptible forams to total CaCO_3
Gibbs et al. (2010)	Southern Ocean, Pacific Ocean, and offshore New Jersey	PETM	Reconstruct acidification and coccolith production	Sr/Ca (production rate), coccolith abundances based on production rate and correction factors, correction factor = abundance lost through dissolution, wt. $\%\text{CaCO}_3$
Pälike et al. (2012)	Equatorial Pacific	Cenozoic	CCD curve	CaCO_3 and mass accumulation rates, paleo-depths, modeling
Lalicata and Lea (2011)	Equatorial Pacific	Pleistocene	Relative calcite dissolution fluctuations	$\%$ coarse fraction, foram shell fragmentation
Mekik et al. (2012)	Tropical and subtropical Pacific, Atlantic, and Indian Oceans	Late Quaternary	Deglacial calcite preservation maximum missing in some locations	Size-normalized foram shell weight, <i>G. menardii</i> fragmentation index, ^{230}Th -normalized carbonate accumulation rate, sediment-water modeling
Zamelczyk et al. (2012)	Fram Strait	Late Quaternary	Paleoceanographic changes and CaCO_3 dissolution	Distribution of planktonic forams, <i>N. pachyderma</i> and <i>T. quinqueloba</i> mean shell weight, test fragmentation index, total carbon, total organic carbon, $\delta^{13}\text{C}$, and $\delta^{18}\text{O}$
Slotnick et al. (2015)	Indian Ocean	Early Paleogene	CCD curve	Coccolith abundances, CaCO_3 content, bulk CaCO_3 $\delta^{13}\text{C}$

Table 1. Examples of approaches used to study calcite dissolution.

Sample # (Fig. 1)	Surface sediment sample	Latitude (N)	Longitude (W)	Water Depth (m)	Sources A = alkenones; C = coccoliths
1	2013 029 068 BC 0-1 cm	72.82	-77.43	1074	this study ^{A,C}
2	2013 029 063 TWC 0-1 cm	72.45	-72.91	833	this study ^{A,C}
3	2008 029 014 BC 0-1 cm	70.46	-64.66	2060	this study ^{A,C}
4	2013 029 0075 TWC 0-1 cm	69.55	-64.64	1484	this study ^{A,C}
5	2013 029 074 TWC 0-1 cm	69.52	-64.64	1482	this study ^{A,C}
6	2013 029 078 BC 0-1 cm	68.31	-63.80	1153	this study ^{A,C}
7	2008 029 070 TWC 0-1 cm	68.23	-57.62	444	this study ^{A,C}
8	2013 029 084 TWC 0-1 cm	67.78	-59.58	1359	this study ^{A,C}
9	2013 029 0048 TWC 0-1 cm	67.61	-61.97	935	this study ^{A,C}
10	2013 029 0051 BC 0-1 cm	67.46	-63.59	700.9	this study ^{A,C}
11	2008 029 008 TWC 0-1 cm	64.39	-58.13	857	this study ^{A,C}
12	SU90 I07 TWC 0-1 cm	63.87	-28.94	1625	Marmen (2000) ^A ; this study ^C
13	2013 029 0018 VVG 0-1 cm	61.54	-60.49	1065	this study ^{A,C}
14	2008 029 004 TWC 0-1 cm	61.46	-58.04	2674	this study ^{A,C}
15	91045 060 BC 0-1 cm	59.85	-33.58	2255	Marmen (2000) ^A ; this study ^C
16	91045 063 BC 0-1 cm	59.68	-30.36	1319	Marmen (2000) ^A ; this study ^C
17	91045 056 BC 0-1 cm	59.63	-36.13	3025	Marmen (2000) ^A ; Levesque (1995) ^C
18	91045 051 BC 0-1 cm	59.49	-39.31	2949	Marmen (2000) ^A ; this study ^C
19	90013 006 BC 0-1 cm	59.49	-45.87	1105	Marmen (2000) ^A ; this study ^C
20	91045 071 BC 0-1 cm	58.94	-28.74	2237	Marmen (2000) ^A ; this study ^C
21	90013 011 BC 0-2 cm	58.91	-47.08	2805	this study ^{A,C}
22	91045 028 BC 0-1 cm	56.62	-49.75	3992	Marmen (2000) ^A ; this study ^C
23	2005 033B 053 TWC 0-2 cm	54.87	-52.01	2357	Filippova (2016) ^A ; this study ^C
24	91045 014 BC 0-1 cm	54.74	-53.73	340	Marmen (2000) ^A ; this study ^C
25	91045 008 BC 0-1 cm	54.72	-55.58	301	Marmen (2000) ^A ; this study ^C
26	2006 040 0005 TWC 0-2 cm	54.71	-52.91	1148	Filippova (2016) ^A ; this study ^C
27	91045 005 BC surface	54.71	-56.45	530	Filippova et al. (2016) ^A ; this study ^C
28	91045 006 BC 0-2 cm	54.70	-56.45	534	this study ^{A,C}
29	91045 090 BC 0-1 cm	53.33	-45.26	3378	Marmen (2000) ^A ; this study ^C
30	91045 080 TWC 0-1 cm	53.06	-33.53	3024	Marmen (2000) ^A ; this study ^C
31	SU90 I03 TWC 0-1 cm	51.88	-39.78	2970	Marmen (2000) ^A ; this study ^C
32	91045 093 BC 0-1 cm	50.20	-45.69	3448	Marmen (2000) ^A ; this study ^C
33	2009 061 0114 PSH 0-2 cm	49.94	-45.05	1404	Filippova et al. (2016) ^A ; this study ^C
34	2003 033 029 TWC 0-2 cm	49.91	-44.88	3900	Filippova (2016) ^A ; this study ^C
35	2004 024 032 TWC 0-2 cm	49.51	-46.87	2805	Filippova et al. (2016) ^A ; this study ^C
36	2004 024 050 TWC 0-2 cm	49.42	-48.18	2293	Filippova et al. (2016) ^A ; this study ^C
37	2010 020 0058 TWC 0-2 cm	48.88	-51.86	346	Filippova et al. (2016) ^A ; this study ^C
38	2009 061 0117 PSH 0-2 cm	48.65	-45.17	1018	Filippova et al. (2016) ^A ; this study ^C
39	2009 061 0119 PSH 0-2 cm	48.61	-45.11	933	Filippova et al. (2016) ^A ; this study ^C
40	2006 039 005 TWC 0-2 cm	46.26	-54.98	184	Filippova (2016) ^A ; this study ^C
41	90031 049 BC 0-1 cm	46.19	-57.94	468	Marmen (2000) ^A ; this study ^C
42	90031 045 BC 0-1 cm	45.85	-57.59	473	Marmen (2000) ^A ; this study ^C
43	SU90 I02 TWC 0-1 cm	45.09	-39.43	1966	Marmen (2000) ^A ; this study ^C
44	2002 046 079 TWC 0-2 cm	44.85	-55.02	1865	Filippova (2016) ^A ; this study ^C
45	2006 048 0029 TWC 0-2 cm	44.77	-55.08	1861	Filippova (2016) ^A ; this study ^C
46	2004 024 006 TWC 0-2 cm	44.76	-54.38	585	Filippova (2016) ^A ; this study ^C
47	2006 048 0020 TWC 0-2 cm	44.75	-55.24	2230	Filippova (2016) ^A ; this study ^C
48	2002 046 077 TWC 0-2 cm	44.70	-54.45	988	Filippova (2016) ^A ; this study ^C
49	2006 048 0024 TWC 0-2 cm	44.63	-55.33	2343	Filippova (2016) ^A ; this study ^C
50	2002 046 074 TWC 0-2 cm	44.39	-55.44	2672	Filippova (2016) ^A ; this study ^C
51	96029 069 TWC 0-2 cm	43.07	-55.83	3506	Filippova et al. (2016) ^A ; this study ^C
52	2000 036 016 TWC 0-2 cm	42.88	-62.17	665	Filippova (2016) ^A ; this study ^C
53	2004 030 003 TWC 1-2 cm	42.83	-62.05	896	Filippova et al. (2016) ^A ; this study ^C
54	99036 052 TWC 0-2 cm	42.74	-62.12	1156	Filippova et al. (2016) ^A ; this study ^C
55	99036 055 TWC 0-2 cm	42.64	-62.12	1517	Filippova et al. (2016) ^A ; this study ^C
56	2006 046 0001 TWC 0-2 cm	42.49	-62.13	2095	Filippova (2016) ^A ; this study ^C
57	2000 036 030 TWC 0-2 cm	42.35	-62.03	2414	Filippova et al. (2016) ^A ; this study ^C
58	98039 010 TWC 0-2 cm	42.34	-54.71	4424	Filippova et al. (2016) ^A ; this study ^C
59	SU90 U01 BC 0-1 cm	42.03	-32.73	1970	Marmen (2000) ^A ; this study ^C
60	91020 013 TWC 0-2 cm	41.83	-62.33	3450	Filippova (2016) ^A ; this study ^C
61	91020 014 TWC 0-2 cm	41.79	-62.34	3532	Filippova et al. (2016) ^A ; this study ^C
62	91020 008 TWC 0-2 cm	41.47	-62.01	4136	Filippova et al. (2016) ^A ; this study ^C
63	98039 009 TWC 0-2 cm	41.44	-54.78	4649	Filippova et al. (2016) ^A ; this study ^C
64	91020 012 TWC 1-2 cm	41.27	-61.81	4342	Filippova et al. (2016) ^A ; this study ^C
65	99036 037 TWC 0-1 cm	40.26	-61.19	4791	Filippova et al. (2016) ^A ; this study ^C
66	99036 035 TWC 0-2 cm	40.22	-61.36	4792	Filippova et al. (2016) ^A ; this study ^C

Table 2. List of surface sediment samples used in this thesis to identify the coccolith assemblages as well as the coccolith and alkenone concentrations.

Core #	Cruise	Latitude (N)	Longitude (W)	Water Depth (m)	Core length (cm)	Core length analyzed (from the top; cm)
174	MSM09/02	68.53	-63.33	1559	777.5	371
170	MSM09/02	68.97	-59.39	1078	683	637
004	HU2008-029	61.46	-58.04	2674	896	699
094	HU91-045	50.20	-45.69	3448	1098	372
080	HU91-045	50.06	-33.53	3024	1735	287

Table 3. The long cores analyzed in this study, ordered by longitude (west to east).

Figure

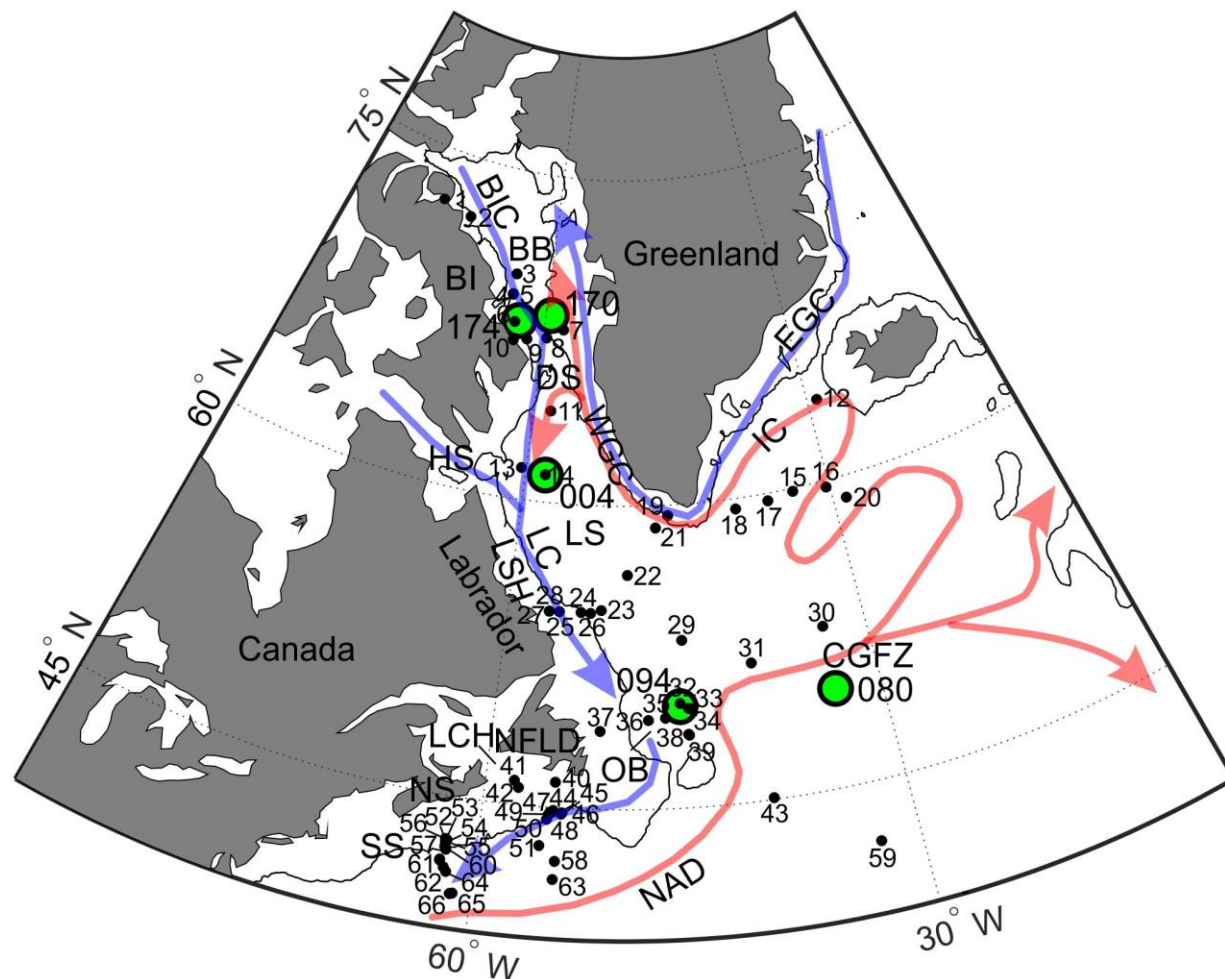


Figure 1. Map of core site locations for all sediment samples used in this thesis. Core-top sediment sites (*Chapter One*) are identified by black dots. The cores taken along the transect studied in *Chapter Three* are represented by the green dots. The sediment core used in *Chapter Two* is a case study of one of those cores: Core 004 from the northwest Labrador Sea. Modern surface currents are identified (warm in red and cold in blue; adapted from Lewis et al., 2012). The solid bathymetric contour marks the 500 m isobath. Acronyms on the map include the Irminger Current (IC), East Greenland Current (EGC), West Greenland Current (WGC), Baffin Bay (BB), Baffin Island (BI), Baffin Island Current (BIC), Davis Strait (DS), Hudson Strait (HS), Labrador Sea (LS), Labrador Current (LC), Labrador Shelf (LSH), Newfoundland (NFLD), Laurentian Channel (LCH), Nova Scotia (NS), Scotian Shelf (SS), North Atlantic Drift (NAD), and the Charlie-Gibbs Fracture Zone (CGFZ).

CHAPTER ONE

Carbonate dissolution and environmental parameters govern coccolith vs. alkenone abundances in surface sediments from the northwest North Atlantic

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Abstract

We determined the abundances and concentrations of coccoliths and alkenones in sixty-six surface sediment samples from the northwest North Atlantic to evaluate the role that surface ocean temperature, salinity, sea-ice cover, and productivity have on the regional distribution of these two biological remains produced by haptophytes in the photic zone. In areas with sea-ice cover of more than one month per year, coccolith and alkenone concentrations in sediments are extremely low to nil. Elsewhere, the distribution of coccolith taxa generally display strong relationships to water temperature, salinity, and productivity. *Coccolithus pelagicus* is associated with low summer sea-surface temperature ($< 8^{\circ}\text{C}$) and relatively high summer sea-surface salinity (> 33.5), whereas *Helicosphaera carteri* seems to follow the path of the North Atlantic Drift. The distribution of *Emiliania huxleyi*, the dominant alkenone producer, is not strongly correlated with that of alkenones. Calcite dissolution in shelf sediments could explain the occurrence of alkenones in the absence of coccoliths. However, alkenone production by non-calcifying haptophytes and alkenone degradation seem to also exert some control on alkenone concentrations in surface sediments, thus blurring alkenone abundance links with coccolithophorid production and relative preservation.

Highlights

- Sea-ice cover limits the accumulation of both coccoliths and alkenones in sediments.
- Calcite dissolution in shelf sediments may explain the accumulation of alkenones in the presence of few or no coccoliths.
- Non-calcifying haptophytes most likely produce alkenones in nearshore environments.

Keywords

Coccoliths, alkenones, dissolution, preservation, Scotian Slope, Labrador Sea, Baffin Bay

1. Introduction

Coccolithophores (Jordan and Chamberlain, 1997) are among the dominant primary producers and the most important pelagic calcifying organisms in the modern ocean (Baumann et al., 2004; Hay, 2004; Monteiro et al., 2016). Coccolithophores take up inorganic carbon in the photic layer. They are grazed upon by zooplankton, die, and sink, thus sequestering carbon to the deep ocean (Ducklow et al., 2001). Calcite export through coccolith production, sinking, and sedimentation thus regulate ocean alkalinity and pH (Schiebel, 2002; Henehan et al., 2016).

Coccolithophores thrive in the photic zone of mostly stratified, oligotrophic water masses (Baumann et al., 2005). They are protected within an exoskeleton composed of numerous calcite plates, or coccoliths, which can be preserved in the sedimentary record and may even represent a substantial part of sedimentary fluxes (Baumann et al., 2005). The species composition of coccolith assemblages on the seafloor depends first on the temperature of the surface waters in which they develop (e.g., McIntyre and Bé, 1967) and may thus preserve valuable paleoceanographic information. However, coccoliths can be highly altered not only when sinking in the water column but also in surface sediments through early diagenetic processes and thus sedimented coccoliths, in abundance and in their taxonomic composition, may represent only a fraction of the coccoliths that were originally produced in surface waters (Andruleit, 1997; Milliman et al., 1999; Kinkel et al., 2000). Nonetheless, coccolith assemblages may be used to infer past ocean conditions as their distribution in sediments is generally tightly linked to the properties of surface water masses (e.g., Eide, 1990; Samtleben and Schröder, 1992; Baumann and Matthiessen, 1992; Levesque, 1995; Baumann et al., 1999; Boeckel and Baumann, 2004).

Some coccolithophore species, including the cosmopolitan *Emiliania huxleyi* and the closely related *Gephyrocapsa oceanica*, synthesize long-chain unsaturated ketones (alkenones;

C₃₇—C₃₉; de Leeuw et al., 1980; Volkman et al., 1980; 1995; Brassell et al., 1986; Conte et al., 1998; Theroux et al., 2010). Among the haptophytes, *Emiliania huxleyi* is the dominant open ocean alkenone producer (Volkman et al., 1980; Marlowe et al., 1984a,b, 1990; Conte et al., 1994; Rosell-Melé et al., 2000). This species is the most widespread coccolithophore in the modern ocean, ranging from the subpolar to the oligotrophic tropical ocean (Winter et al., 2014). Because alkenone unsaturation (the number of double bonds) depends on growth temperatures (Brassell et al., 1986; Prahl and Wakeham, 1987), alkenones are widely used as a means to reconstruct past sea-surface temperatures (e.g., Prahl and Wakeham, 1987; Prahl et al., 1988; Sikes and Volkman, 1993; Müller et al., 1998). Core-top calibrations and laboratory culturing of *Emiliania huxleyi* under controlled temperatures have helped with establishing relationships between alkenone unsaturation and water temperature (Prahl and Wakeham, 1987; Müller et al., 1998). Other taxa have also been cultured, including *Gephyrocapsa oceanica* (Conte et al., 1995; Volkman et al., 1995), *Calcidiscus leptoporus* (Dudley et al., 1986; Candelier et al., 2013), and *Coccolithus pelagicus* (Langer et al., 2006; Marlowe et al., 1990). Among these species, only *Gephyrocapsa oceanica* produce alkenones. It is also worth mentioning that some non-calcifying haptophytes from coastal seas, estuaries, and lakes also produce alkenones (Marlowe et al., 1984a; 1990; Coolen et al., 2004; de Vargas and Probert, 2004; Malinverno et al., 2008; Bendif et al., 2013).

In this study, we analyzed and compiled abundances and compositions of coccoliths and alkenones from surface sediments of the northwest North Atlantic, in relation to surface water mass properties (salinity, temperature, sea-ice cover), local production, and carbonate preservation. This study expands on earlier unpublished work by Levesque (1995) and Marmen

(2000) and is one of the few studies confronting data from both coccoliths and alkenones of surface sediment samples.

2. Regional context

2.1 Hydrography

The study area (Fig. 1) includes Baffin Bay, the Labrador Sea, the Nova Scotian margin off southeast Canada, and offshore sites of the northwest North Atlantic, and is thus characterized by a wide range of sea-surface conditions. Along the Greenland margin, the West Greenland Current flows northwards through eastern Davis Strait, carrying the cold and low saline ($-1.8\text{ }^{\circ}\text{C}$; practical salinity (S_P) ≤ 34.5) waters of the East Greenland Current over the shelf (Cuny et al., 2002), with a warmer and saltier subsurface deeper component originating from the Irminger Current (Ribergaard et al., 2008). In the west, off Baffin Island, the Baffin Island Current flows southward through the western Davis Strait and into the Labrador Sea, where it mixes with Hudson Strait outflow water to become the Labrador Current (Percy and Fife, 1993; Lazier and Wright, 1993). The Labrador Current then flows southwards along the Labrador coastline, parallel to the coastline of eastern Newfoundland and along the Scotian Slope, off Nova Scotia.

In central Baffin Bay, the water column is strongly stratified, and intermediate-to-bottom waters are undersaturated with respect to both calcite and aragonite (Azetsu-Scott et al., 2010). In contrast, in the Labrador Sea, intermediate-to-deep waters are well ventilated and supersaturated with respect to calcite (Azetsu-Scott et al., 2010). Vertical convection in the

central Labrador Sea through the formation of the Labrador Sea Water (LSW) contributes to the formation of North Atlantic Deep Water as part of the Atlantic Meridional Overturning Circulation (Yashayaev, 2007; Jones and Anderson, 2008; Yashayaev and Loder, 2009). The LSW forms following surface cooling during severe winters (Clarke and Gascard, 1983) and spreads throughout the deep North Atlantic (Yashayaev et al., 2007).

Spring blooms, which occur in May through June in the Labrador Sea (Balaguru et al., 2018), in response to the seasonal increases in irradiance and stratification, track poleward into summer and are one of the most characteristic features of seasonal primary productivity in the North Atlantic (Yool et al., 2015). The magnitude and timing of blooms play a fundamental role in the marine food webs and in carbon export (Cushing, 1990; Briggs et al., 2011; Cole et al., 2012).

2.2 Previous coccolith studies from the study area

Coccolith assemblages have been documented in a few surface samples (the first centimeter) from the Labrador Sea by Levesque (1995) and Solignac et al. (2008). Data from these samples suggest that the abundance of *Coccolithus pelagicus* is inversely related to sea-surface temperatures.

Records of coccolith abundances in cored sequences have also been used to infer changes through time in surface ocean conditions. For example, Rahman and de Vernal (1994) proposed that the dearth of coccoliths in the glacial section of two sediment cores collected in the Labrador Sea (91045-094PC, 50.20 °N, 45.69 °W, adjacent to #32; and 90013-013PC, 58.21 °N, 48.37 °W, adjacent to #21; Fig. 1) was due to the very low surface water temperatures of the Last Glacial Maximum (LGM). Increased abundances of coccoliths in the deglacial section of the

same cores were linked to the penetration of relatively warm North Atlantic water into the Labrador Sea (Rahman and de Vernal, 1994).

Rosell-Melé et al. (2011) studied both alkenones and coccoliths from glacial debris flows deposited in the Labrador Sea and in Baffin Bay. They proposed that the ratio of allochthonous coccoliths (identified by the presence of extinct pre-Quaternary species found using a polarized light microscope) to total alkenone concentrations might be useful to identify sites in the glacial North Atlantic that had received significant amounts of allochthonous alkenones through ice-rafted debris. Although, these authors also concluded that the presence of allochthonous coccoliths in cored sediments does not lead to necessarily infer the presence of allochthonous alkenones.

2.3 Previous alkenone studies from the study area

A few studies have reported alkenone concentrations in sediment samples retrieved from the northwest North Atlantic including the Labrador Sea, Baffin Bay, and Nova Scotia margins. The above-mentioned study, Rosell-Melé et al. (2011), yielded relatively warm temperature estimates based on the alkenone unsaturation ratio; this led Rosell-Melé et al. (2011) to infer advection with ice-rafted debris, in the imbedding glacial debris, of reworked alkenones.

Marmen (2000) and Filippova et al. (2016) presented alkenone data from several surface sediment samples (0–2 cm) of the Labrador Sea that revealed a relationship between alkenone unsaturation and sea-surface temperatures similar to those reported in other studies from the North Atlantic (Marlowe et al., 1984a; Brassell et al., 1986; Prahl and Wakeham, 1987; Müller et al., 1998; Sikes and Sire, 2002; Sire et al., 2002; see Filippova et al., 2016 for synthesis).

Marmen (2000) noted that sediments from Nova Scotia margin sites have elevated alkenone concentrations, suggesting high coccolithophore productivity, whereas sediments collected on the Labrador Shelf contain no traces of alkenones.

Studies have related the abundance of $C_{37:4}$ in marine sediments, thus coccolithophore production in the overlying surface ocean, to salinity or meltwater inputs (using $\%C_{37:4}$). For example, Sicre et al. (2002) showed that $\%C_{37:4}$ was correlated to both temperature and (even more so) to salinity, however, their samples were suspended sediment samples obtained in the surface waters of the North Atlantic and the Nordic Seas. Moros et al. (2016) studied sediment cores in Disko Bugt and reported low alkenone concentrations and a relatively high $\%C_{37:4}$. They used $\%C_{37:4}$ as an indicator of changes in meltwater discharge from the Greenland Ice Sheet, as the $\%C_{37:4}$ is higher at lower surface salinities in polar and subpolar waters (Rosell-Melé, 1998; Rosell-Melé et al., 2002; Sicre et al., 2002; Harada et al., 2003; Bendle et al., 2005; Blanz et al., 2005). Lochte et al. (2019, 2020) interpreted downcore changes in $C_{37:4}$ abundance to imply variable sea-ice conditions and related changes in species composition along the Labrador Shelf during the Holocene.

3. Materials and Methods

Sixty-six core-top (surface, 0–1 cm, and 0–2 cm) sediment samples from trigger weight (TWC), box (BC), push (PSH), and Van Veen Grab (VVG) cores, collected during several oceanographic research cruises between 1990 and 2013 were analyzed in this study (Table 1). Alkenone data were available for fifty of these samples (Filippova et al., 2016), we thus simply added coccolith counts for these samples. We determined both alkenones and coccoliths in

fifteen other samples. Both alkenone and coccolith data were available for one sample; published respectively by Marmen (2000) and Levesque (1995).

Coccolith specimens were counted on microscope slides after preparation of ca. 0.01 g of dry sediment, using a polarizing optical transmitted light microscope at a magnification of 1250x (de Vernal et al., 2010). Coccolith census data were gathered by counting all coccolith specimens, with identification at the species level. Identifications were made based on the Nannotax3 online database of polarized light microscope images (Young et al., 2017; see Appendix A for the list of taxa identified). *Gephyrocapsa muelleri* and *Gephyrocapsa oceanica* were counted and reported as *Gephyrocapsa* spp. due to their small size and minor abundance. When coccoliths were rare or absent, 100 optical fields were examined. Once 300 counts of *Emiliania huxleyi* coccoliths per sample were obtained or after counting all coccoliths in 100 optical fields, total coccolith concentrations (coccoliths g⁻¹), assuming even distribution in the petri dish, were computed as follows:

$$(1) \quad C_{\text{coccolith}} = [(n \times (rp)^2)/(nc \times (rc)^2)]/p_{\text{se}}$$

where n is the total number of counted coccoliths, rp is the radius of the petri dish (cm), nc is the number of optical fields (no units), rc is the radius of the optical field (cm), and p_{se} is the weight of the subsample (g).

Total alkenone concentrations in bulk sediments were measured using high performance liquid gas chromatography (GC) at Dalhousie University following the protocol described in Kienast et al. (2006, 2012). Total lipids were extracted from 2 to 4 grams of dried and homogenized sediments using an Accelerated Solvent Extraction (ASE) system. Extracts were saponified and purified by elution through a silica column. The purified extracts were then

analyzed by GC using flame ionization detection (GC-FID) to obtain total alkenone concentrations ($C_{37:2}$, $C_{37:3}$, and $C_{37:4}$ alkenones; ng g^{-1} ; detection limit $< 10 \text{ ng g}^{-1}$). We report the percentage of $C_{37:4}$ alkenones ($\%C_{37:4}$) as the relative abundance of $C_{37:4}$ alkenones to the total abundances of $C_{37:2}$, $C_{37:3}$, and $C_{37:4}$ alkenones. Alkenone-based temperatures ($U_{37}^{\text{K}}\text{-SST}$) were derived from the alkenone unsaturation index (U_{37}^{K}):

$$(2) \quad U_{37}^{\text{K}} = ([C_{37:2}] - [C_{37:4}]) / [C_{37:2} + C_{37:3} + C_{37:4}]$$

where $C_{37:2}$, $C_{37:3}$, and $C_{37:4}$ are the di-, tri-, and tetra-unsaturated C_{37} alkenones, respectively (Brassell et al., 1986). Temperatures ($^{\circ}\text{C}$) were estimated using the following equation (Müller et al., 1998):

$$(3) \quad U_{37}^{\text{K}}\text{-SST} = [(U_{37}^{\text{K}} - 0.044) / 0.033]$$

Forty-four of the sixty-six sediment samples were analyzed for their organic and inorganic carbon contents (dry weight percentages) and for their stable carbon isotopic composition of the organic matter ($\delta^{13}\text{C}_{\text{org}}$; ‰ vs. Vienna Pee Dee Belemnite or VPDB, the international reference standard for carbon isotopes). The other samples were not analyzed for their carbon content because not enough material was available. First, 9 to 10 mg of bulk sediments were analyzed for their total carbon and nitrogen contents using an NC 2500TM elemental analyzer (Carlo-ErbaTM). The procedures for organic carbon and $\delta^{13}\text{C}_{\text{org}}$ measurement were as follows: Several hundred milligrams of bulk dry sediments were treated with 10 ml of a 1 N HCl solution in centrifuge tubes. Acidified solutions were mixed twice by hand, centrifuged for ten minutes, and the supernatant was siphoned off by pipette. The sediments were then rinsed twice with distilled water, and subsequently frozen and freeze-dried. Treated and freeze-dried sediments were subsampled (5–6 mg) into tin capsules for organic carbon analysis with the

elemental analyzer. The average of the standard deviations of eight duplicates for total organic carbon measurements was ± 0.1 %. Inorganic carbon percentages (%CaCO₃) were obtained by subtracting the organic carbon from the total carbon. For $\delta^{13}\text{C}_{\text{org}}$ measurements, acid-treated sediment samples (~ 25 mg) weighed in tin capsules were analyzed at GEOTOP (Montréal, Québec) with a Micromass Isoprime 100 isotope ratio mass spectrometer coupled to an Elementar Vario MicroCube elemental analyser in continuous flow mode. Two internal reference materials ($\delta^{13}\text{C}_{\text{org}} = -28.73 \pm 0.06$ ‰ and -11.85 ± 0.04 ‰) were used to normalize the results on the NBS19-LSVEC scale (Coplen et al., 2006). The analytical uncertainty ($\pm 1\sigma$) was better than ± 0.1 ‰, using a reference material ($\delta^{13}\text{C}_{\text{org}} = -17.04 \pm 0.11$ ‰) that was analyzed as an unknown to assess the accuracy of the normalization. The average standard deviation for $\delta^{13}\text{C}_{\text{org}}$ of the study samples, based on seventeen duplicate measurements, was ± 0.16 ‰.

For each sample location, the mean of winter (January-February-March) and summer (July-August-September) sea-surface salinities (SSS) and temperatures (SST; °C) were compiled from the 1955–2012 $1/4^\circ$ dataset of the World Ocean Atlas (WOA 2013 V2; cf. Locarnini et al., 2013; Zweng et al., 2013; <https://www.nodc.noaa.gov/OC5/woa13/woa13data.html>) (summers and winters defined as per de Vernal et al., 2020). Sea-ice cover (months yr⁻¹ with greater than 50 % sea-ice concentration) was compiled from the 1955–2012 gridded dataset of the National Snow and Ice Data Center (NSIDC V1; Walsh et al., 2015; <http://nsidc.org/data/G10010>). Annual primary productivity (gC m⁻² yr⁻¹; 2002–2017) was compiled from the monthly files of Moderate Resolution Imaging Spectroradiometer (MODIS) R2018 (Behrenfeld and Falkowski, 1997; <http://orca.science.oregonstate.edu/2160.by.4320.monthly.xyz.vgpm.m.chl.m.sst.php>).

Multivariate analyses were performed using CANOCO 5 software (ter Braak and Šmilauer, 2012; Šmilauer and Lepš, 2014) to describe the relationship between coccolith

assemblages (percentages) and six environmental variables. A square root transformation was applied to the coccolith percentages to reduce the effect of dominant species (Palmer, 1993) and closed-sum percentage data (Butler, 1981), due to some 100 % totals. Rare taxa (*Pontosphaera discopora*, *Rhabdosphaera clavigera*, *Syracosphaera pulchra*, and *Reticulofenestra producta*) were included in the analysis so that the percentage sum of species equaled 100 %. The data were constrained, and the standard deviation was ± 1.7 %, so a linear redundancy analysis (RDA) (ter Braak and Šmilauer, 1998) was performed on the percentages of all coccolith species and environmental variables.

4. Results

4.1 Coccolith distribution

Coccoliths were found in forty-nine of the sixty-six sediment samples (Table 2; Fig. 2). The seventeen samples that did not contain coccoliths include all ten samples from Baffin Bay, one from Davis Strait, five from the Labrador Shelf, and one from the Scotian Slope (Fig. 2A). Samples from the North Atlantic and far offshore in the Labrador Sea contain up to 4.7×10^8 coccoliths g^{-1} , while sediment samples within 250 km of the coastline contain scarce or no coccoliths (Fig. 2B). The total coccolith concentrations increase offshore and are positively correlated to the %CaCO₃ of the sediments ($r^2 = 0.45$; $p = 0.00$; Fig. 2C), suggesting that micrite linked to coccolith production is a major component of the pelagic carbonate fluxes. Representative scanning electron micrographs of samples contain nil to abundant coccoliths (Appendix B).

The coccolith assemblages are dominated by a few species, including *Coccolithus pelagicus*, *Emiliania huxleyi*, and *Calcidiscus leptoporus* (Fig. 3; Table 3). *Coccolithus pelagicus* is the dominant species in sediment samples from the Labrador Sea/Orphan Basin. *Emiliania huxleyi* is the second most abundant species in all samples containing coccoliths. *Calcidiscus leptoporus* occurs in most samples, with its highest abundances along the Scotian Shelf (Fig. 3). *Umbilicosphaera sibogae* and *Helicosphaera carteri* are sparse and only found in a few samples from southern sites containing up to an average of ~ 2 % of coccolith species.

A redundancy analysis (RDA), a method to detect linear relationships on (dis)similarities generated by measures which may be non-linear (Legendre and Anderson, 1999), was performed with coccolith assemblages (percentage of taxa) and sea-surface parameters (salinity, temperature, sea-ice cover, and productivity) for forty-nine sites (Fig. 4; Tables 4 and 5), excluding the seventeen samples with no coccoliths. The first, second, and third RDA axes explain 75.6 %, 12.1 % and 9.0 % of the variance, respectively. Axis 1 is characterized by negative scores for most taxa except *Coccolithus pelagicus*. It shows a negative correlation with sea-surface temperatures ($r = -0.547$ and -0.735 in winter and summer, respectively) and annual primary productivity ($r = -0.577$), and a positive correlation with sea-ice cover ($r = 0.355$). Axis 2 is characterized by negative scores for *Calcidiscus leptoporus* and positive scores for *Helicosphaera carteri*, *Gephyrocapsa* spp. and *Umbilicosphaera sibogae*. It correlates positively with sea-surface salinities ($r = 0.726$ and 0.611 in winter and summer, respectively).

4.2 Alkenones and organic carbon

The total alkenone concentrations vary from less than 10 ng g^{-1} to greater than $11,000 \text{ ng g}^{-1}$ (Fig. 5; Table 6). Alkenones were detected in all samples but three - two of which are from

the Labrador Shelf and one is from offshore the Scotian Shelf. The highest alkenone concentrations ($> 11,000 \text{ ng g}^{-1}$) were recorded on the continental shelf within and close to the Laurentian Channel (Gulf of St. Lawrence; Fig. 1) - an area of high biological productivity (e.g., Sourisseau et al., 2006). Overall, alkenone concentrations are higher in core-top sediments from southern sites than from northern sites (Fig. 5).

The samples with high alkenone concentrations from the Scotian Slope and Laurentian Channel are characterized by relatively high organic carbon content ($> 2 \%$) (Figs. 5 and 6). A positive correlation between alkenone and organic carbon content is recorded among the ten samples containing more than 1000 ng g^{-1} of alkenones ($r^2 = 0.55$; $p = 0.02$; Fig. 7). The relationship is, however, not significant for samples containing less than 1000 ng g^{-1} of alkenones ($r^2 = 0.00$; $p = 0.82$).

The weak relationship between total alkenone concentration and organic carbon content for the full range of alkenone concentrations from our study area may reflect the diversity of the organic matter sources, possibly including terrestrial material. Significant contributions of terrestrial organic matter is certainly implied by the low $\delta^{13}\text{C}_{\text{org}}$ values reaching as low as -25.7 at some sites (Fig. 5). At a number of the coastal sites in the Labrador Sea and Baffin Bay, where high organic carbon and low alkenone content are recorded, the $\delta^{13}\text{C}_{\text{org}}$ values range from -22.8 to -22.0 ‰ , suggesting a predominantly marine origin of organic carbon. Other samples from the Scotian Slope with high organic carbon ($2.1\text{--}4.0 \%$) and high alkenone concentrations (3500 to 6260 ng g^{-1}), have $\delta^{13}\text{C}_{\text{org}}$ values as low as -24.7 ‰ , indicative of some contributions from terrestrial organic matter (e.g., Meyers, 1997). Fourteen samples from the continental margins of Newfoundland and Labrador are characterized by $\delta^{13}\text{C}_{\text{org}}$ values as low as -25.7 ‰ , also suggesting significant contributions of terrestrial inputs (Fig. 5). Offshore, in the Labrador Sea

and the North Atlantic, the $\delta^{13}\text{C}_{\text{org}}$ values average -23 ‰, which reflect mostly marine inputs (Meyers, 1997; Muzuka and Hillaire-Marcel, 1999).

Our dataset includes $\text{C}_{37:4}$ alkenone concentrations for many samples (Fig. 5 and 6) throughout our study area (Table 6). There are eight samples where $\text{C}_{37:4}$ alkenones were not detected (nd) and thirty-one samples where $\text{C}_{37:4}$ alkenones were not determined (Table 1). High $\%\text{C}_{37:4}$ ($> 10\%$) occur in some Baffin Bay and Labrador Sea/Shelf sites (Fig. 6), generally corresponding to sites where the sediments contain few or no coccoliths (Fig. 3) and low alkenone concentrations (Fig. 5).

4.3 Alkenone-coccolith relationships

The alkenone concentrations were compared to the coccolith concentrations and major species percentages (Fig. 8). There is a positive relationship between total alkenone and coccolith concentrations along the Scotian Slope/Rise sites ($r^2 = 0.77$; $p = 0.00$; red circles; Fig. 8A-B). However, when considering the overall study area, there is no relationship between alkenone and coccolith concentrations ($r^2 = 0.00$; $p = 0.99$; Fig. 8C). This is also the case for total alkenones versus *Gephyrocapsa* spp. ($r^2 = 0.00$; $p = 0.78$) and the non-alkenone producer *Coccolithus pelagicus* ($r^2 = 0.00$; $p = 0.47$). Over the entire study area, the alkenone-species relationship is only weakly positive for total alkenones versus *Emiliana huxleyi* ($r^2 = 0.20$; $p = 0.00$) and for total alkenones versus the non-alkenone producer *Calcidiscus leptoporus* ($r^2 = 0.24$; $p = 0.00$).

4.4 Sea-surface properties versus alkenone and coccolith concentrations

The study area encompasses temperate to arctic zones characterized by a wide range of sea-surface conditions (Fig. 9; Appendix C). At sites where the practical salinity is lower than 32, there are no coccoliths and the alkenone concentrations are exceptionally low, with the exception of the Laurentian Channel sites, where there are rare coccoliths but high alkenone concentrations (Fig. 9A-B). Coccolith concentrations peak in samples at locations where summer sea-surface temperatures range from 7 to 21 °C, whereas alkenone concentrations are high in samples from locations where summer sea-surface temperatures are 15 to 17 °C (Fig. 9C-D). At sites where seasonal sea-ice cover spans more than one month per year, there are rare or no coccoliths and almost no alkenones (Fig. 9E-F). Hence, both coccoliths and alkenones seem excluded from cold regions marked by sea-ice cover as well as from zones characterized by low salinity, with exception of the Laurentian Channel (Fig. 9). Coccoliths are abundant in sediments at sites where primary productivity ranges from 155–280 gC m⁻² yr⁻¹, but alkenone concentrations are highest in sediments where surface productivity is ~ 240 gC m⁻² yr⁻¹ (Fig. 9G-H). Sea-surface conditions also affect the unsaturation of alkenones, specifically the percentages of the tetra-unsaturated alkenone (%C_{37:4}; Fig. 10). The %C_{37:4} values are generally low where the alkenone and coccolith concentrations are high (see Fig. 10A-B) but they are high (> 20 %) at some sites characterized by the presence of sea-ice cover (> 1 month yr⁻¹) and low sea-surface temperatures (< 5 °C in the summers), salinities (< 34 year-round), and productivity (< 175 gC m⁻² yr⁻¹) (Fig. 10C-F).

4.5 Alkenone unsaturation and temperature

The alkenone-based U_{37}^K -SST estimates range from -17.5 to +29.0 °C. The extreme values (-17.5 °C and +29.0 °C) are outright unrealistic or impossible. Moreover, most samples yield large discrepancies (twice the standard deviation) of the calculated T_{summer} values with WOA summer SSTs, with both warm and cold biases in Baffin Bay, the southeast Labrador Sea, and Orphan Basin, as well as along the Scotian Slope and rise and offshore southern Greenland (Fig. 11), in agreement with the synthesis by Filippova et al. (2016). Only nine of the study samples provided U_{37}^K -derived SST estimates that fall within two standard deviations of observed WOA summer SSTs (Table 4). Among the fifty-seven samples characterized by significant departures, forty-four yielded U_{37}^K -SSTs lower than expected and thirteen recorded U_{37}^K -SSTs higher than expected. Most of these samples are from Baffin Bay and the Labrador Shelf, at sites where alkenone concentrations are low and coccoliths are absent. The largest SST residuals (defined as U_{37}^K -SST — T_{summer}) occur mostly in samples with either high %C_{37:4} or no C_{37:4} alkenones (Fig. 12; Table 6). Baffin Bay samples with remarkably high %C_{37:4} tend to underestimate SSTs whereas low or undetectable (nd) %C_{37:4} overestimate SSTs (Tables 4 and 6).

5. Discussion

5.1 Coccoliths and surface water mass properties

Baumann and Matthiessen (1992) studied coccolith assemblages in five cores collected in the Norwegian-Greenland seas and found low species diversity, with a dominance of *Coccolithus pelagicus* and *Emiliania huxleyi*. We obtained similar results and observed that only a handful of

species dominate the surface sediments. In our study area, *Emiliania huxleyi* is one of the two dominant taxa (Fig. 3) and is particularly abundant in the high productivity areas as demonstrated by the RDA analysis (Fig. 4C; Appendix C).

Coccolithus pelagicus is known to live in cool (6–18 °C) waters and upwelling regions (Haq, 1980; Giraudeau and Bailey, 1995; Cachão and Moita, 2000; Parente et al., 2004; Sato et al., 2004; Palumbo et al., 2019). *Coccolithus pelagicus* dominates in our samples from the Labrador Sea, the Orphan Basin, and some samples from the Scotian Slope, where low sea-surface temperatures (< 8 °C in the summers) and relatively high sea-surface salinity ($S_P > 33.5$ in the summers) are found (Fig. 3; Appendix C). The distribution of *Coccolithus pelagicus* shows an inverse relationship with sea-surface temperatures, as previously reported by Levesque (1995) and Solignac et al. (2008) for the North Atlantic.

Other species that are useful indicators of water temperature include *Umbilicosphaera sibogae* and *Helicosphaera carteri*. These species are rare, but nonetheless are the fifth and sixth most abundant species in our study area (Fig 3). *Umbilicosphaera sibogae* is indicative of warm and oligotrophic surface waters (López-Otálvaro et al., 2008; Amore et al., 2012; Palumbo et al., 2013a, b, 2019). It is found exclusively in our southernmost samples (Fig. 3). *Helicosphaera carteri*, a warm-water species of high productivity settings (Ziveri et al., 2004), is observed in samples from the open ocean (#43 and #59; Fig. 1), with sparse occurrences in samples from the Scotian Slope. *Helicosphaera carteri* appears to be associated with the North Atlantic Drift, which is a relatively productive (Yool et al., 2015), warm (> 8 °C) and salty ($S_P > 35$) surface water current (Breckenfelder et al., 2017).

Calcidiscus leptoporus is a cosmopolitan species (McIntyre and Bé, 1967) that never reaches high abundances in surface waters but may dominate coccolith assemblages in sediments

due to its resistance to dissolution (McIntyre and McIntyre, 1971). Renaud and Klaas (2001) studied *Calcidiscus leptoporus* near Bermuda, and their results yielded no significant correlation between environmental parameters and the *Calcidiscus leptoporus* abundance in sediments. However, an earlier study of a global set of Holocene sediment samples indicated that temperature might be a significant factor influencing the distribution of *Calcidiscus leptoporus* (Knappertsbusch et al., 1997). In our study region, *Calcidiscus leptoporus* is common along the Scotian Slope and its occurrence corresponds to the warmest summer sea-surface temperatures ($> 17^{\circ}\text{C}$), but relatively low salinity ($S_P = 31\text{--}34$) (Fig. 3; Appendix C).

Overall, the distribution of coccolith taxa in our study area is in part consistent with their known affinity for certain environmental conditions. The strong correlations of RDA axes 1 and 2 with sea-surface temperature and salinity, respectively, suggests that larger datasets might be suitable for the development of transfer functions and analogue techniques to quantitatively reconstruct surface water properties. There are sources of errors to consider with the total annual primary productivity data (de Vernal et al., 2020). For example, these data were calculated using the vertical generalized production model (VGPM) algorithm applied to the 2002-2017 chlorophyll data provided by MODIS and the VGPM is a “chlorophyll-based” model that estimates primary productivity from chlorophyll, SST, and photosynthetically active radiation (Behrenfield and Falkowski, 1997). Linear interpolation is used where there are data gaps, within an 80 km radius (de Vernal et al., 2020). While there are errors associated with the values calculated from the model, another important point is to consider that the primary productivity values here are for total annual primary productivity, and not only coccolithophore productivity.

5.2 Coccoliths versus alkenones

Since *Emiliania huxleyi*, the second dominant species after *Coccolithus pelagicus*, is the main alkenone producer (Volkman et al., 1980), coccoliths and alkenones are expected to have similar distributions. However, alkenone concentrations are not strongly correlated to coccolith abundances in the study area overall ($r^2 = 0.00$; $p = 0.99$; Fig. 8C), except in the Scotian Slope area alone ($r^2 = 0.77$; $p = 0.00$; Fig. 8A-B). In the Baffin Bay and Labrador Shelf samples containing no coccoliths, alkenone concentrations range up to 81 ng g^{-1} . In samples from the Laurentian Channel, in which coccolith abundances are minimal, alkenone concentrations peak at $11,200 \text{ ng g}^{-1}$ (Fig. 8A). In contrast, at most sites in the deep Labrador Sea, where coccoliths are abundant, alkenone concentrations remain low ($< 208 \text{ ng g}^{-1}$). Hence, our results suggest that the fluxes of alkenones and of *Emiliania huxleyi* to the seafloor and their subsequent preservation in the sediments are driven by processes that differ between each other and from one region to another. These processes, discussed in detail below, could include any combination of calcite dissolution, preferential degradation of alkenones, and/or alkenone production by non-calcifying haptophytes.

5.2.1 Calcite dissolution

As foraminiferal tests and coccoliths settle through the water column, their calcite structures weaken or dissolve. Based on common dissolution markers (planktonic foraminifer data, their degree of fragmentation, and benthic-to-planktonic foraminifer ratios; Thunell, 1976; Berger, 1979) analyzed in ODP core 646 (Fig. 1), Aksu et al. (1989) inferred that there has been no substantial calcareous fossil dissolution in sediments of the northeast Labrador Sea over the past one million years. In contrast, the degree of preservation and fragmentation of foraminiferal

shells and the relative abundances of dissolution-resistant and dissolution-susceptible foraminifers imply significant calcite dissolution in Baffin Bay during interglacials according to Aksu (1983). The ratio of organic linings to benthic foraminifers has also been used to show dissolution throughout the Holocene in Davis Strait (de Vernal et al., 1992). It is noted, however, that Subhas et al. (2019) found that foraminifer dissolution does not necessarily imply coccolith dissolution, as these authors documented a preferential dissolution of foraminifers relative to coccoliths.

Figure 13 shows regional calcite dissolution based on the synthesis of prior studies using a variety of different foraminifer dissolution indices (Table 7). The compiled data indicate foraminifer calcite dissolution in shelf sediment samples throughout the study region, and in all Baffin Bay sediment samples (Fig. 13). Mucci et al. (2000) and Nesbitt and Mucci (2020) have shown that diagenesis-driven dissolution results from oxic remineralization of organic matter, which likely explains the presence of alkenones in the absence of coccoliths in organic-rich shelf sediments. This is consistent with carbonate dissolution in Labrador Shelf sediments preventing radiocarbon dating of postglacial sediments (e.g., Levac and de Vernal, 1997). Baffin Bay is also an area of intense modern calcite dissolution, as evidenced by undersaturated bottom waters (Azetsu-Scott et al., 2010) and dissolution indices from recent sediments (Table 7). In Baffin Bay, as on the Labrador Shelf, calcite dissolution can thus explain the presence of alkenones in the absence of coccoliths. This interpretation is also consistent with the overall correlation of coccolith abundances and carbonate concentrations in the samples reported here (Fig. 2C).

5.2.2 Lateral transport and allochthonous materials

Coccoliths and alkenones in sediments could be allochthonous (e.g., Benthien and Müller, 2000), advected laterally by ocean currents (Rühlemann and Butzin, 2006). Ice rafting also transports allochthonous material, including alkenones and pre-Quaternary coccolith species to the open ocean, especially during glacial stages (Rosell-Melé et al., 2011), but our core-top samples are inferred to represent recent deposition. IRD can also cause increased particle adhesion that can increase the flux of carbon to the seafloor. Bottom undercurrents, such as the Western Boundary Undercurrent (WBUC), can also cause winnowing, downslope transport, and re-deposition of sediment, as suggested by Müller et al. (1998) and documented for the Labrador Sea by Hillaire-Marcel et al. (1994a). However, transport by deep currents affects both dissolved organic compounds (such as alkenones) and coccoliths, as documented for the Labrador Sea by Maccali et al. (2020), based on deep-sea coral data, of course unless the material was impacted by dissolution in the source region. As a general rule, differential transport of alkenones and coccoliths should not be expected since both are brought to the seafloor rapidly within zooplankton fecal pellets (Roth et al., 1975; Honjo, 1976; Langer et al., 2007) and marine snow (e.g., Turner, 2002), regardless of the differences in settling velocities of individual coccoliths (Zhang et al., 2018; Gutiérrez et al., 2019). However, it cannot be totally discarded along the eastern North American margins due, in particular, to the strong influence of the WBUC on sedimentary processes along the margin down to the abyssal plain. Thus, some subsequent decoupling of alkenones vs. coccolith fluxes cannot be discarded but remain difficult to estimate based on the available information.

5.2.3 Preferential degradation

Preferential degradation of alkenones in marine sediments has been shown to yield higher (1 to 2 °C) than expected temperatures, as indicated by investigations of sediments where extensive degradation has occurred (Freeman and Wakeham, 1992; Gong and Hollander, 1999; Hoefs et al., 1998; Herbert, 2003; Freitas et al., 2017). Furthermore, $C_{37:4}$ is thought to degrade at a somewhat higher rate than $C_{37:3}$ and $C_{37:2}$ (the more unsaturated the alkenone, the higher the degradation rate; $C_{37:4} > C_{37:3} > C_{37:2}$; Rosell-Melé et al., 1995). Although $C_{37:4}$ is not present everywhere, consideration of $C_{37:4}$ alkenones in the temperature equation appears to improve the temperature estimates in cold regions (Rosell-Melé, 1998; Bendle and Rosell-Melé, 2004; Filippova et al., 2016), but the absence or scarcity of the $C_{37:4}$ alkenone in some northern sediment samples (e.g., Baffin Bay) could explain why extreme temperature offsets or double-digit residuals are observed for Baffin Bay sediments (Table 6; Fig. 13). It remains that the temperature residuals that we report are far higher than any offsets expected from potential alkenone degradation. It is also possible that extremely low alkenone concentrations in some of our study samples are the result of extensive overall alkenone degradation. As such, alkenone-derived temperature estimates should be used with caution with low alkenone concentrations ($< 10 \text{ ng g}^{-1}$) that yield large differences in sea-surface temperatures (Freitas et al., 2017). Alkenones, however, are generally well preserved, even in highly oxidizing sediments (Prahl et al., 2003). Given the relatively high organic carbon content of our samples (mean: 1 %; up to 4 %), high differential degradation rates of the relatively refractory alkenones (in comparison with amino acids, for example, see Muzuka and Hillaire-Marcel, 1999) is not the likely scenario to explain the alkenone distributions in our study area.

5.2.4 Non-calcifying haptophytes

Coccolith and alkenone abundance data from the water column in the Gulf of California led Malinverno et al. (2008) to conclude that the calcifying taxa *Emiliania huxleyi* and *Gephyrocapsa oceanica* account regionally for the observed alkenone concentrations. This may not apply everywhere: only a fraction of the coccolithophore populations calcify (Paasche, 2002), and not all alkenone producers calcify (Eltgroth et al., 2005; Versteegh et al., 2001). Even when alkenones are used as a proxy of coccolithophore production (Schneider et al., 1996; Schulte et al., 1999; Villanueva et al., 1998; Malinverno et al. 2008), mismatches between total alkenone and total coccolith production are possible (Ding et al., 2019), as appears to be the case in our study area. We suggest that the alkenones that accumulate in some of the surface sediments in our study area may have been produced by non-calcifying haptophytes (i.e., the Laurentian Channel). Non-calcifying haptophytes are commonly reported in littoral and coastal areas (de Vargas and Probert, 2004), which might disproportionally impact sample sites close to the coastline and/or samples influenced by riverine inputs, such as within the Laurentian Channel. Hence, the proliferation of non-calcifying haptophytes may best explain the extremely high alkenone concentrations in sediment samples #41 and #42 from the Laurentian Channel (Fig. 8).

Freshwater alkenone-producing haptophytes have attracted great interest in the paleoclimate community because their alkenones may be used as indicators of past climate (Schulz et al., 2000; Zink et al., 2001; Chu et al., 2005; Hou et al., 2016; Warden et al., 2016; Longo et al., 2016, 2018; Araie et al., 2018), notably for paleo-salinity reconstructions, as in the Baltic Sea (Warden et al., 2016), the Okhotsk Sea (Harada et al., 2008), and offshore West Greenland (Moros et al., 2016) in relation to freshwater or meltwater inputs. Our $\delta^{13}\text{C}_{\text{org}}$ dataset

indicates that terrestrial inputs are likely near the coast where the transport of freshwater alkenones to the sediments could be linked to fluvial inputs (e.g., the St. Lawrence Estuary and the Laurentian Channel). Terrestrial material, including freshwater alkenones, may have been transported to the sea, thus impacting alkenone-derived temperature estimates. Hence, the mixing of alkenones of various origins could explain the extreme temperature offsets and, to some extent, the weak positive correlation ($r^2 = 0.23$; $p = 0.01$) between %C_{37:4} and summer salinities. Rosell-Melé (1998) proposed that salinity variations could modulate changes in the sedimentary records of C_{37:4} alkenone abundances, a hypothesis supported by Sicre et al. (2002) who reported a strong correlation between %C_{37:4} and salinity. However, this salinity ‘indicator’ has not been consistent across the world oceans or even in the North Atlantic (Filippova et al., 2016), and any correlation between C_{37:4} alkenone abundance and salinity could be an artifact of the co-variation of salinities and temperatures in the North Atlantic (Sikes and Sicre, 2002).

6. Summary and Conclusions

Haptophytes are a significant component of the primary productivity in the ocean and their micropaleontological (coccolith) and biomarker (alkenone) remains in sediments are commonly used as proxies of paleoceanographic conditions, notably to reconstruct past sea-surface temperatures for alkenones. In high latitudes and subpolar basins, their use is equivocal because sea-ice cover, freshening/meltwater discharge, the saturation state of overlying waters with respect to carbonate minerals, differential long-distance transport by boundary currents, and diagenesis may modify their production and preservation. In this study, we presented the regional distribution of coccoliths and alkenones, both produced by haptophytes, in surface sediments from the northwest North Atlantic and adjacent subpolar seas. Our objectives were to

analyze and compile abundances and compositions of coccoliths and alkenones from surface sediments of the northwest North Atlantic, in relation to surface water mass properties (salinity, temperature, sea-ice cover), local production, and carbonate preservation.

Our main findings are as follows: The sparse to nil occurrences of both alkenones and coccoliths in areas covered by sea ice for one or more months per year serve to illustrate how significant biogenic fluxes related to haptophyte production are restricted to ice-free environments. In addition, the distribution of *Emiliania huxleyi*, which is the dominant alkenone producer in the modern ocean, does not correspond with that of alkenones. Whereas high alkenone concentrations and low coccolith abundances in some settings could suggest calcite dissolution, the U^{K}_{37} -SST values also point to heterogeneous sources of alkenones, especially in shallow areas. Our results indicate that coccolith assemblages correspond with summer temperatures, with *Coccolithus pelagicus* related to cold waters and *Helicosphaera carteri* likely associated with the North Atlantic Drift. Our results also suggest sedimentation of alkenones produced by non-calcifying haptophytes and/or freshwater alkenone sources along the coastal margins. Calcite dissolution in shelf sediments could explain the occurrence of alkenones in the absence of coccoliths. However, alkenone production by non-calcifying haptophytes and alkenone degradation seem to also exert some control on alkenone concentrations in surface sediments, thus blurring alkenone abundance links with coccolithophorid production and relative preservation.

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Figures

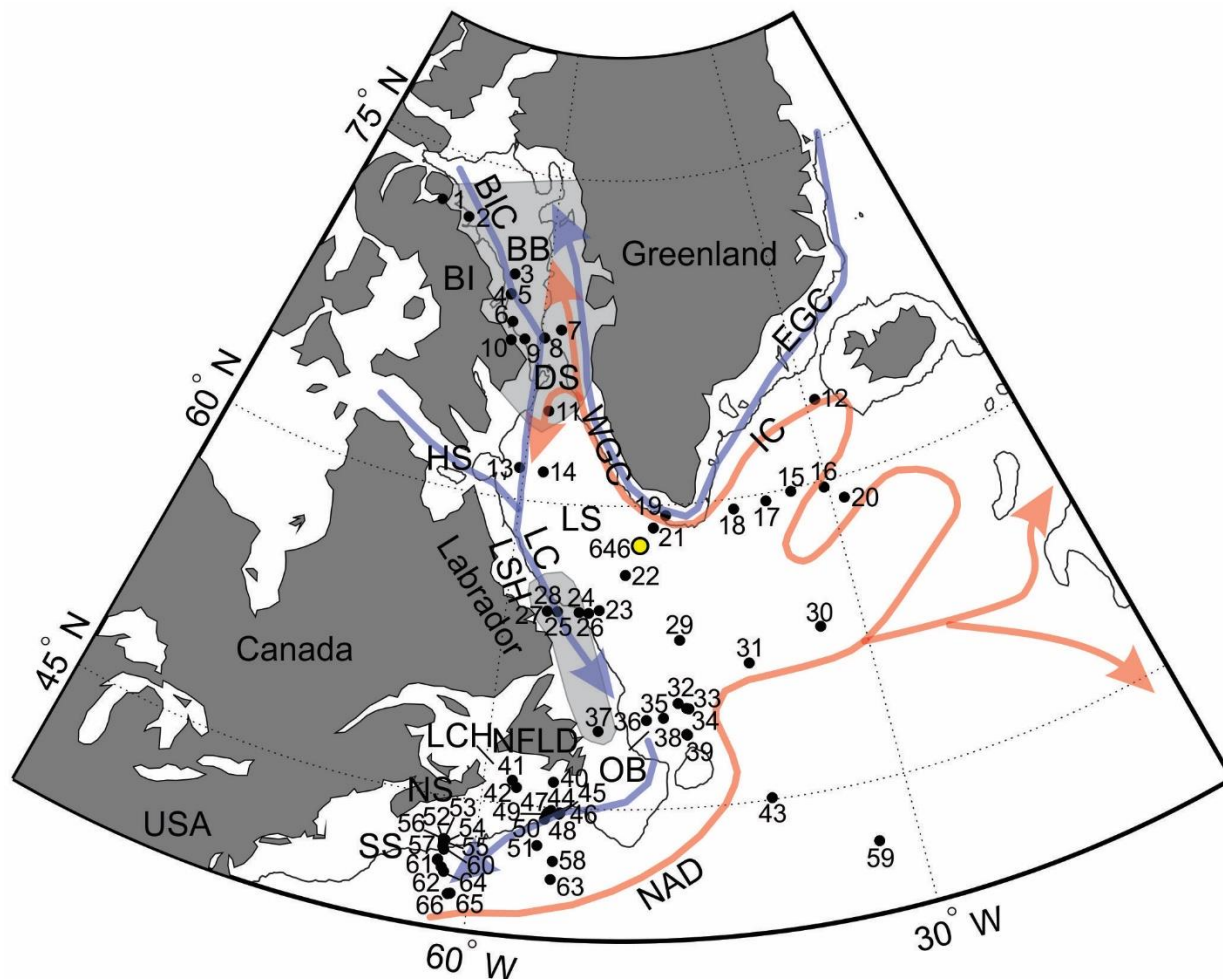


Figure 1. Map of the core site locations in the northwest North Atlantic. The black dots correspond to surface sediment samples (Table 1). The ODP core 646 is identified by the yellow dot. Relatively warm currents are in red and the relatively cold currents are in blue (adapted from Lewis et al., 2012). The light grey zones correspond to areas where surface sediment samples are devoid of coccoliths. The solid bathymetric contour marks the 500 m isobath. Acronyms on the map include the Irminger Current (IC), East Greenland Current (EGC), West Greenland Current (WGC), Baffin Bay (BB), Baffin Island (BI), Baffin Island Current (BIC), Davis Strait (DS), Hudson Strait (HS), Labrador Sea (LS), Labrador Current (LC), Labrador Shelf (LSH), Scotian Slope (SS), Newfoundland (NFLD), Orphan Basin (OB), Laurentian Channel (LCH), North Atlantic Drift (NAD), and Nova Scotia (NS).

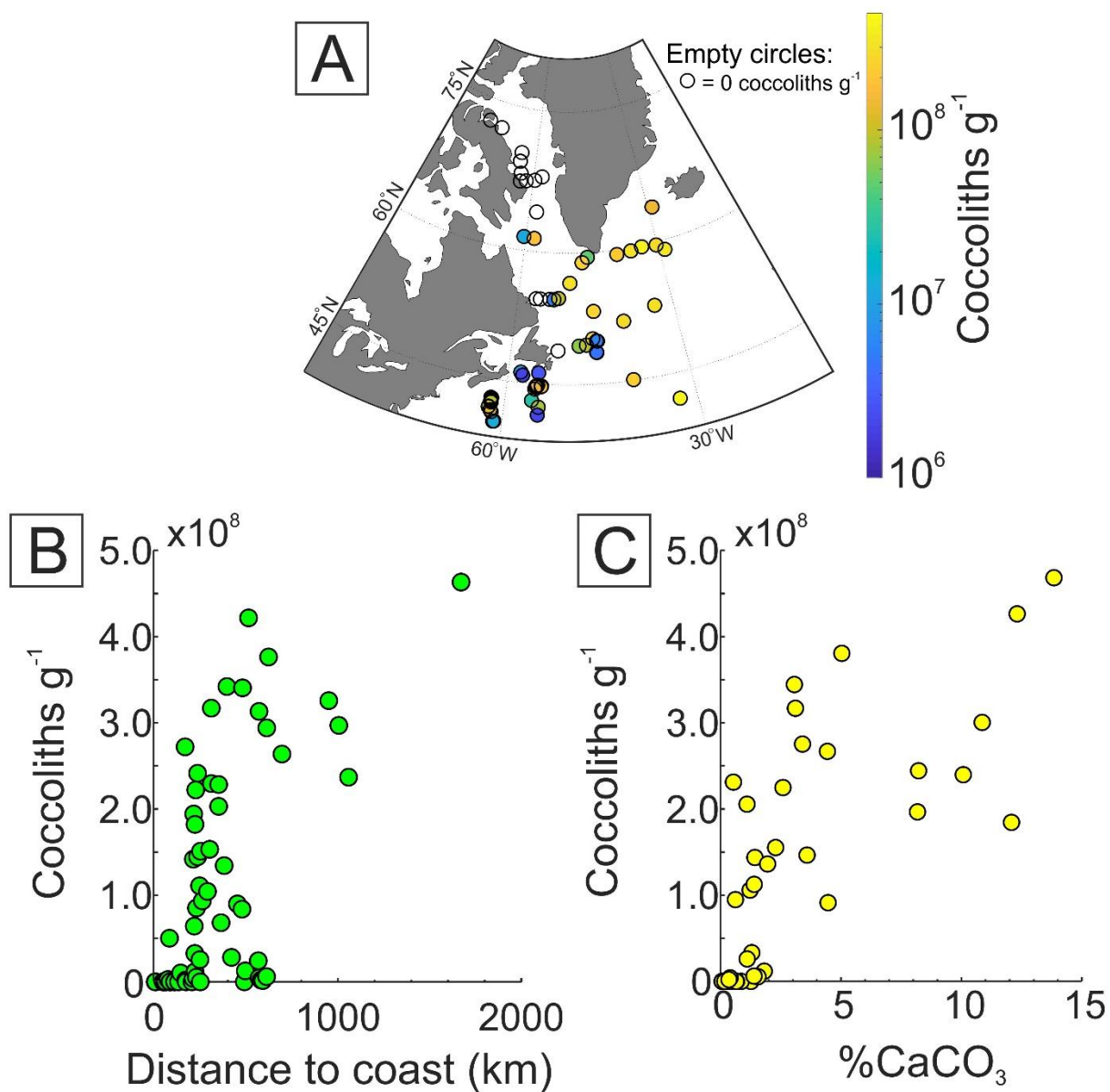


Figure 2. Coccoliths concentrations in geographical space (A), versus distance to coast (km) (B), and versus percentages of inorganic carbon ($\% \text{CaCO}_3$) (C). Empty circles on the map (A) indicate the absence of coccoliths.

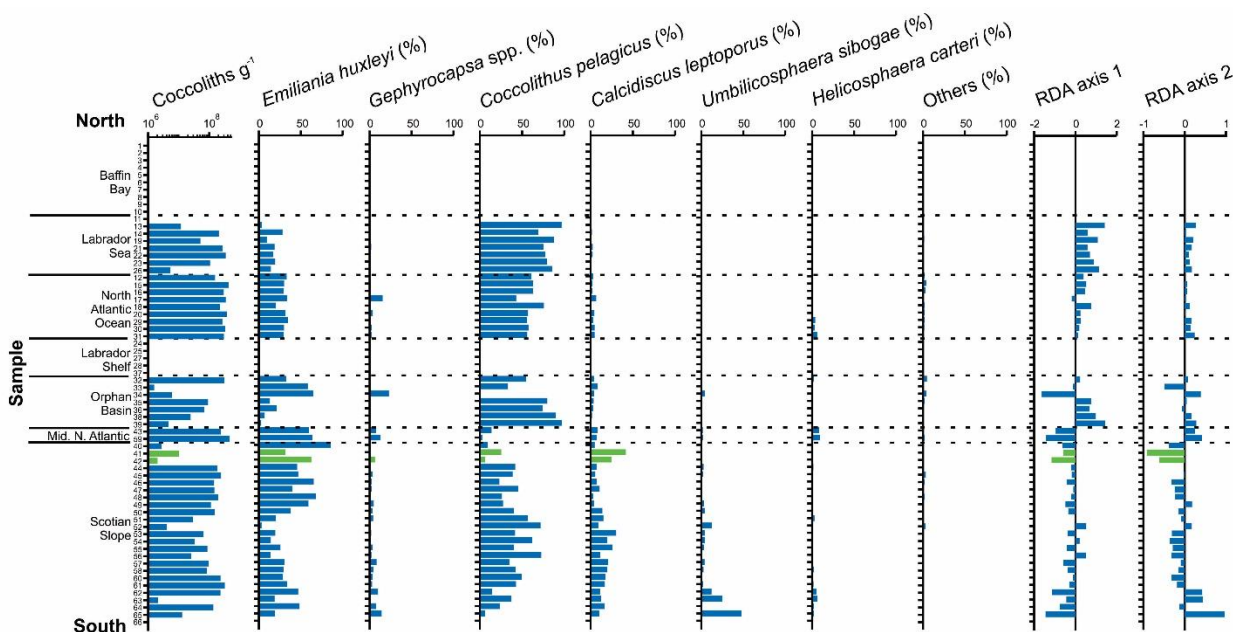


Figure 3. Coccolith concentrations and percentages of the main coccolith taxa in surface sediment samples plotted versus latitude. The scores of RDA axes 1 and 2 are derived from a redundancy analysis (RDA) of coccolith species and six environmental variables used in this study (Table 4). Data from the Laurentian Channel samples are shown in green. The data are zero if no data are presented among the coccolith concentrations and percent abundances. No data presented along the RDA axes mean that those particular samples were not analyzed in the RDA.

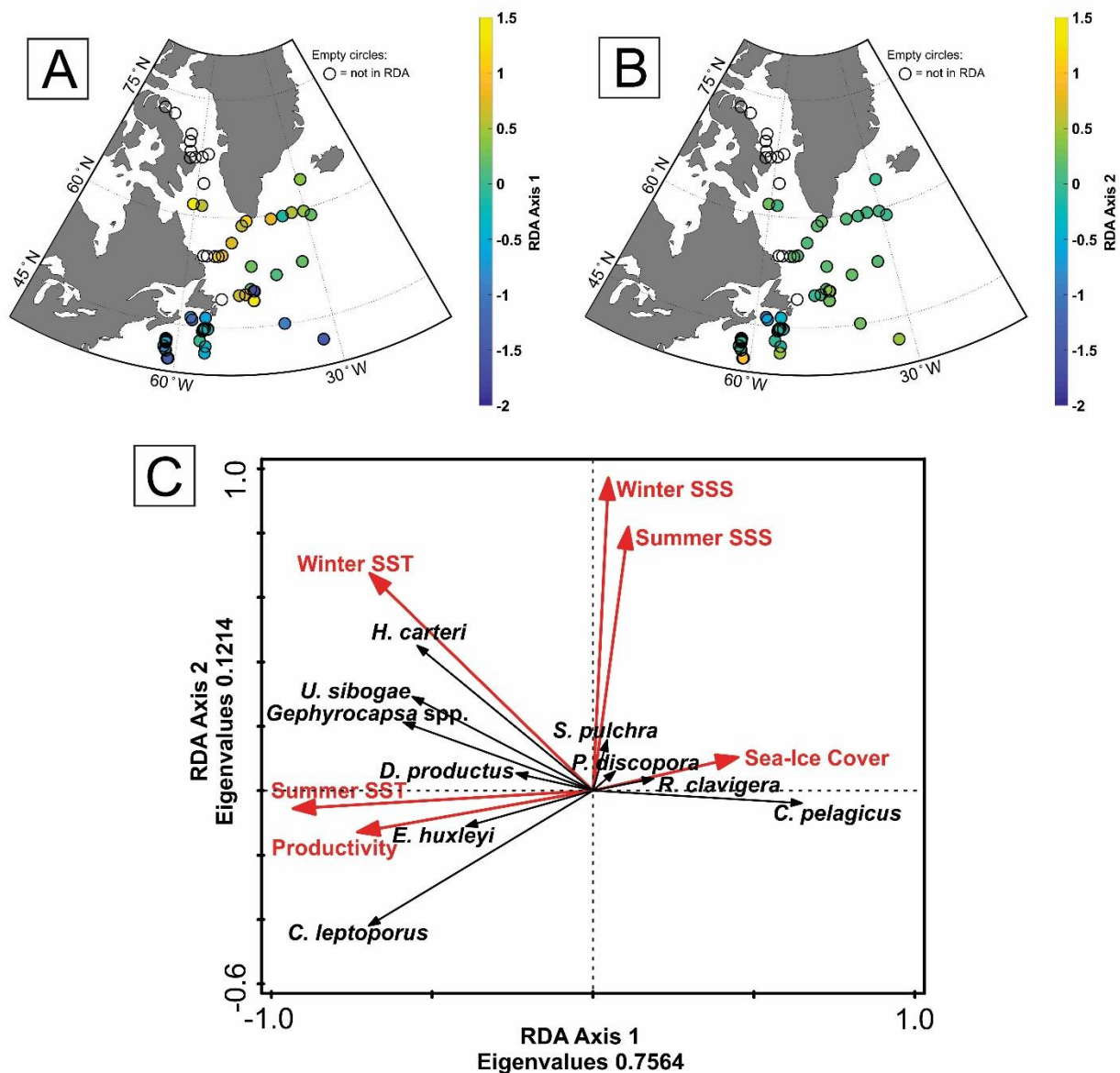


Figure 4. Results from the redundancy analysis (RDA) performed on coccolith taxa percentages and environmental parameters: distribution map of axis 1 (A) and axis 2 (B) scores. (C) Ordination diagram of axes 1 and 2.

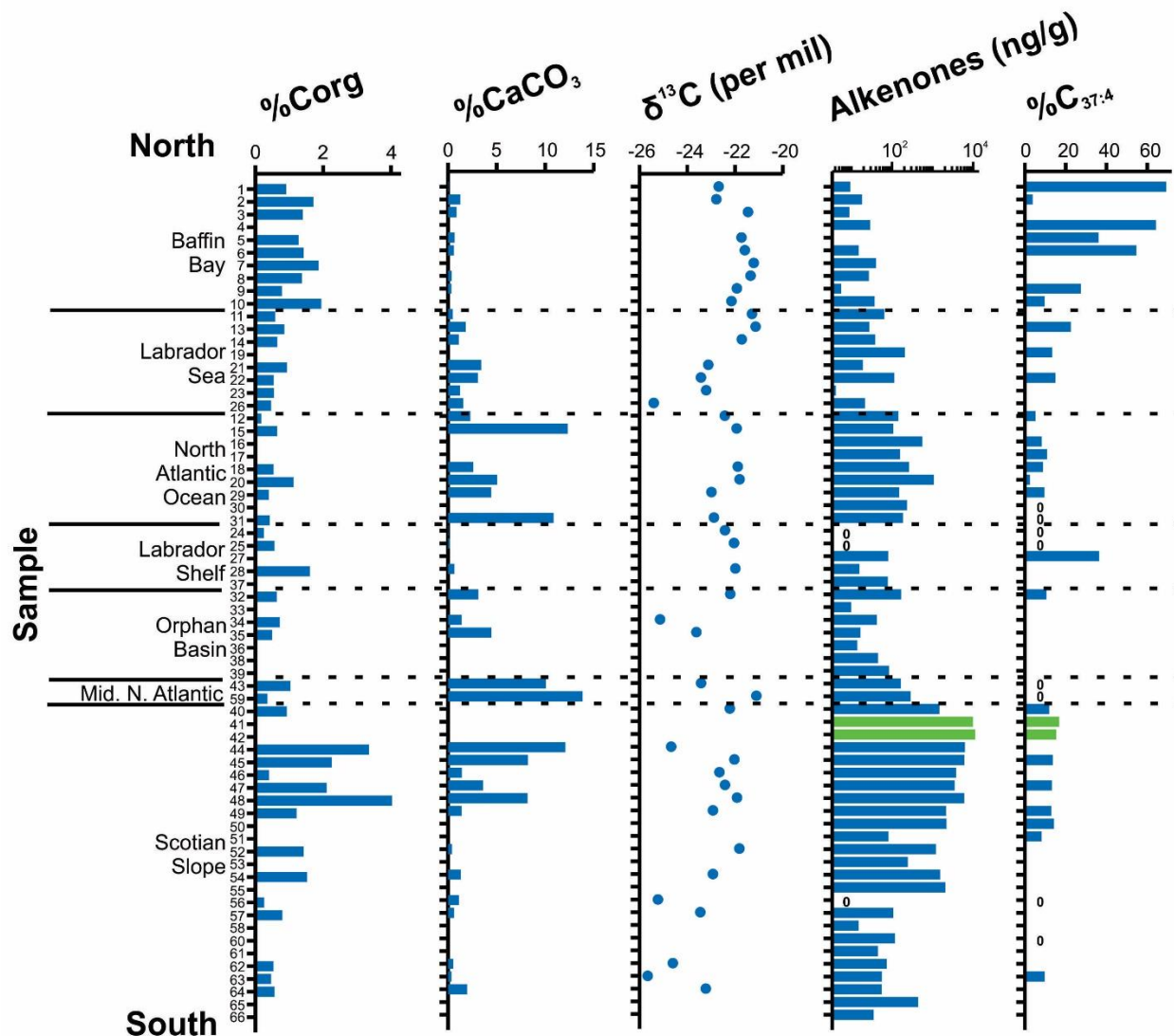


Figure 5. Organic carbon, inorganic carbon, $\delta^{13}\text{C}_{\text{Corg}}$, alkenone concentrations, and %C_{37:4}, where measured, are plotted versus latitude as in Figure 3. Data from the Laurentian Channel samples are shown in green. The zeros along the y-axis for alkenone concentrations and %C_{37:4} indicate zero ng g⁻¹ and zero %C_{37:4} for those samples. For %C_{37:4} only, no zero and no data presented indicate that %C_{37:4} was either not analyzed or is unknown. No data presented for %Corg indicate not analyzed, and no data for %CaCO₃ indicate not calculated.

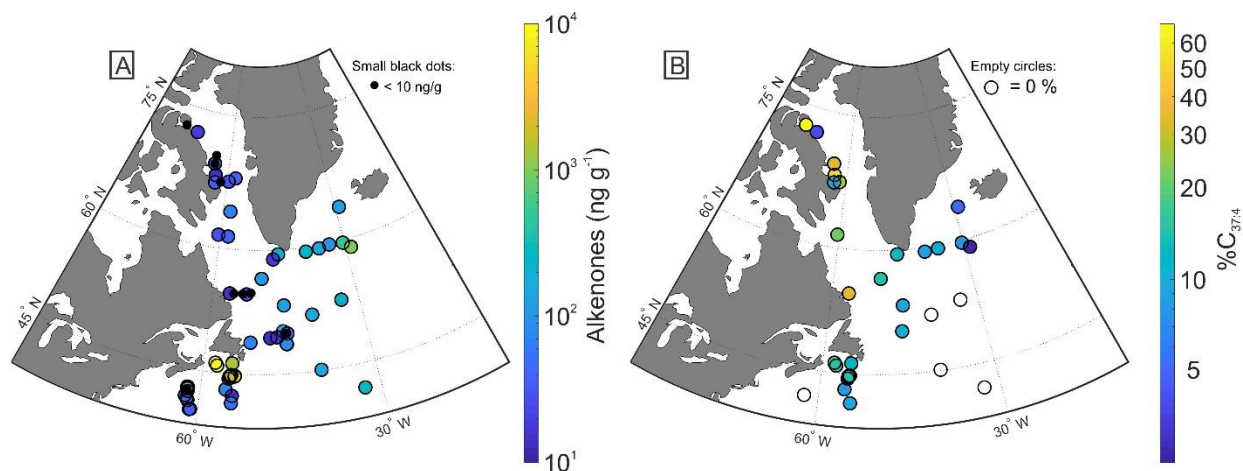


Figure 6. Distribution maps of (A) alkenone concentrations and (B) $\%C_{37:4}$. The small black dots indicate concentrations $< 10 \text{ ng g}^{-1}$ and empty circles indicate zero $\%C_{37:4}$. The samples where $C_{37:4}$ is not quantified/unknown (no $C_{37:4}$ data in Table 6) are not included here.

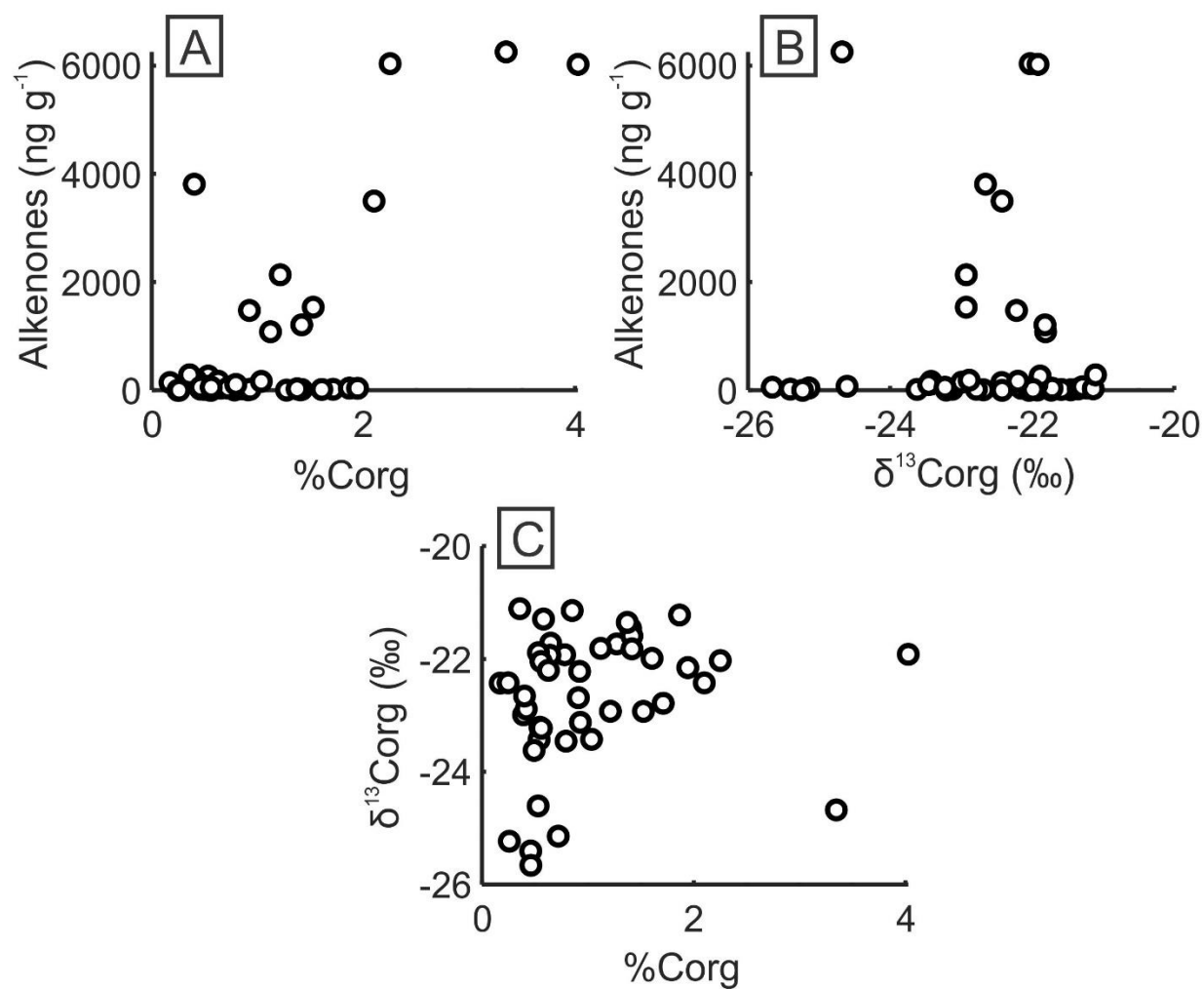


Figure 7. Alkenone concentrations vs. (A) %Corg and (B) $\delta^{13}\text{C}_{\text{Corg}}$, and (C) $\delta^{13}\text{C}_{\text{Corg}}$ vs. %Corg.

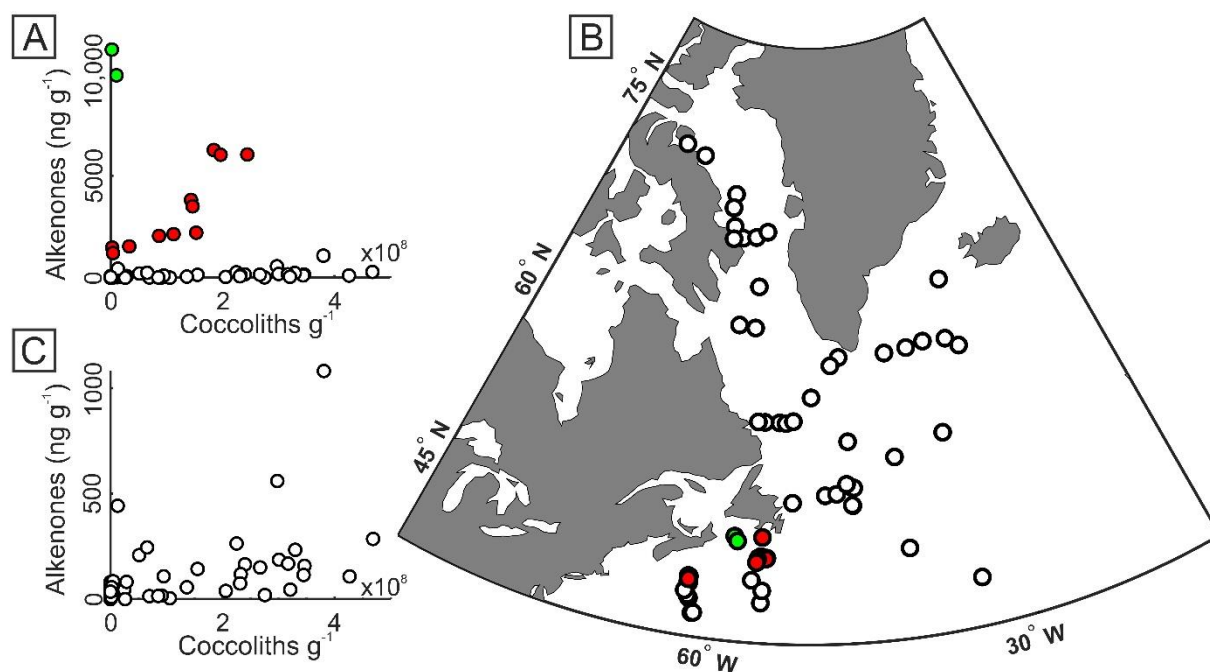


Figure 8. Concentrations of alkenones versus coccoliths. A) Alkenones represented on a scale from 0 to 10,000 ng g⁻¹ with eleven samples (red) showing a strong correlation ($r^2 = 0.77$; $p = 0.00$) and two samples (green) characterized by abundant alkenones but minimal coccoliths are identified on the map (B). C) The alkenone concentrations on a y-scale up to 1000 ng g⁻¹.

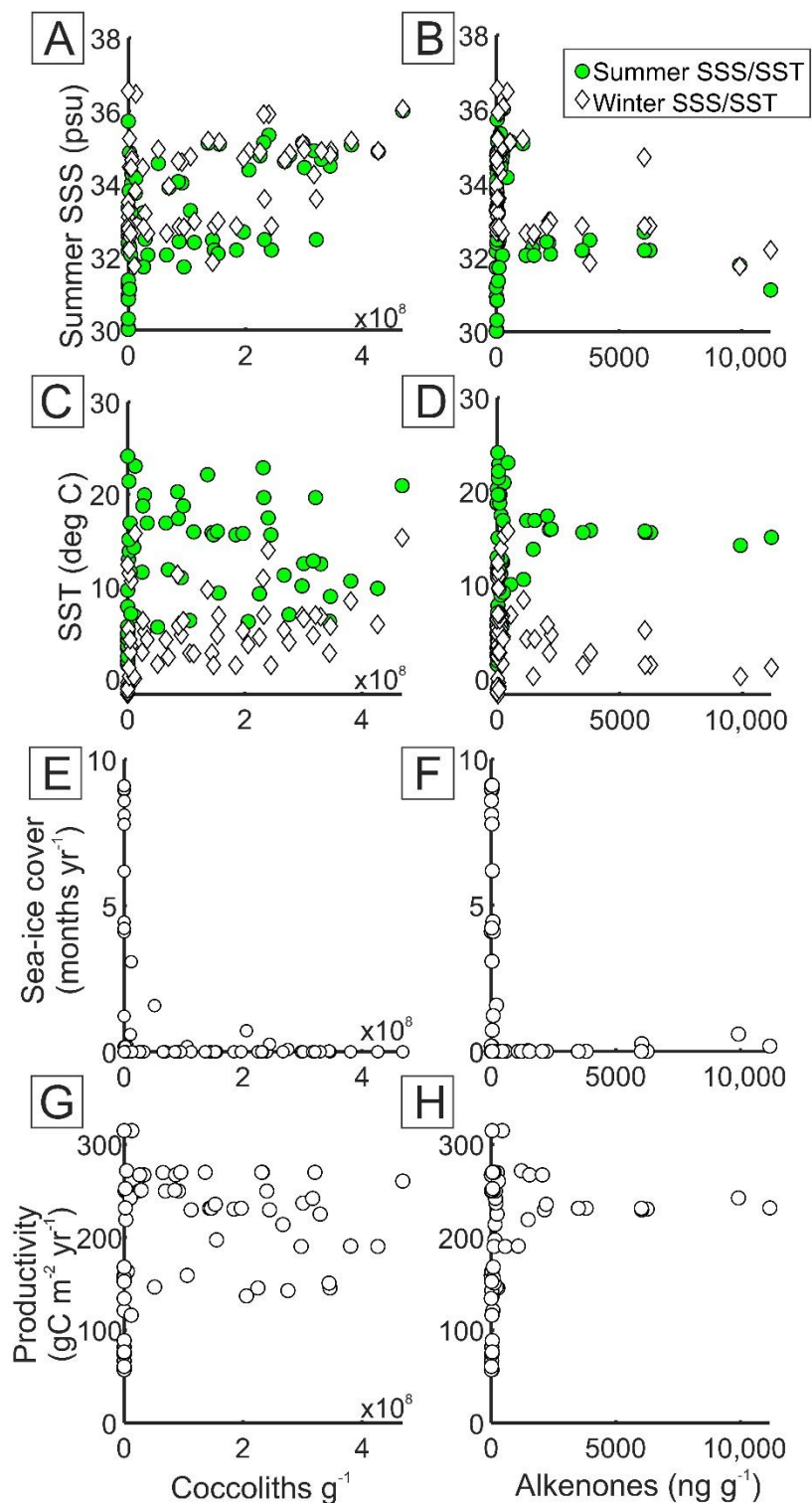


Figure 9. Coccolith and alkenone concentrations versus sea-surface parameters (salinities (SSS; A-B), sea-surface temperatures (SST; C-D), sea-ice cover (E-F), and annual surface productivity (G-H)). Summer SSS and SST are displayed as green circles. Winter SSS and SST are shown as white diamonds (A-D).

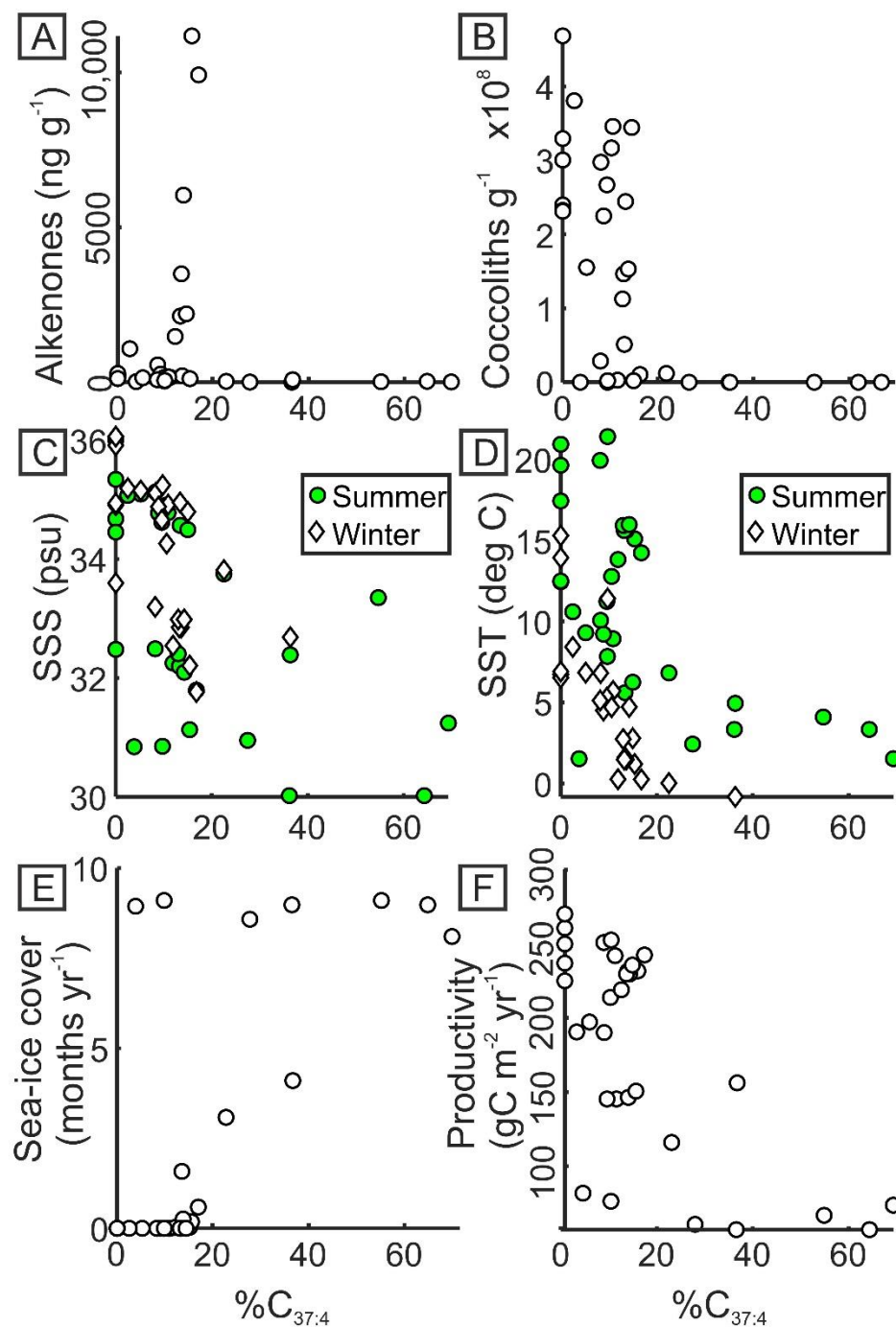


Figure 10. Concentrations of alkenones and coccoliths, sea-surface parameters (salinity (SSS), sea-surface temperatures (SST), sea-ice cover, and productivity) vs. $\%C_{37:4}$ alkenones. SSS and SST are provided for summer (green circles) and winter (white diamonds).

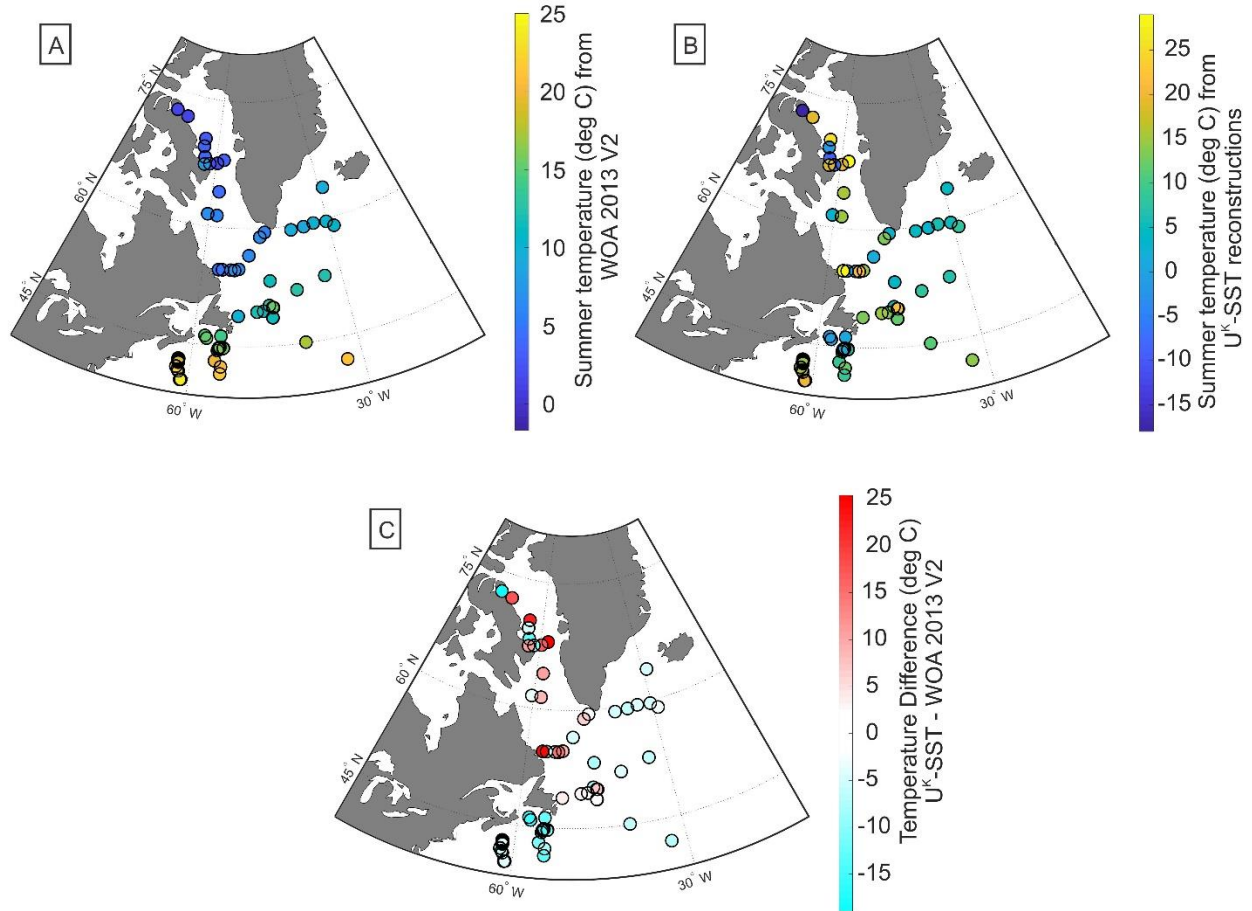


Figure 11. Temperature differences. A) Summer temperatures observed from WOA 2013 V2. B) Reconstructed summer temperatures from U^K_{37} -SST. The temperature difference map (C) shows regions where U^K_{37} -SST reconstructed temperatures are either warmer (red) or colder (light blue) than the WOA 2013 V2. Temperatures with little or no differences are identified by the white circles.

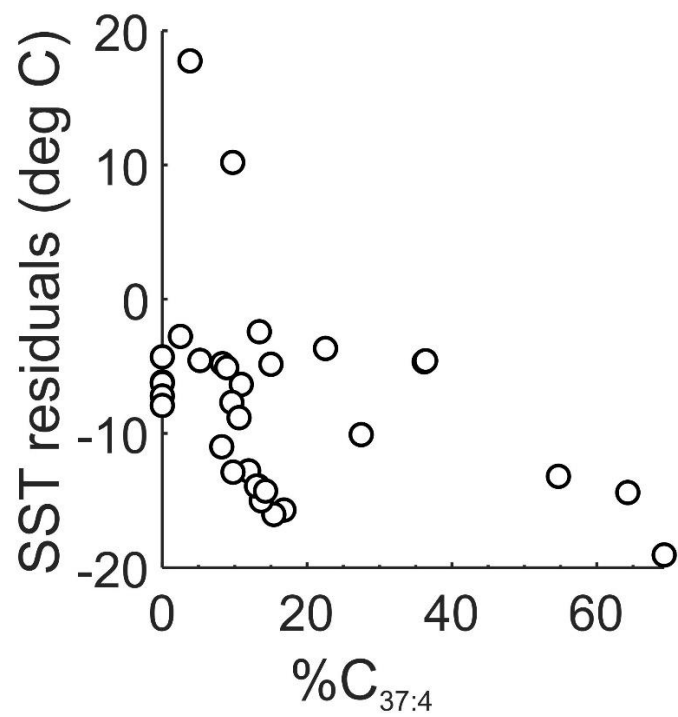


Figure 12. The residuals in sea-surface temperatures (U_{37}^K -SST - WOA 2013 V2) versus %C_{37:4}.

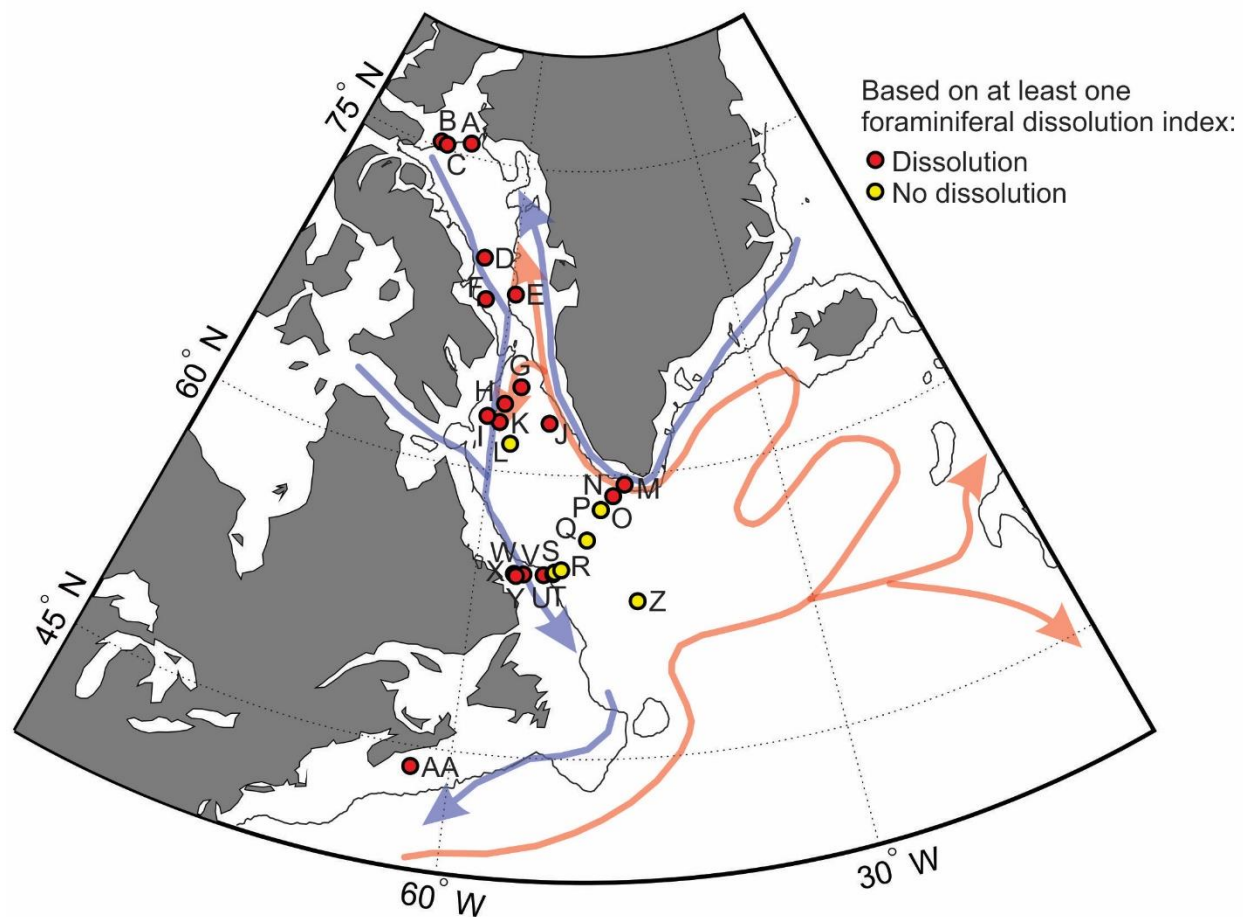


Figure 13. Sites where surface sediments display evidence of calcite dissolution (red) and calcite preservation (yellow) (see Table 7).

Tables

Sample # (Fig. 1)	Surface sediment sample	Latitude (°N)	Longitude (°W)	Water Depth (m)	Sources A = alkenones; C = coccoliths
1+	2013 029 068 BC 0-1 cm	72.82	77.43	1074	this study ^{A,C}
2+	2013 029 063 TWC 0-1 cm	72.45	72.91	833	this study ^{A,C}
3+?	2008 029 014 BC 0-1 cm	70.46	64.66	2060	this study ^{A,C}
4	2013 029 0075 TWC 0-1 cm	69.55	64.64	1484	this study ^{A,C}
5+	2013 029 074 TWC 0-1 cm	69.52	64.64	1482	this study ^{A,C}
6+	2013 029 078 BC 0-1 cm	68.31	63.80	1153	this study ^{A,C}
7+?	2008 029 070 TWC 0-1 cm	68.23	57.62	444	this study ^{A,C}
8+?	2013 029 084 TWC 0-1 cm	67.78	59.58	1359	this study ^{A,C}
9+	2013 029 0048 TWC 0-1 cm	67.61	61.97	935	this study ^{A,C}
10+	2013 029 0051 BC 0-1 cm	67.46	63.59	700.9	this study ^{A,C}
11+?	2008 029 008 TWC 0-1 cm	64.39	58.13	857	this study ^{A,C}
12+	SU90 107 TWC 0-1 cm	63.87	28.94	1625	Marmen (2000) ^A ; this study ^C
13+	2013 029 0018 VVG 0-1 cm	61.54	60.49	1065	this study ^{A,C}
14+?	2008 029 004 TWC 0-1 cm	61.46	58.04	2674	this study ^{A,C}
15+?	91045 060 BC 0-1 cm	59.85	33.58	2255	Marmen (2000) ^A ; this study ^C
16	91045 063 BC 0-1 cm	59.68	30.36	1319	Marmen (2000) ^A ; this study ^C
17	91045 056 BC 0-1 cm	59.63	36.13	3025	Marmen (2000) ^A ; Levesque (1995) ^C
18+	91045 051 BC 0-1 cm	59.49	39.31	2949	Marmen (2000) ^A ; this study ^C
19	90013 006 BC 0-1 cm	59.49	45.87	1105	Marmen (2000) ^A ; this study ^C
20+	91045 071 BC 0-1 cm	58.94	28.74	2237	Marmen (2000) ^A ; this study ^C
21+?	90013 011 BC 0-2 cm	58.91	47.08	2805	this study ^{A,C}
22+	91045 028 BC 0-1 cm	56.62	49.75	3992	Marmen (2000) ^A ; this study ^C
23+?	2005 033B 053 TWC 0-2 cm	54.87	52.01	2357	Filippova (2016) ^A ; this study ^C
24+*	91045 014 BC 0-1 cm	54.74	53.73	340	Marmen (2000) ^A ; this study ^C
25+*	91045 008 BC 0-1 cm	54.72	55.58	301	Marmen (2000) ^A ; this study ^C
26+?	2006 040 0005 TWC 0-2 cm	54.71	52.91	1148	Filippova (2016) ^A ; this study ^C
27	91045 005 BC surface	54.71	56.45	530	Filippova (2016) ^A ; this study ^C
28+?	91045 006 BC 0-2 cm	54.70	56.45	534	this study ^{A,C}
29+	91045 090 BC 0-1 cm	53.33	45.26	3378	Marmen (2000) ^A ; this study ^C
30*	91045 080 TWC 0-1 cm	53.06	33.53	3024	Marmen (2000) ^A ; this study ^C
31+*	SU90 103 TWC 0-1 cm	51.88	39.78	2970	Marmen (2000) ^A ; this study ^C
32+	91045 093 BC 0-1 cm	50.20	45.69	3448	Marmen (2000) ^A ; this study ^C
33?	2009 061 0114 PSH 0-2 cm	49.94	45.05	1404	Filippova et al. (2016) ^A ; this study ^C
34+?	2003 033 029 TWC 0-2 cm	49.91	44.88	3900	Filippova (2016) ^A ; this study ^C
35+?	2004 024 032 TWC 0-2 cm	49.51	46.87	2805	Filippova et al. (2016) ^A ; this study ^C
36?	2004 024 050 TWC 0-2 cm	49.42	48.18	2293	Filippova et al. (2016) ^A ; this study ^C
37?	2010 020 0058 TWC 0-2 cm	48.88	51.86	346	Filippova et al. (2016) ^A ; this study ^C
38?	2009 061 0117 PSH 0-2 cm	48.65	45.17	1018	Filippova et al. (2016) ^A ; this study ^C
39?	2009 061 0119 PSH 0-2 cm	48.61	45.11	933	Filippova et al. (2016) ^A ; this study ^C
40+	2006 039 005 TWC 0-2 cm	46.26	54.98	184	Filippova (2016) ^A ; this study ^C
41	90031 049 BC 0-1 cm	46.19	57.94	468	Marmen (2000) ^A ; this study ^C
42	90031 045 BC 0-1 cm	45.85	57.59	473	Marmen (2000) ^A ; this study ^C
43+*	SU90 102 TWC 0-1 cm	45.09	39.43	1966	Marmen (2000) ^A ; this study ^C
44+?	2002 046 079 TWC 0-2 cm	44.85	55.02	1865	Filippova (2016) ^A ; this study ^C
45+	2006 048 0029 TWC 0-2 cm	44.77	55.08	1861	Filippova (2016) ^A ; this study ^C
46+?	2004 024 006 TWC 0-2 cm	44.76	54.38	585	Filippova (2016) ^A ; this study ^C
47+	2006 048 0020 TWC 0-2 cm	44.75	55.24	2230	Filippova (2016) ^A ; this study ^C
48+?	2002 046 077 TWC 0-2 cm	44.70	54.45	988	Filippova (2016) ^A ; this study ^C
49+	2006 048 0024 TWC 0-2 cm	44.63	55.33	2343	Filippova (2016) ^A ; this study ^C
50	2002 046 074 TWC 0-2 cm	44.39	55.44	2672	Filippova (2016) ^A ; this study ^C
51	96029 069 TWC 0-2 cm	43.07	55.83	3506	Filippova et al. (2016) ^A ; this study ^C
52+?	2000 036 016 TWC 0-2 cm	42.88	62.17	665	Filippova (2016) ^A ; this study ^C
53?	2004 030 003 TWC 1-2 cm	42.83	62.05	896	Filippova et al. (2016) ^A ; this study ^C
54+?	99036 052 TWC 0-2 cm	42.74	62.12	1156	Filippova et al. (2016) ^A ; this study ^C
55?	99036 055 TWC 0-2 cm	42.64	62.12	1517	Filippova et al. (2016) ^A ; this study ^C
56+*	2006 046 0001 TWC 0-2 cm	42.49	62.13	2095	Filippova (2016) ^A ; this study ^C
57+?	2000 036 030 TWC 0-2 cm	42.35	62.03	2414	Filippova et al. (2016) ^A ; this study ^C
58?	98039 010 TWC 0-2 cm	42.34	54.71	4424	Filippova et al. (2016) ^A ; this study ^C
59+*	SU90 U01 BC 0-1 cm	42.03	32.73	1970	Marmen (2000) ^A ; this study ^C
60*	91020 013 TWC 0-2 cm	41.83	62.33	3450	Filippova (2016) ^A ; this study ^C
61?	91020 014 TWC 0-2 cm	41.79	62.34	3532	Filippova et al. (2016) ^A ; this study ^C
62+?	91020 008 TWC 0-2 cm	41.47	62.01	4136	Filippova et al. (2016) ^A ; this study ^C
63+	98039 009 TWC 0-2 cm	41.44	54.78	4649	Filippova et al. (2016) ^A ; this study ^C
64+?	91020 012 TWC 1-2 cm	41.27	61.81	4342	Filippova et al. (2016) ^A ; this study ^C
65?	99036 037 TWC 0-1 cm	40.26	61.19	4791	Filippova et al. (2016) ^A ; this study ^C
66?	99036 035 TWC 0-2 cm	40.22	61.36	4792	Filippova et al. (2016) ^A ; this study ^C

Table 1. List of surface sediment samples used in this study with coordinates, water depth, and references. Sample numbers correspond to the locations on the map (Fig. 1). The samples for which $\delta^{13}\text{C}_{\text{org}}$ data are available are identified by a plus (+) sign. The samples for which $\text{C}_{37:4}$ alkenone data are not detected (nd or 0 ng g⁻¹ in the data; eight samples) are identified by the asterisk (*) and the $\text{C}_{37:4}$ data that were either not determined or unknown are identified by the question mark (?; thirty-one samples).

#	Total coccoliths g ⁻¹ (10 ⁶)	<i>Coccolithus pelagicus</i> g ⁻¹ (10 ⁶)	<i>Emiliania huxleyi</i> g ⁻¹ (10 ⁶)	<i>Calcidiscus leptopus</i> g ⁻¹ (10 ⁶)	<i>Gephyrocapsa spp.</i> g ⁻¹ (10 ⁶)	<i>Umbilicosphaera sibogae</i> g ⁻¹ (10 ⁶)	<i>Helicosphaera carteri</i> g ⁻¹ (10 ⁶)	<i>Pontosphaera discopora</i> g ⁻¹ (10 ⁶)	<i>Rhabdosphaera clavigera</i> g ⁻¹ (10 ⁶)	<i>Syracosphaera pulchra</i> g ⁻¹ (10 ⁶)	<i>Reticulofenestra producta</i> g ⁻¹ (10 ⁶)
1	0	0	0	0	0	0	0	0	0	0	0
2	0	0	0	0	0	0	0	0	0	0	0
3	0	0	0	0	0	0	0	0	0	0	0
4	0	0	0	0	0	0	0	0	0	0	0
5	0	0	0	0	0	0	0	0	0	0	0
6	0	0	0	0	0	0	0	0	0	0	0
7	0	0	0	0	0	0	0	0	0	0	0
8	0	0	0	0	0	0	0	0	0	0	0
9	0	0	0	0	0	0	0	0	0	0	0
10	0	0	0	0	0	0	0	0	0	0	0
11	0	0	0	0	0	0	0	0	0	0	0
12	155	94.2	51.4	4.27	2.05	0.17	0.51	1.71	0.34	0.68	0
13	11.8	11.4	0.39	0	0	0	0	0	0	0	0
14	206	142	57.8	3.16	0.74	1.67	0	0	0	0	0
15	4.26	270	128	8.05	0.42	1.70	2.12	14.8	0	1.70	0
16	298	187	88.5	7.30	3.50	0.29	1.75	6.72	0	1.75	0.29
17	346	149	120	23.0	11.5	0.61	0.61	-	-	-	-
18	2.25	171	44.8	2.84	0.60	0.45	0.90	3.58	0	1.19	0
19	51.1	44.9	4.81	0.26	0	0.39	0	0.26	0	0.52	0
20	381	217	121	17.2	14.1	0.39	1.95	2.34	0	6.25	0
21	275	208	51.4	6.93	4.90	1.69	2.88	0	0	0	0
22	344	267	58.4	8.64	4.03	0.58	1.54	1.92	0	1.54	0.77
23	106	84.4	20.6	0.39	0.39	0.13	0	0	0	0	0
24	0	0	0	0	0	0	0	0	0	0	0
25	0	0	0	0	0	0	0	0	0	0	0
26	5.41	4.64	0.77	0	0	0	0	0	0	0	0
27	0	0	0	0	0	0	0	0	0	0	0
28	0	0	0	0	0	0	0	0	0	0	0
29	267	149	91.9	8.03	1.49	1.49	9.52	3.27	0	1.79	0
30	330	190	98.6	15.4	7.56	1.64	11.2	2.96	0	1.97	0.33
31	300	169	87.9	14.0	7.30	2.63	17.5	1.75	0	0.58	0
32	317	174	104	13.1	3.10	1.38	6.90	13.4	0	1.03	0.34
33	1.58	0.53	0.92	0.13	0	0	0	0	0	0	0
34	6.12	0	3.95	0.25	1.40	0.25	0	0	0	0	0.25
35	91.3	73.0	12.0	3.25	0.52	0.26	0.39	1.95	0	0	0
36	69.3	51.7	14.7	1.93	1.03	0	0	0	0	0	0
37	0	0	0	0	0	0	0	0	0	0	0
38	24.7	22.2	1.71	0.39	0.26	0.13	0	0	0	0	0
39	4.64	4.51	0.13	0	0	0	0	0	0	0	0
40	2.76	0.26	2.37	0.13	0	0	0	0	0	0	0
41	10.4	2.63	3.29	4.34	0.13	0	0	0	0	0	0
42	2.04	0.13	1.28	0.51	0.13	0	0	0	0	0	0
43	240	33.3	143	19.7	18.3	3.75	19.7	0.47	0	1.41	0
44	185	77.2	84.2	12.9	2.68	4.83	2.68	0	0	0	0
45	244	95.0	115	13.5	7.88	5.26	0	3.00	0.37	0.37	4.51
46	144	33.3	94.0	10.3	3.11	0	0	0	0	0.31	2.80
47	147	66.9	59.0	15.7	3.67	0.77	0.39	0	0	0.19	0
48	197	51.7	134	6.13	1.31	0	0	0	0.88	0	3.07
49	113	30.9	66.6	4.38	5.48	3.72	1.10	0.44	0	0	0
50	153	61.8	58.2	21.3	5.02	6.38	0.39	0	0	0	0
51	28.6	16.3	5.78	4.34	1.18	0.26	0.79	0	0	0	0
52	4.12	2.96	0.13	0.39	0	0.51	0	0.13	0	0	0
53	65.3	27.3	12.8	19.8	1.15	2.68	0.89	0.25	0	0.51	0
54	33.2	20.6	4.73	6.57	0	1.31	0	0	0	0	0
55	86.5	34.6	22.2	22.2	2.99	2.73	0.65	0.26	0	0.78	0
56	26.0	18.9	3.55	3.02	0.53	0	0	0	0	0	0
57	94.9	33.1	29.2	19.7	7.75	3.55	1.31	0.13	0	0.13	0
58	85.2	35.8	24.9	16.5	4.12	2.19	1.55	0	0	0.13	0
59	468	13.1	299	33.8	61.0	8.45	44.1	0	0	0.94	7.51
60	232	115	66.6	40.6	7.11	1.11	1.55	0	0	0	0.22
61	321	137	108	54.0	9.23	6.75	4.97	0.35	0	0	0
62	231	33.4	108	25.8	23.0	27.9	11.8	0	0	0.35	0.70
63	2.08	0.78	0.39	0.26	0	0.52	0.13	0	0	0	0
64	136	32.0	66.1	22.3	10.5	1.53	3.07	0.44	0	0	0.22
65	12.9	0.79	2.50	1.31	1.84	6.18	0.26	0	0	0	0
66	0	0	0	0	0	0	0	0	0	0	0

Table 2. Coccolith concentrations in the surface sediment samples.

#	<i>Coccolithus pelagicus</i> (%)	<i>Emiliania huxleyi</i> (%)	<i>Calcidiscus leptoporus</i> (%)	<i>Gephyrocapsa</i> spp. (%)	<i>Umbilicosphaera sibogae</i> (%)	<i>Helicosphaera carteri</i> (%)	<i>Pontosphaera discopora</i> (%)	<i>Rhabdosphaera clavigera</i> (%)	<i>Syracosphaera pulchra</i> (%)	<i>Reticulofenestra producta</i> (%)
1	-	-	-	-	-	-	-	-	-	-
2	-	-	-	-	-	-	-	-	-	-
3	-	-	-	-	-	-	-	-	-	-
4	-	-	-	-	-	-	-	-	-	-
5	-	-	-	-	-	-	-	-	-	-
6	-	-	-	-	-	-	-	-	-	-
7	-	-	-	-	-	-	-	-	-	-
8	-	-	-	-	-	-	-	-	-	-
9	-	-	-	-	-	-	-	-	-	-
10	-	-	-	-	-	-	-	-	-	-
11	-	-	-	-	-	-	-	-	-	-
12	60.66	33.08	2.75	1.32	0.11	0.33	1.10	0.22	0.44	0
13	96.70	3.30	0	0	0	0	0	0	0	0
14	69.17	28.12	1.54	0.36	0.81	0	0	0	0	0
15	63.22	30.02	1.89	0.10	0.40	0.50	3.48	0	0.40	0
16	63.00	29.74	2.45	1.18	0.10	0.59	2.26	0	0.59	0.10
17	43.08	33.45	6.65	15.41	0.18	0.18	0	0	0	0
18	75.83	19.92	1.26	0.27	0.20	0.40	1.59	0	0.53	0
19	87.79	9.41	0.51	0	0.76	0	0.51	0	1.02	0
20	57.13	31.79	4.51	3.69	0.10	0.51	0.62	0	1.64	0
21	75.37	18.67	2.52	1.78	0.61	1.04	0	0	0	0
22	77.54	16.95	2.51	1.17	0.17	0.45	0.56	0	0.45	0.22
23	79.65	19.48	0.37	0.37	0.12	0	0	0	0	0
24	-	-	-	-	-	-	-	-	-	-
25	-	-	-	-	-	-	-	-	-	-
26	85.71	14.29	0	0	0	0	0	0	0	0
27	-	-	-	-	-	-	-	-	-	-
28	-	-	-	-	-	-	-	-	-	-
29	55.96	34.45	3.01	0.56	0.56	3.57	1.23	0	0.67	0
30	57.63	29.91	4.69	2.29	0.50	3.39	0.90	0	0.60	0.10
31	56.17	29.25	4.66	2.43	0.87	5.83	0.58	0	0.19	0
32	54.84	32.75	4.13	0.98	0.44	2.18	4.24	0	0.33	0.11
33	33.33	58.33	8.33	0	0	0	0	0	0	0
34	0.00	64.58	4.17	22.92	4.17	0	0	0	0	4.17
35	79.91	13.11	3.56	0.57	0.28	0.43	2.14	0	0	0
36	74.54	21.19	2.79	1.49	0	0	0	0	0	0
37	-	-	-	-	-	-	-	-	-	-
38	89.89	6.91	1.60	1.06	0.53	0	0	0	0	0
39	97.22	2.78	0	0	0	0	0	0	0	0
40	9.52	85.71	4.76	0	0	0	0	0	0	0
41	25.32	31.65	41.77	1.27	0	0	0	0	0	0
42	6.25	62.50	25.00	6.25	0	0	0	0	0	0
43	13.89	59.69	8.22	7.63	1.57	8.22	0.20	0	0.59	0
44	41.86	45.64	6.98	1.45	2.62	1.45	0	0	0	0
45	38.86	46.85	5.53	3.23	2.15	0	1.23	0.15	0.15	1.84
46	23.16	65.37	7.14	2.16	0	0	0	0	0.22	1.95
47	45.65	40.24	10.69	2.51	0.53	0.26	0	0	0.13	0.00
48	26.28	67.93	3.12	0.67	0	0	0	0.45	0	1.56
49	27.43	59.14	3.89	4.86	3.31	0.97	0.39	0	0	0
50	40.40	38.01	13.89	3.28	4.17	0.25	0	0	0	0
51	56.88	20.18	15.14	4.13	0.92	2.75	0	0	0	0
52	71.88	3.13	9.38	0	12.50	0	3.13	0	0	0
53	41.80	19.53	30.27	1.76	4.10	1.37	0.39	0	0.78	0
54	62.06	14.23	19.76	0	3.95	0	0	0	0	0
55	40.00	25.71	25.71	3.46	3.16	0.75	0.30	0	0.90	0
56	72.73	13.64	11.62	2.02	0	0	0	0	0	0
57	34.90	30.75	20.78	8.17	3.74	1.39	0.14	0	0.14	0
58	42.06	29.20	19.36	4.84	2.57	1.82	0	0	0.15	0
59	2.81	63.93	7.21	13.03	1.80	9.42	0	0	0.20	1.60
60	49.52	28.68	17.50	3.06	0.48	0.67	0	0	0	0.10
61	42.86	33.67	16.83	2.88	2.10	1.55	0.11	0	0	0
62	14.46	46.84	11.14	9.94	12.05	5.12	0	0	0.15	0.30
63	37.50	18.75	12.50	0	25.00	6.25	0	0	0	0
64	23.47	48.55	16.40	7.72	1.13	2.25	0.32	0	0	0.16
65	6.12	19.39	10.20	14.29	47.96	2.04	0	0	0	0
66	-	-	-	-	-	-	-	-	-	-

Table 3. Percentages of coccolith taxa.

#	Summer SSS	Winter SSS	Summer SST (°C)	Summer SST SD (°C)	Winter SST (°C)	Sea-Ice Cover (months yr ⁻¹)	Annual productivity (gC m ⁻² yr ⁻¹)	RDA axis 1	RDA axis 2
1	31.240	-	1.524	1.272	-	8.10	73.76	-	-
2	30.842	-	1.521	1.081	-	8.94	81.85	-	-
3	30.075	-	3.265	1.438	-	8.98	60.47	-	-
4	30.020	-	3.333	0.067	-	8.98	57.31	-	-
5	30.020	-	3.333	0.067	-	8.98	57.31	-	-
6	33.350	-	4.097	0.733	-	9.10	66.95	-	-
7	32.580	33.100	3.564	1.073	-1.68	6.18	88.76	-	-
8	30.313	-	1.928	1.022	-	7.78	74.40	-	-
9	30.950	-	2.427	0.000	-	8.58	60.86	-	-
10	30.850	-	7.837	0.000	-	9.10	76.44	-	-
11	32.777	33.502	4.357	2.373	-0.413	4.44	121.06	-	-
12	35.094	35.151	9.300	0.000	6.840	0.00	197.19	0.3983	0.0017
13	33.750	33.813	6.820	0.000	0.016	3.08	116.04	1.4154	0.2723
14	34.388	34.888	6.200	0.000	3.727	0.72	136.78	0.5867	0.0312
15	34.891	34.907	9.803	0.932	5.885	0.00	190.07	0.5269	0.0572
16	35.128	35.109	10.066	1.095	6.813	0.00	190.07	0.4573	0.0531
17	34.779	34.915	8.930	1.564	5.703	0.00	145.33	-0.1826	0.0314
18	34.775	34.881	9.215	0.769	4.504	0.00	145.33	0.7682	0.1251
19	34.567	34.951	5.600	1.973	1.599	1.58	146.40	1.0791	0.2044
20	35.071	35.191	10.596	0.618	8.425	0.00	190.47	0.2394	0.0082
21	34.785	34.839	6.960	0.640	3.97	0.08	142.59	0.5834	0.1722
22	34.494	34.794	6.245	0.000	2.784	0.03	150.59	0.7004	0.0993
23	33.277	34.748	6.323	1.854	2.784	0.18	158.90	0.8894	0.119
24	32.167	32.869	4.795	1.167	-1.133	0.18	158.90	-	-
25	31.200	33.280	5.670	1.709	-1.428	4.10	134.14	-	-
26	34.025	34.760	7.039	0.979	3.536	0.18	163.37	1.1537	0.1731
27	32.388	32.685	4.944	3.559	-0.83	4.10	156.19	-	-
28	32.388	32.685	4.944	3.559	-0.83	4.22	152.30	-	-
29	34.620	34.658	11.234	1.191	5.280	0.00	213.71	0.2496	0.1589
30	34.677	34.905	12.449	0.947	6.730	0.00	224.93	0.1876	0.1476
31	34.453	34.937	12.492	0.000	6.526	0.00	236.82	0.122	0.2403
32	34.909	34.260	12.78	1.171	4.706	0.00	241.84	0.2314	0.0802
33	33.817	34.463	12.965	2.664	5.394	0.00	249.75	-0.115	-0.4866
34	34.272	34.663	15.028	2.586	10.591	0.00	249.75	-1.6345	0.3893
35	34.034	34.600	10.968	2.627	4.762	0.00	249.75	0.7583	0.0484
36	33.904	33.940	11.836	0.976	2.352	0.00	249.75	0.6773	-0.0732
37	31.372	32.778	9.609	2.653	-1.108	1.22	167.87	-	-
38	33.210	34.448	11.564	2.186	2.885	0.00	249.75	0.9863	0.1638
39	33.210	34.448	11.564	2.186	2.885	0.00	249.75	1.4323	0.2803
40	32.250	32.543	13.824	2.719	0.255	0.02	218.94	-0.6267	-0.3845
41	31.794	31.763	14.236	1.822	0.245	0.59	242.33	-0.5997	-0.9188
42	31.129	32.207	15.095	1.701	1.1930	0.18	231.59	-1.1584	-0.6208
43	35.340	35.913	17.439	0.000	13.933	0.00	249.75	-0.958	0.2521
44	32.196	32.850	15.613	1.984	1.471	0.00	230.41	-0.2003	0.0031
45	32.196	32.850	15.613	1.984	1.471	0.26	229.52	-0.1827	-0.0303
46	32.471	31.860	15.843	1.905	2.780	0.00	231.26	-0.4278	-0.3238
47	32.196	32.850	15.613	1.984	1.471	0.00	231.26	-0.1051	-0.242
48	32.690	34.703	15.759	3.088	5.194	0.00	231.26	-0.2225	-0.2313
49	32.405	32.983	15.930	1.462	2.736	0.00	229.52	-0.4873	0.1768
50	32.095	32.984	15.984	1.185	4.719	0.00	235.55	-0.35	-0.1441
51	32.490	33.195	19.939	2.412	5.113	0.00	250.72	-0.0127	-0.0838
52	32.061	32.653	16.896	2.201	4.246	0.00	271.62	0.5247	0.1722
53	32.061	32.653	16.896	2.201	4.246	0.00	269.67	-0.3878	-0.3103
54	32.061	32.653	16.896	2.201	4.246	0.00	267.18	0.2021	-0.37
55	32.429	32.832	17.374	1.883	5.744	0.00	267.18	-0.419	-0.2896
56	31.732	32.816	18.744	3.194	6.292	0.00	267.18	0.508	-0.3188
57	31.732	32.816	18.744	3.194	6.292	0.00	269.95	-0.5832	-0.0981
58	34.069	34.630	20.274	2.654	11.324	0.00	250.72	-0.3848	-0.1447
59	36.006	36.060	20.938	0.933	15.300	0.00	260.49	-1.4225	0.4177
60	32.480	33.595	19.644	2.769	6.903	0.00	269.95	-0.1174	-0.319
61	32.480	33.595	19.644	2.769	6.903	0.00	269.95	-0.3076	-0.1857
62	35.137	35.910	22.881	2.493	10.889	0.00	269.95	-1.1382	0.4257
63	34.862	35.243	21.414	0.935	11.413	0.00	252.43	-0.4421	0.4393
64	35.120	35.190	22.133	1.655	9.659	0.00	269.95	-0.7597	-0.1329
65	34.160	36.470	23.05	2.245	15.75	0.00	314.78	-1.4522	0.9647
66	35.721	36.550	24.131	1.458	12.378	0.00	314.78	-	-

Table 4. Environmental data from the World Ocean Atlas (WOA), the Snow and Ice Data Center (NSIDC), and the Moderate Resolution Imaging Spectroradiometer (MODIS) (see Section 3) as well as axes 1 and 2 scores from the RDA analyses performed on the taxa and environmental data. Standard deviations are only provided for summer SST.

Summary results	RDA axis 1	RDA axis 2	RDA axis 3	RDA axis 4
Eigenvalues	0.331	0.053	0.039	0.008
Explained variation (cumulative)	33.06	38.37	42.29	43.14
Pseudo-canonical correlation	0.788	0.746	0.510	0.471
Explained fitted variation (cumulative)	75.64	87.78	96.75	98.70
Correlation matrix (r)				
Summer sea-surface salinity	0.086	0.611	-0.244	-0.133
Winter sea-surface salinity	0.038	0.726	-0.105	-0.008
Summer sea-surface temperature	-0.735	-0.041	0.1024	0.017
Winter sea-surface temperature	-0.547	0.504	-0.0253	-0.068
Sea-ice cover	0.355	0.078	0.2123	-0.045
Annual primary productivity	-0.577	-0.096	0.1770	-0.101

Table 5. RDA results for 10 taxa, 6 environmental parameters, and 49 sites.

#	Dist. To coast (km)	Alkenone conc. (ng g ⁻¹)	U ^K ₃₇	U ^K ₃₇ -SST (°C)	%C _{37:4}	%C _{tot} (dw%)	%CaCO ₃	%C _{org}	δ ¹³ C _{org} (‰)
1	6	9.38	-0.53	-17.53	69.31	0.92	0.06	0.9	-22.7
2	62	18.30	0.68	19.28	3.82	1.86	1.26	1.7	-22.8
3	110	8.91	0.89	25.63		1.51	0.86	1.4	-21.5
4	85	28.67	-0.32	-11.07	64.28				
5	85	3.36	0.001	-1.33	36.17	1.35	0.66	1.3	-21.7
6	52	15.20	-0.26	-9.10	54.70	1.49	0.58	1.4	-21.6
7	173	40.39	0.999	28.97		1.85		1.9	-21.2
8	131	26.92	0.67	18.84		1.42	0.37	1.4	-21.4
9	47	5.63	-0.21	-7.66	27.47	0.82	0.34	0.8	-21.9
10	4	37.26	0.64	18.03	9.72	1.90		1.9	-22.2
11	250	65.05	0.54	15.14		0.64	0.48	0.6	-21.3
12	301	143.04	0.20	4.74	5.20	0.45	2.29	0.2	-22.4
13	221	27.93	0.15	3.15	22.54	1.07	1.81	0.8	-21.1
14	350	38.64	0.52	14.36		0.78	1.10	0.6	-21.7
15	513	107.50	0.23	5.54		2.12	12.31	0.6	-21.9
16	612	560.20	0.22	5.26	8.30				
17	395	157.80	0.13	2.58	10.90				
18	225	263.67	0.18	4.12	8.90	0.84	2.59	0.5	-21.9
19	82	208.55	0.15	3.18	13.40				
20	622	1081.95	0.30	7.83	2.50	1.73	5.03	1.1	-21.8
21	167	19.12	0.49	13.53		1.34	3.40	0.9	-23.1
22	480	114.47	0.09	1.40	15.00	0.91	3.06	0.5	-23.4
23	289	4.12	0.53	14.77		0.69	1.23	0.5	-23.2
24	205	0.00	-	-	0.00	0.25	0.07	0.2	-22.4
25	109	0.00	-	-	0.00	0.58	0.18	0.6	-22.0
26	228	21.58	0.75	21.53		0.65	1.55	0.5	-25.4
27	60	80.73	0.06	0.36	36.36				
28	47	15.61	0.999	28.97		1.68	0.65	1.6	-22.0
29	695	150.11	0.16	3.550	9.60	0.92	4.43	0.4	-23.0
30	950	233.60	0.25	6.28	0.00				
31	1005	186.70	0.31	8.19	0.00	1.72	10.86	0.4	-22.9
32	570	168.23	0.17	3.96	10.60	1.00	3.11	0.6	-22.2
33	591	9.89	0.54	15.02					
34	611	42.42	0.77	21.90		0.89	1.39	0.72	-25.1
35	452	16.73	0.48	13.08		1.03	4.46	0.49	-23.6
36	363	13.96	0.53	14.63					
37	86	78.86	0.47	13.04					
38	566	45.10	0.46	12.73					
39	570	84.49	0.43	11.61					
40	74	1476.68	0.08	1.03	11.92	0.92		0.92	-22.2
41	143	9918.99	0.001	-1.47	16.80				
42	170	11179.43	0.01	-0.94	15.40				
43	1058	164.80	0.41	11.19	0.00	2.24	10.07	1.0	-23.4
44	221	6256.22	0.23	5.53		4.80	12.08	3.3	-24.7
45	235	6038.78	0.06	0.60	13.66	3.24	8.22	2.2	-22.0
46	212	3810.28	0.23	5.56		0.57	1.42	0.4	-22.7
47	235	3498.19	0.10	1.70	13.23	2.53	3.59	2.1	-22.4
48	214	6026.16	0.24	6.03		5.01	8.18	4.0	-21.9
49	246	2136.10	0.11	2.01	13.01	1.38	1.39	1.2	-22.9
50	251	2206.65	0.10	1.69	14.26				
51	420	81.81	0.34	8.95	8.21				
52	210	1209.61	0.32	8.32		1.46	0.40	1.4	-21.8
53	217	244.90	0.34	9.04					
54	220	1536.38	0.34	9.04		1.68	1.30	1.5	-22.9
55	229	2050.59	0.32	8.47					
56	248	0.00	-	-	0.00	0.39	1.10	0.3	-25.2
57	261	108.20	0.53	14.74		0.87	0.62	0.8	-23.5
58	477	15.00	0.45	12.40					
59	1671	285.30	0.50	13.73	0.00	2.02	13.84	0.4	-21.1
60	310	118.36	0.43	11.73	0.00				
61	310	44.97	0.54	14.93					
62	350	74.23	0.71	20.16		0.59	0.53	0.5	-24.6
63	581	56.54	0.32	8.53	9.76	0.50	0.35	0.5	-25.7
64	380	56.39	0.57	15.94		0.80	1.95	0.6	-23.2
65	495	442.72	0.59	16.51					
66	490	35.25	0.69	19.72					

Table 6. Distance to coast, alkenones, U^K, and organic and inorganic carbon content data.

Letter (on map)	Samples	Latitude (°N)	Longitude (°W)	Water depth (m)	frag. %	B/P	log(OL/B)	Calcite Dissolution?	Reference
A	76F	75.58	-70.80	560	n.d.	n.d.	3.2	yes	Hamel et al., 2002
B	66F	75.27	-76.08	405	n.d.	n.d.	2.9	yes	Hamel et al., 2002
C	68D	75.22	-74.93	499	n.d.	n.d.	2	yes	Hamel et al., 2002
D	85-027-016 TWC	70.51	-64.52	2091	n.d.	high (infinity)	n.d.	yes	Scott et al., 1989
E	SL-170	68.97	-59.39	1078	n.d. (no PF)	high (infinity)	n.d.	yes	R. Jackson, <i>unpub.</i>
F	SL-174	68.53	-63.33	1559	n.d. (no PF)	high (infinity)	n.d.	yes	R. Jackson, <i>unpub.</i>
G	Core 87-033-007	64.40	-57.42	823	n.d.	n.d.	high (infinity)	yes	de Vernal et al., 1992
H	Core 77-1-2	63.47	-59.10	880	n.d.	high	n.d.	yes	Aksu and Mudie, 1985
I	Core 77-5-1	62.74	-60.90	750	n.d.	high	n.d.	yes	Aksu and Mudie, 1985
J	Core 87-033-008	62.65	-53.88	2424	n.d.	n.d.	high (high OL)	yes	Hillaire-Marcel and de Vernal, 1989;
K	Core 87-033-009	62.52	-59.45	1434	n.d.	No P	n.d.	yes	Hillaire-Marcel et al., 1994a
L	Core 2008-029-004	61.46	-58.04	2674	low (2.79)	low (0.04)	low (-0.936)	no	Gibb et al., 2014;
M	90-013-006-BX 0-1 cm	59.49	-45.87	1105	n.d.	n.d.	1.00	yes	Marshall et al., <i>subm.</i>
N	90-013-011 0-1 cm	58.91	-47.08	2805	n.d.	low (0.03)	0.10	yes	Hillaire-Marcel et al., 1994b
O	ODP 646	58.21	-48.37	3460	low	<< 1	n.d.	no	Hillaire-Marcel et al., 1994a,b
P	91-045-017-BX 0-1 cm	58.21	-48.36	3379	n.d.	n.d.	0.00	no	Aksu et al., 1989;
Q	90-013-013-P	58.21	-48.36	3379	n.d.	n.d.	low (-0.65)	no	Hillaire-Marcel and de Vernal, 1989
R	91-045-028-BX 0-1 cm	56.62	-49.75	3992	n.d.	n.d.	-0.70	no	Hillaire-Marcel et al., 1994b
S	91-045-018-BX 0-1 cm	55.03	-52.13	2648	n.d.	n.d.	0.00	no	Hillaire-Marcel et al., 1994b
T	91-045-023-BX 0-1 cm	54.90	-52.74	1984	n.d.	n.d.	0.72	yes	Hillaire-Marcel et al., 1994b
U	91-045-016-BX 0-1 cm	54.82	-52.87	1364	n.d.	n.d.	0.64	yes	Hillaire-Marcel et al., 1994b
V	91-045-014-BX 0-1 cm	54.74	-53.73	340	n.d.	n.d.	1.30	yes	Hillaire-Marcel et al., 1994b
W	91-045-008-BX 0-1 cm	54.72	-55.58	301	n.d.	n.d.	0.70	yes	Hillaire-Marcel et al., 1994b
X	91-045-005-BX 0-1 cm	54.71	-56.45	530	n.d.	n.d.	1.00	yes	Hillaire-Marcel et al., 1994b
Y	73-027-12	54.60	-56.26	540	n.d.	high (4)	n.d.	yes	Scott et al., 1984
Z	ODP 647	53.33	-45.26	3870	low	<< 1	n.d.	no	Aksu et al., 1989
AA	77-002-20	43.89	-62.88	271	n.d.	high (2.5)	n.d.	yes	Scott et al., 1984

Table 7. Core-top sediment samples with at least one foraminiferal dissolution index applied to indicate ‘yes’ or ‘no’ to calcite dissolution (Fig. 13).

Appendices

Appendix A: Coccolith Taxonomy

Division HAPTOPHYTA
Class PRYMNESIOPHYCEAE
Order COCCOLITHALES
Family CALCIDISCACEAE
Genre <i>Calcidiscus</i>
<i>Calcidiscus leptoporus</i>
Genre <i>Umbilicosphaera</i>
<i>Umbilicosphaera sibogae</i>
Family COCCOLITHACEAE
Genre <i>Coccolithus</i>
<i>Coccolithus pelagicus</i>
Order ISOCHRYSIDALES
Family NOELAERHABDACEAE
Genre <i>Emiliana</i>
<i>Emiliana huxleyi</i>
Genre <i>Gephyrocapsa</i>
<i>Gephyrocapsa muelleriae</i>
<i>Gephyrocapsa oceanica</i>
Genre <i>Reticulofenestra</i>
<i>Reticulofenestra producta</i>
Order SYRACOSPHERALES
Family RHABDOSPHAERACEAE
Genre <i>Rhabdosphaera</i>
<i>Rhabdosphaera clavigera</i>
Family SYRACOSPHAERACEAE
Genre <i>Syracosphaera</i>
<i>Syracosphaera pulchra</i>
Order ZYGODISCALES
Family HELICOSPHAERACEAE
Genre <i>Helicosphaera</i>
<i>Helicosphaera carteri</i>
Family PONTOSPHAERACEAE
Genre <i>Pontosphaera</i>
<i>Pontosphaera discopora</i>

Table A1. Coccolith taxonomy of all species identified in our study. See the Nannotax3 database for more details and species references (Young et al., 2017).

Appendix B: Scanning electron micrographs of core-top sediments

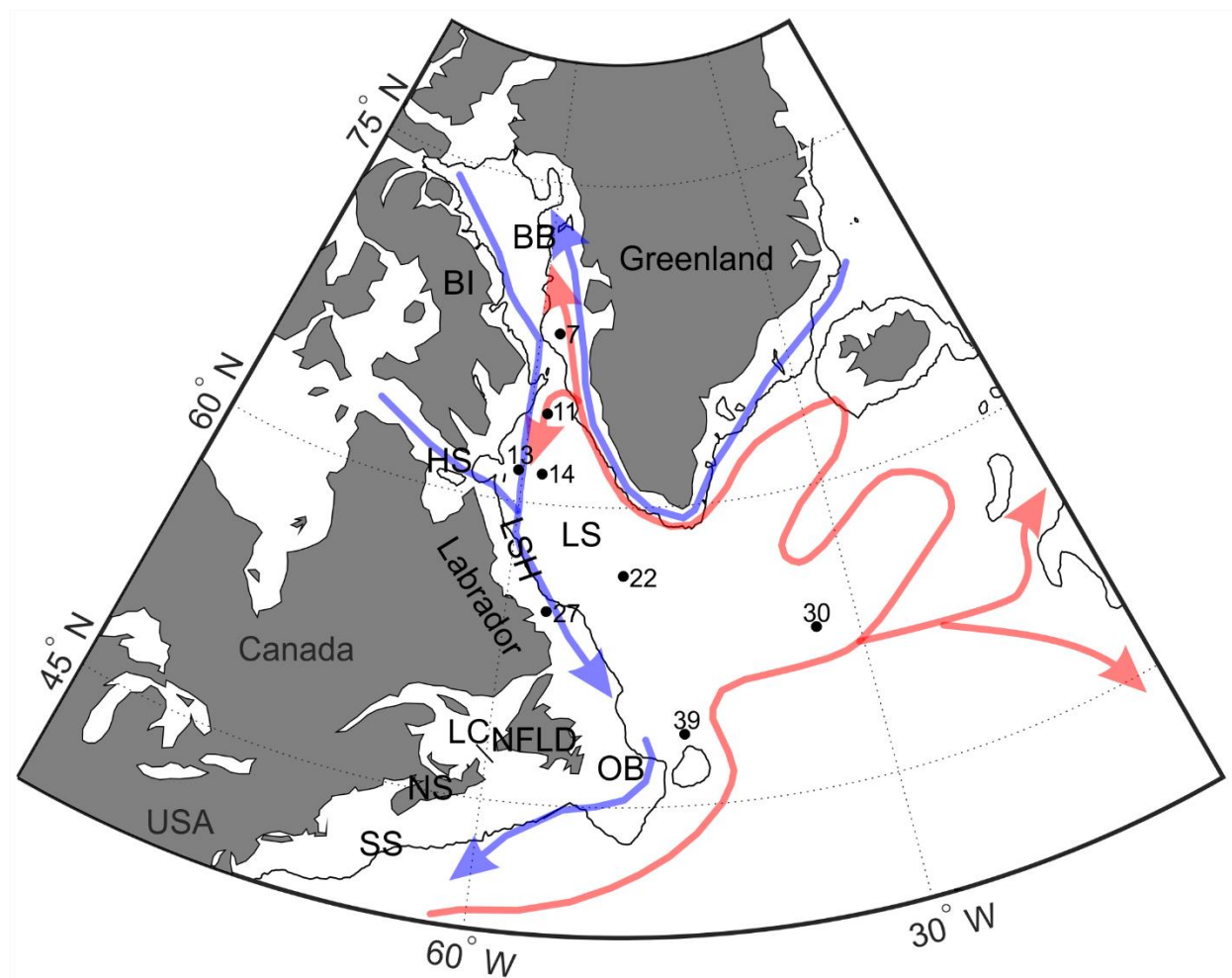


Figure B1. The core locations of the samples in the scanning electron micrographs.

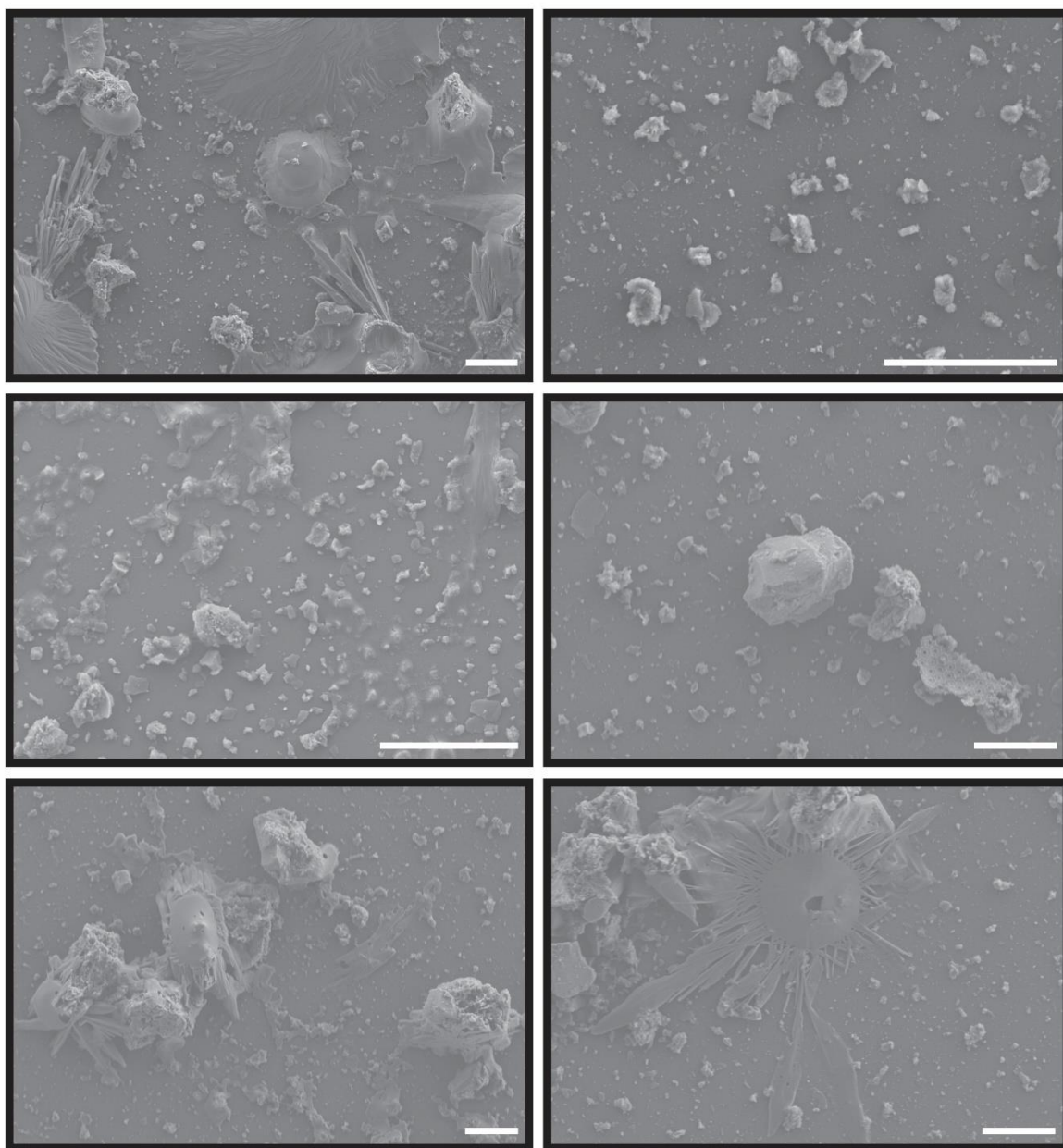


Figure B2. 2008 029 070 TWC 0-1 cm (#7 on map; Baffin Bay). Scale bars are all 20 μm . This sample is barren of coccoliths.

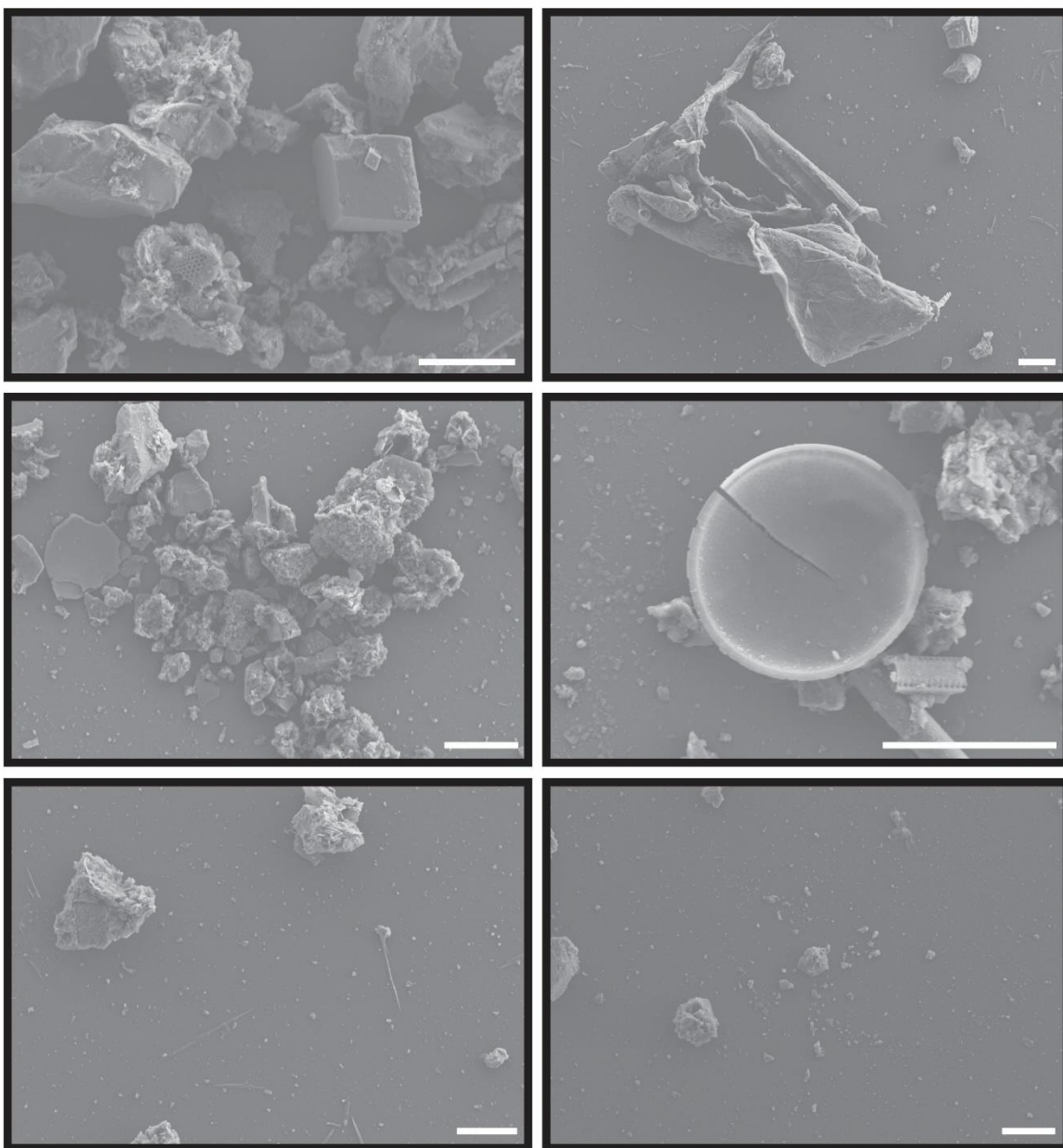


Figure B3. 2008 029 008 TWC 0-1 cm (#11 on map; Davis Strait). Scale bars are all 20 μm . This sample is barren of coccoliths.

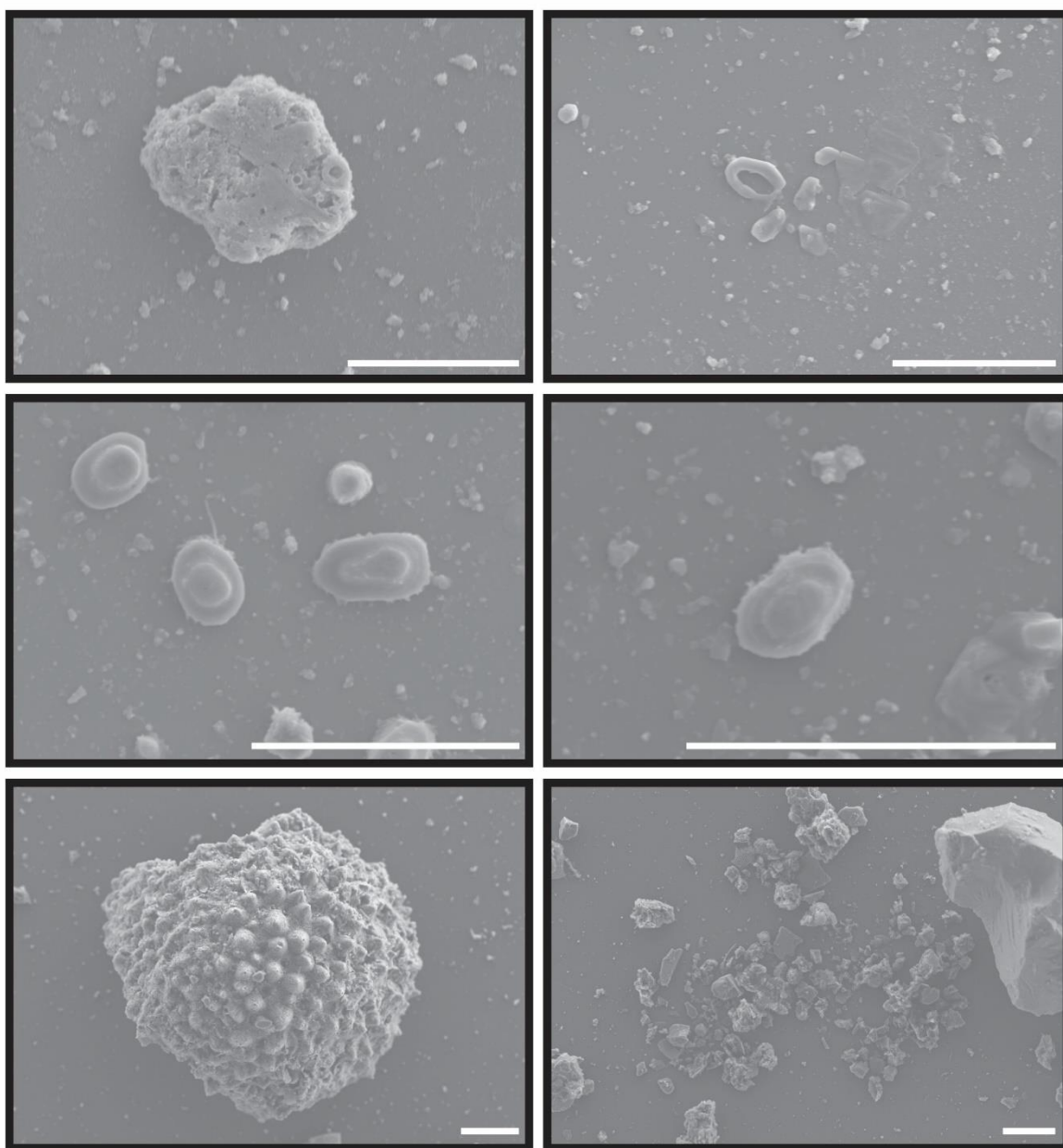


Figure B4. 2013 029 0018 VVG 0-1 cm (#13 on map; northwest Labrador Sea). Scale bars are all 20 μm . This sample contains coccoliths.

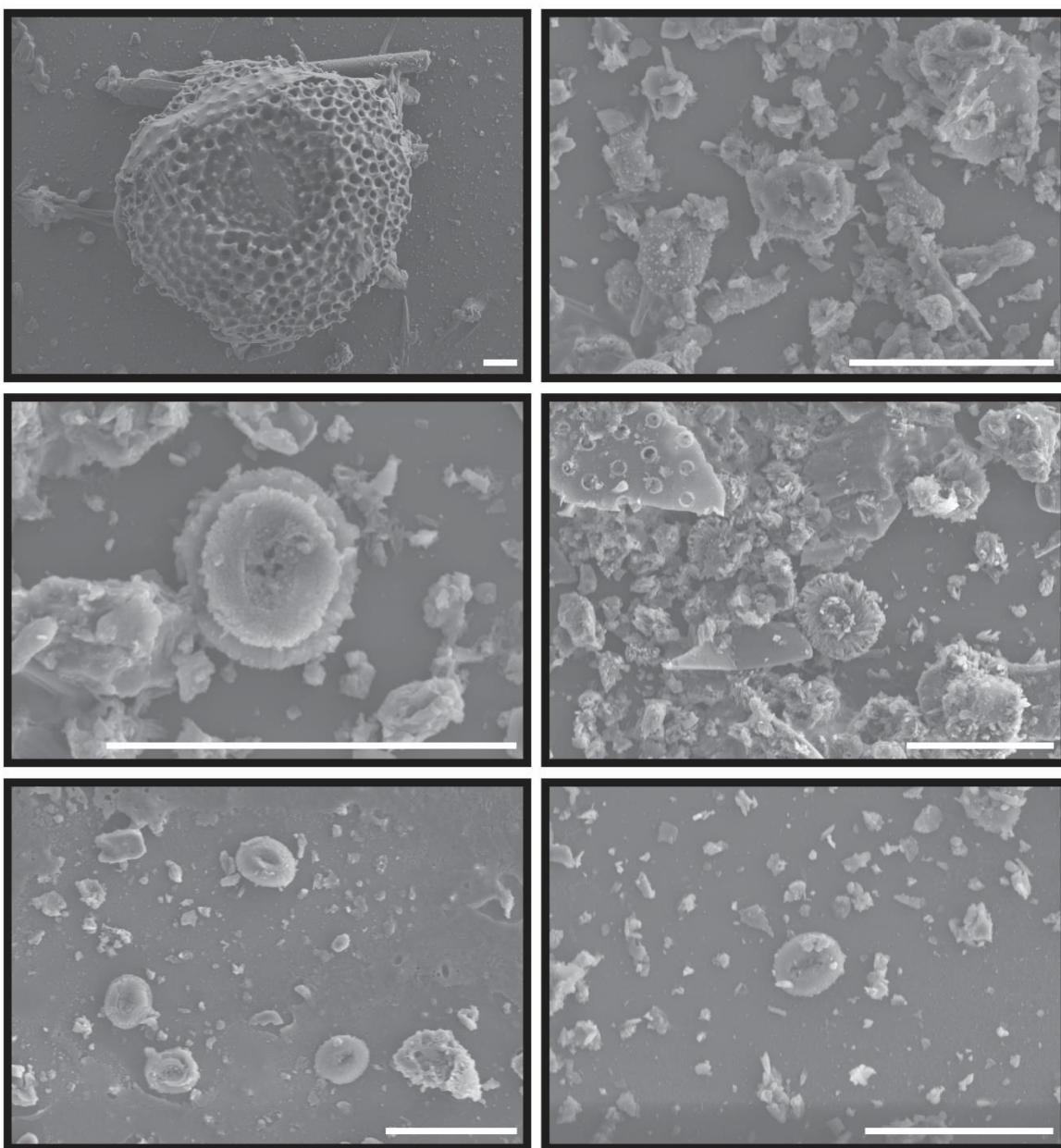


Figure B5. 2008 029 004 TWC 0-1 cm (#14 on map; northwest Labrador Sea). Scale bars are all 20 μm . This sample contains coccoliths.

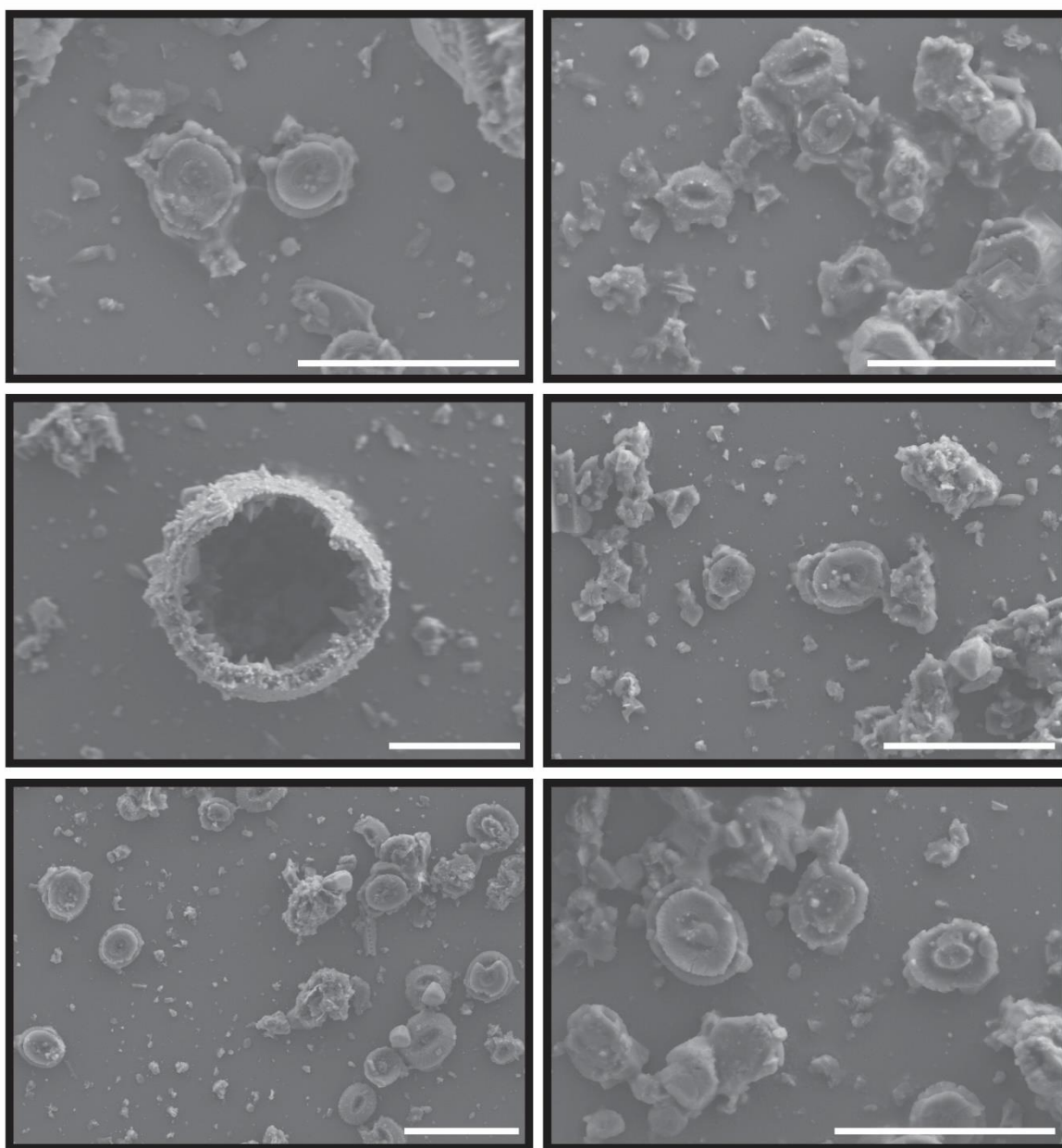


Figure B6. 91045 028 BC 0-1 cm (#22 on map; central Labrador Sea). Scale bars are all 20 μm . This sample contains abundant coccoliths.

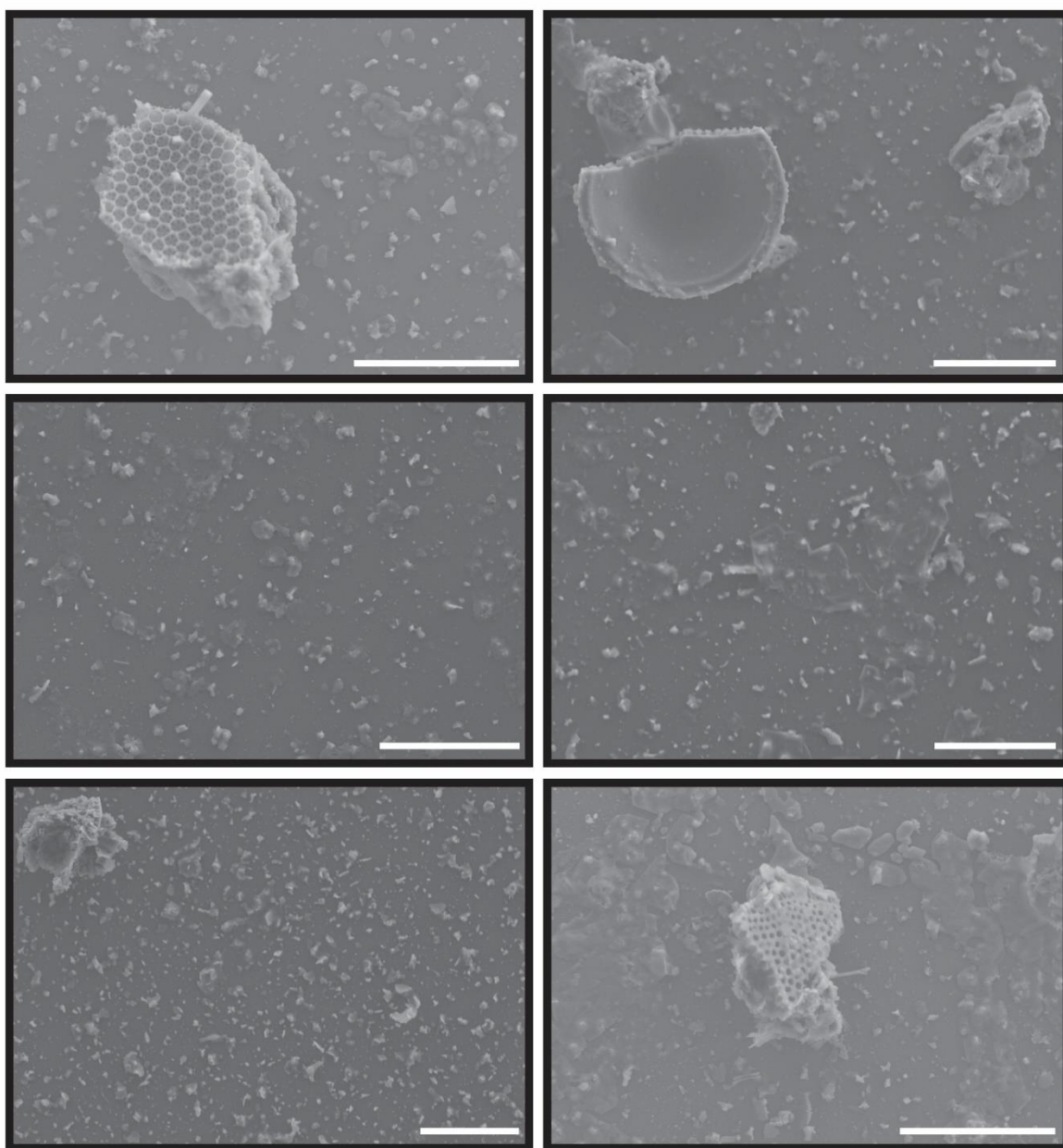


Figure B7. 91045 005 BC surface (#27 on map; Labrador Shelf). Scale bars are all 20 μm . This sample is barren of coccoliths.

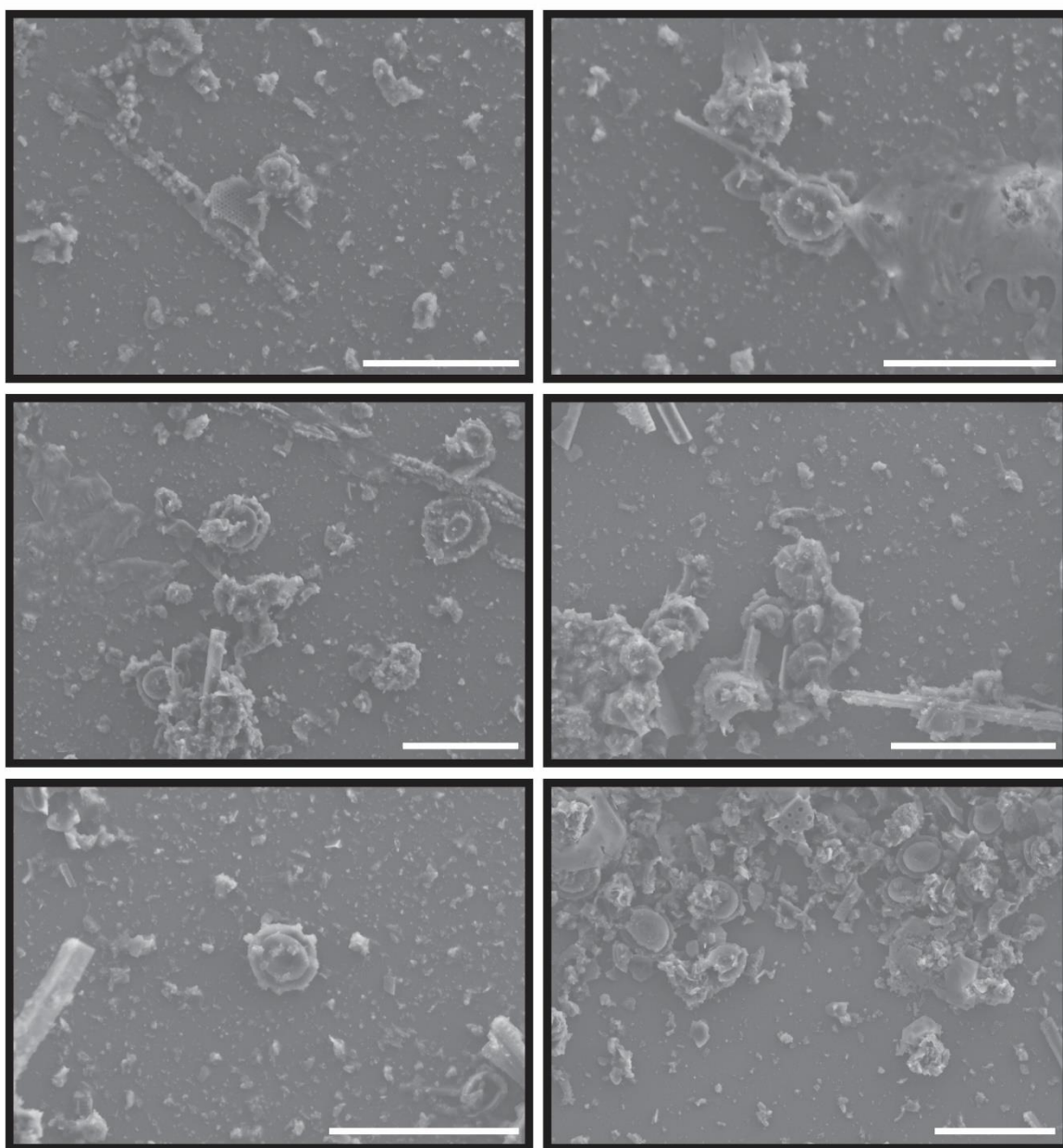


Figure B8. 91045 080 TWC 0-1 cm (#30 on map; central North Atlantic). Scale bars are all 20 μm . This sample contains abundant coccoliths.

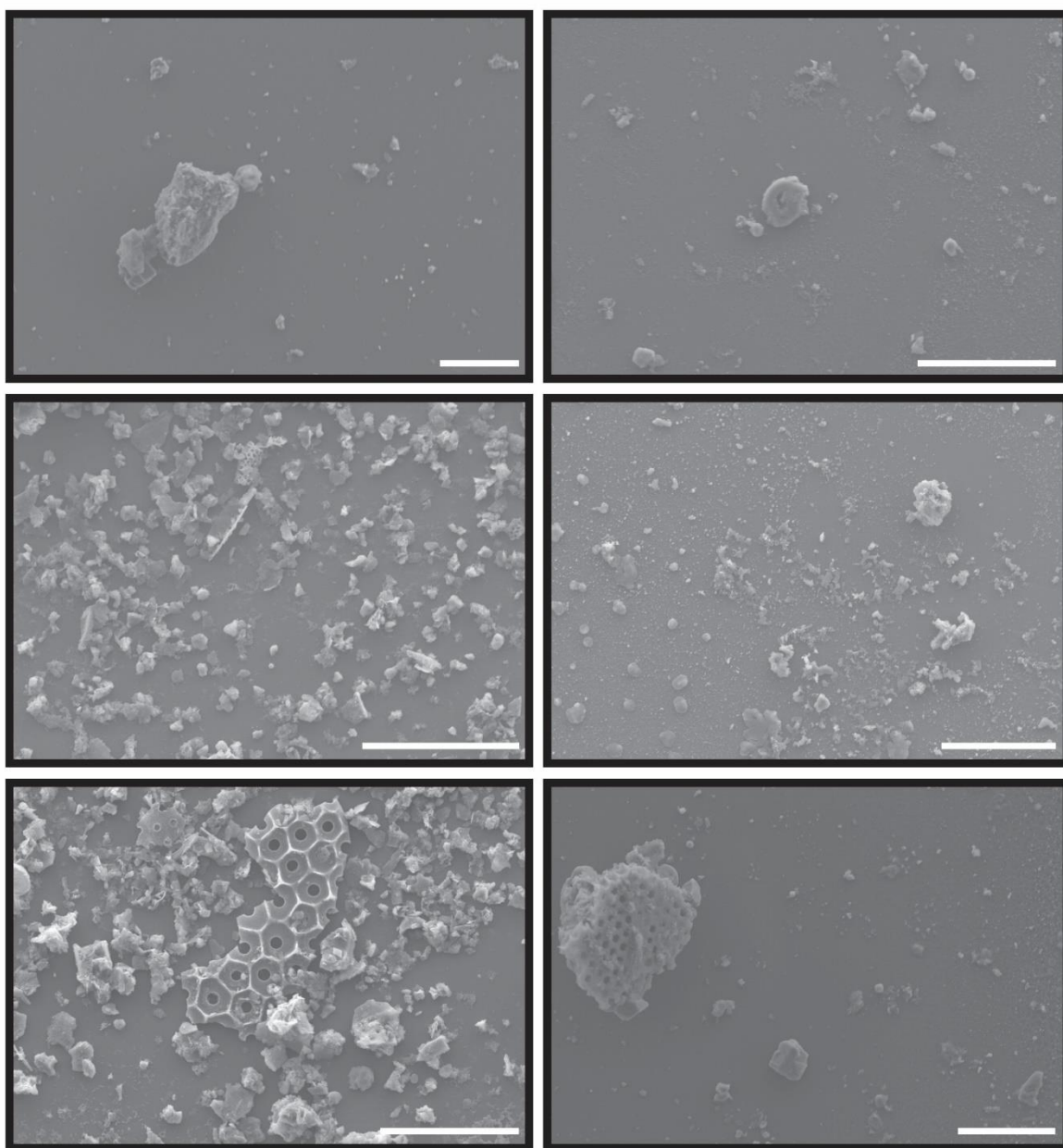


Figure B9. 2009 061 0119 PSH 0-2 cm (#39 on map; southeast Labrador Sea). Scale bars are all 20 μm . This sample contains coccoliths.

Appendix C: Oceanographic parameters

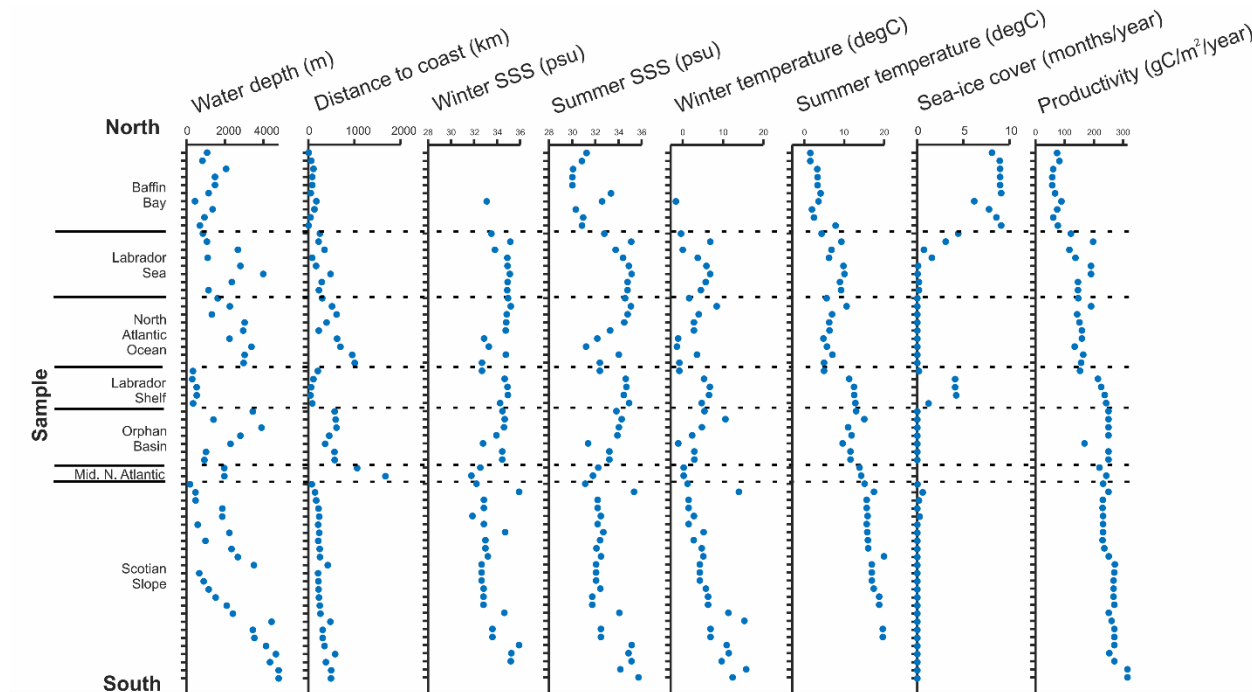


Figure C1. Oceanographic parameters including water depth, distance to coast, seasonal sea-surface salinities and temperatures, sea-ice cover, and annual productivity are shown here on a north-south diagram. The Scotian Slope region has warmer summer temperatures and a more uniform salinity range compared to that of the Labrador Sea. Overall, annual productivity increases with decreasing latitude.

CHAPTER TWO

Biogenic carbonate fluxes and preservation in the northwest Labrador Sea since the Last Glacial Maximum

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Abstract

Coccoliths, alkenones, and foraminifers were analyzed in core HU2008-029-004 PC (61.46 °N and 58.04 °W; water depth = 2674 m) from the northwest Labrador Sea to document linkages between hydrographic conditions, biogenic carbonate fluxes to the seafloor, and their preservation/dissolution during the last 25,000 years. Large changes in coccolith and foraminifer concentrations are recorded, with sediments from the last ice age containing significantly less carbonate microfossils ($9.5 \pm 3.9 \times 10^5$ coccoliths g^{-1} and 2860 ± 580 planktonic foraminifers g^{-1}) than sediments from the deglacial and postglacial intervals (up to 3.1×10^8 coccoliths g^{-1} and 2.9×10^4 foraminifers g^{-1}). Three foraminifer-based calcite dissolution indices were used to evaluate biogenic carbonate preservation: the planktonic foraminifer fragmentation index, the ratio of benthic-to-planktonic foraminifers (B/P), and the ratio of organic linings to benthic foraminifers (OL/B). Fragmentation remained low throughout the postglacial (mean of 4%) but reached up to 8 % in the deglacial and peaked at 16 % in samples from the Bølling-Allerød of the late glacial interval. Samples from the Bølling-Allerød and the deglacial interval also depict a slightly elevated B/P index (> 0.15), in contrast with the near zero values characterizing most of the sequence. The OL/B index is close to zero throughout the whole core, suggesting particularly good biogenic carbonate preservation, which implies that the low biogenic carbonate and coccolith content in sediments of the glacial stage are primarily related to the low productivity of calcifying organisms. The Bølling-Allerød recorded elevated fragmentation that may stem from mechanical abrasion from ice-rafted debris, while the elevated fragmentation during the deglacial interval, a time of elevated productivity and low percentages of ice-rafted debris, suggests possible oxic remineralization in sediment porewaters causing calcite undersaturation in the sediments. We also identify a large decoupling of coccolith and alkenone concentrations throughout the core. Colder-than-expected $U^{K_{37}}$ -SST estimates from the alkenones of the glacial interval rule out possible allochthonous inputs from lower-latitude locations and we suggest alkenone production by non-calcifying haptophytes, at least during the glacial interval.

Highlights

- Non-calcifying haptophytes most likely produced the alkenones in the glacial interval.
- Mechanical abrasion can explain the elevated fragmentation during the Bølling-Allerød.
- Porewater calcite dissolution may have occurred during the deglacial interval.

Keywords

Coccoliths, alkenones, foraminifers, dissolution, preservation, productivity, Labrador Sea

1. Introduction

Biogenic carbonate shells make up most of the calcite that accumulates on the seafloor (Dutkiewicz et al., 2015). Coccolithophores and foraminifers produce calcite shells, which upon death or grazing, may dissolve in the water column, at the seafloor, or during diagenesis (Jansen et al., 2002). Calcite shell preservation in marine sediments depends on the calcite saturation state of the overlying waters and sediment porewaters (Morse and Mackenzie, 1990; Mucci et al., 2000). The calcite saturation state typically decreases with depth because calcite solubility increases with pressure (Pytkowicz and Connors, 1964) while pH decreases in response to the accumulation of metabolic carbon dioxide resulting from the microbial decay of organic matter settling through the water column (Emerson and Bender, 1981).

In the Labrador Sea, exceptionally large changes in ocean conditions occurred from the last ice age to present, leading to increases in productivity (Hillaire-Marcel et al., 1994a, b; Rahman and de Vernal, 1994; Rahman, 1995; Gibb et al., 2014). In sediments of glacial age, low biogenic carbonate (e.g., foraminifers and coccoliths) content has been interpreted to result from low productivity (piston cores 013, 094, and 009; Fig. 1; Hillaire-Marcel et al., 1994a, b; Rahman and de Vernal, 1994; Rahman, 1995). At the scale of the subpolar North Atlantic, some studies also suggest low biogenic carbonate productivity during glacial times based on planktonic and benthic foraminifers (Thomas et al., 1995; Rasmussen et al., 2002; Wollenburg et al., 2004). Some studies, however, also show that calcium carbonate preservation affects the foraminiferal content of sediments from Baffin Bay (Aksu, 1983; de Vernal et al., 1992; Hamel et al., 2002) and the Nordic Seas (Zamelczyk et al., 2012, 2013).

Here, we have analyzed coccoliths, alkenones, and foraminifers in piston core HU2008-029-004 (hereafter 004) collected in the northwest Labrador Sea (Fig. 1), in order to document

their concentrations and fluxes in the sediments and to examine the linkages with hydrographic conditions, productivity, and the potential effect of preservation/dissolution. Core 004 encompasses the past ~ 36,000 years. It was previously studied by Gibb et al. (2014, 2015) who provided records of stable isotopes ($\delta^{18}\text{O}$ -Npl and $\delta^{13}\text{C}$ -Npl) in planktonic foraminifers (*Neoglobobulimina pachyderma* left-coiled; Npl) as well as palynological data, and by Nuttin et al. (2015) who measured U, Th, and Pa isotopes along with sedimentological parameters such as detrital materials (e.g., carbonate). Our study expands from these prior analyses, allowing us to not only document biogenic carbonate production but also calcite preservation while providing insights into the regional variations of the water column calcite saturation state and ocean circulation. Biogenic carbonate production was evaluated from counts of foraminifer shells and coccoliths, while variations in calcite preservation were evaluated from three different foraminiferal indices including the fragmentation of foraminiferal shells (Berger, 1968; Thunell, 1976), the benthic-to-planktonic foraminiferal shell (B/P) ratio (Aksu et al., 1989), and the proportion of organic linings versus carbonate shells of benthic foraminifers (de Vernal et al., 1992; Hamel et al., 2002). Coccolithophore productivity was assessed using not only coccolith counts but also alkenones (biomarkers of specific coccolithophore species - *Emiliania huxleyi* and *Gephyrocapsa oceanica*; Volkman et al., 1980a, b; Marlowe et al., 1984a, b, 1990), which are two variables not commonly investigated together. In this study, we questioned whether calcite dissolution may have occurred in the northwest Labrador Sea during the last glacial episode, which would explain the low calcite shell abundances in the sediments from that time. Thus, untangling productivity and preservation of biogenic carbonate at our core site was a major goal of our study. Here, we employ multiple proxy tools to assess whether the northwest Labrador Sea acts more like the open northwest North Atlantic (production dominated) or more

like Baffin Bay (dissolution dominated), and how this changes through time as the Labrador Sea emerged from a glacial state, most similar to Baffin Bay, to a key sub-basin component of the northwest North Atlantic.

2. Oceanographic setting

The Labrador Sea is located between southwest Greenland and the coastline of Newfoundland and Labrador (Fig. 1). Along the Greenland coast, the West Greenland Current is formed from the mixing of the warm and saline Irminger Current, a side branch of the North Atlantic Current, with the East Greenland Current that flows northwards into eastern Baffin Bay (Fig. 1) (Buch, 1981; Buch et al., 2004; Tang et al., 2004). In western Baffin Bay, the Baffin Island Current flows southwards through western Davis Strait and onto the Labrador Shelf, where it mixes with water from Hudson Strait and a branch of the West Greenland Current to become the Labrador Current (Myers et al., 1990; Percy and Fife, 1993; Lazier and Wright, 1993). As seen in modeling studies (Wu and Tang, 2011; Wu et al., 2012), the subsurface and bottom waters in the northwest Labrador Sea flowed in from offshore west Greenland. In the central Labrador Sea, the Labrador Sea Water (LSW) forms by vertical convection following strong surface cooling during severe winters and spreads throughout the deep North Atlantic, contributing to the North Atlantic Deep Water (NADW) as a part of the Atlantic Meridional Overturning Circulation (AMOC) (Clarke and Gascard, 1983; Yashayaev, 2007; Jones and Anderson, 2008; Yashayaev and Loder, 2009). Today, the formation of well-ventilated NADW makes the Atlantic Ocean less corrosive to calcite than the rest of the global ocean (Archer, 1991). A weakening in the strength of NADW formation during the glacial is thought to be a

major reason for decreasing carbonate preservation in the Atlantic Ocean at that time (Crowley, 1983; Mix and Fairbanks, 1985; Diester-Haas, 1985; Henrich, 1998).

The modern calcite saturation state varies throughout the North Atlantic. Arctic water has a low saturation state as it enters the North Atlantic via outflow along the western Baffin Bay and Davis Strait (Azetsu-Scott et al., 2010). In Baffin Bay, the calcite saturation horizon ($\Omega_{\text{cal}} = 1$; the depth below which calcite begins to dissolve) is found at depths of 1,000 m (west) to 1,500 m (east), reflecting the cyclonic circulation in Baffin Bay with the addition of Arctic outflow with low saturation states in the west (Azetsu-Scott et al., 2010) and the influence of the relatively warm and salty Atlantic waters mixed into the West Greenland Current that flows northwards in the east (Yashayaev, 2007). In the Labrador Sea, the position of the saturation horizon is controlled by the depths of local convection, ventilation age, and advective time scales of the intermediate and deep-water masses of the North Atlantic. In the Labrador Sea, there is no calcite saturation horizon since Ω_{cal} is ~ 1.2 near the seafloor (Azetsu-Scott et al., 2010). The Ω_{cal} is ~ 2.2 over the Labrador Shelf and the Ω_{cal} is ~ 2.8 over the Greenland Shelf (Azetsu-Scott et al., 2010).

3. Materials and Methods

Core HU2008-029-004 PC (hereafter core 004; 61.46 °N and 58.04 °W; 2674 m of water depth; 8.96 m long; Fig. 1) was collected in 2008 during a cruise of the *CCGS Hudson* (Campbell and de Vernal, 2009). The core chronology (Fig. 2; Table S1) was developed with thirteen ^{14}C dates using the Bacon software (Blaauw and Christen, 2011) with a marine reservoir correction of 400 years and no additional correction (ΔR) for the regional air-sea reservoir difference (see supplementary material of Hillaire-Marcel et al., 2007). Data from core 004 will

only be reported for the past 25,000 years, even though core 004 extends backs ~ 36,000 years (Gibb et al., 2014).

We also adopted the stratigraphic nomenclature of Gibb et al. (2014), defined from the micropaleontological, geochemical, and isotopic signatures of the core. The “glacial” includes 25 ka until 12.2 ka; the “deglacial” spans from 12.2 to 8.3 ka, and the “postglacial” corresponds to the last 8300 years. In addition, we refer in the text and figures to Heinrich events 1 and 2 (H1 and H2), which are dated respectively at ~ 15–17.6 ka and ~ 22–25 ka (Bond et al., 1993, 1999; Rashid et al., 2003; Andrews et al., 2012; Marshall et al., 2014; Gibb et al., 2014; Hesse and Khodabakhsh, 2017), and to the warm stadial of the Bølling-Allerød of 14.75–12.7 ka (Thiagarajan et al., 2014; Dendievel et al., 2019).

Concentrations and fluxes of coccoliths, alkenones, and benthic and planktonic foraminifers were determined in fifty-two sediment samples from core 004 taken at 10–20 cm intervals throughout the glacial unit and 4–10 cm intervals throughout the deglacial and postglacial units. The samples analyzed in this study were aliquots of the samples used by Gibb et al. (2014) and the alkenones were from Filippova (2016).

Mean concentrations and fluxes of coccoliths, alkenones, and foraminifers for the glacial, deglacial, and postglacial intervals excluded barren samples and were reported as mean $\pm$ one standard error. We compared our data with records of organic linings of foraminifers, subsurface-dwelling Npl- $\delta^{13}\text{C}$ and Npl- $\delta^{18}\text{O}$, as well as sea-ice cover, annual productivity, and summer sea-surface temperatures reconstructed from dinocysts, as published by Gibb et al. (2014, 2015).

3.1 Coccoliths

Coccoliths were counted on microscope slides prepared from 0.01 grams of dry sediments dispersed in petri dishes, using a polarizing optical transmitted-light microscope at a high magnification (1250x) (de Vernal et al., 2010). Up to 100 fields of view, with a diameter of 160 μm , were examined. A minimum of 300 coccoliths of *Emiliana huxleyi* per sample were counted. In the case of rare occurrences, all coccoliths in 100 fields of view were counted.

Coccolith concentrations ($C_{\text{coccolith}}$; coccoliths g^{-1}) were computed according to:

$$(1) \quad C_{\text{coccolith}} = [(n \times (rp)^2) / (nc \times (rc)^2)] / p_{\text{se}}$$

where n is the total number of counted coccoliths, rp is the radius of the petri dish (cm), nc is the number of counted optical fields, rc is the radius of the optical field (cm), and p_{se} is the weight of the subsample (g). Coccolith fluxes ($F_{\text{coccolith}}$; coccoliths $\text{cm}^{-2} \text{yr}^{-1}$) were computed by multiplying the concentrations ($C_{\text{coccolith}}$) by the dry bulk sediment densities (ρ ; g cm^{-3} ; dry sediment weights in g of estimated 1 cm^3 volumes) and the sedimentation rates (cm yr^{-1}) reported by Gibb et al. (2014). The two *Gephyrocapsa* coccolith species identified (*Gephyrocapsa muellerae* and *Gephyrocapsa oceanica*) were reported together as *Gephyrocapsa* spp.

3.2 Alkenones

At Dalhousie University, total lipids were extracted from 2 to 4 grams of dried and homogenized sediments using an Accelerated Solvent Extraction (ASE) system. Extracts were saponified and purified by elution through a silica column. The purified extracts were then analyzed by flame ionization detection (GC-FID) (limit of detection $< 10 \text{ ng g}^{-1}$) to obtain total alkenone concentrations ($\text{C}_{37:2}$, $\text{C}_{37:3}$, and $\text{C}_{37:4}$ alkenones; ng g^{-1} ; Kienast et al., 2006, 2012).

Alkenone fluxes (F_{alkenone} ; $\text{ng cm}^{-2} \text{ yr}^{-1}$) were calculated by multiplying alkenone concentrations by the dry bulk sediment densities (ρ ; g cm^{-3}) and the sedimentation rates (cm yr^{-1}).

3.3 Foraminifers

Benthic foraminifers were counted using the $> 106 \mu\text{m}$ fraction of the 5 cm^3 wet-sieved bulk sediments and planktonic foraminifers were counted using the $> 150 \mu\text{m}$ size fraction (Gibb et al., 2014). The benthic and planktonic foraminifer concentrations (C_{foram} ; shells g^{-1}) are:

$$(2) \quad C_{\text{foram}} = (n \times a \times s) / p_{\text{se}}$$

where n is the total number of counted foraminifers, a is the reciprocal of the aliquot fraction, s is the ratio of plate area over the area analyzed, and p_{se} is the weight (g) of the subsample analyzed. Foraminifer fluxes (F_{foram} ; shells $\text{cm}^{-2} \text{ yr}^{-1}$) were computed by multiplying concentrations (C_{foram}) by the dry bulk sediment densities (ρ ; g cm^{-3}) and the sedimentation rates (cm yr^{-1}).

3.4 Calcium carbonate dissolution indices

Three dissolution indices were used: 1) the fragmentation index based on counts of planktonic foraminifer fragments and whole shells (Williams et al., 1985; Malmgren, 1987; Le and Shackleton, 1992; Wang et al., 2016), 2) the benthic-to-planktonic foraminifer ratio based on counts of whole shells (Conan et al., 2002; Kucera, 2007), and 3) the ratio of benthic foraminifer organic linings to benthic foraminifer calcite shells (de Vernal et al., 1992).

To determine the extent of planktonic foraminifer fragmentation (in %), whole and fragments (less than one half of the original shell) (Malmgren, 1987) of planktonic foraminifer shells were counted in populations of > 300 specimens, where possible, in the $> 150 \mu\text{m}$ size

fraction (Le and Shackleton, 1992; Kucera, 2007). The following fragmentation index was applied:

$$(3) \quad \text{Fragmentation index} = (\text{fragments}/8)/((\text{fragments}/8) + \text{whole shells}) \times 100$$

The number of fragments was divided by eight because it was assumed that a planktonic foraminifer breaks into eight fragments (Le and Shackleton, 1992). Even though planktonic foraminifer populations in subpolar regions are of low biodiversity and are almost entirely comprised of *Npl*, which is the planktonic foraminifer species the most resistant to dissolution (Malmgren, 1983/84), this fragmentation index can still be used to infer dissolution at high latitudes (e.g., Malmgren, 1983/84; Henrich, 1986; Zamelczyk et al., 2012, 2013). Also, the fragmentation index is a measure of calcite dissolution intensity that is independent of the composition of planktonic foraminiferal faunas (Kucera et al., 1997). Fragmentation represents the weakening of shells from dissolution (Conan et al., 2002) although foraminifers can also break apart from abrasion during sedimentation or from sieving (de Vernal et al., 1992; Kotler et al., 1992) independent of any dissolution effects. Fragmentation in excess of 40 % was set as the boundary for strong calcite dissolution (Kucera et al., 1997) because Malmgren (1987) noted that dissolution effects on the composition of planktonic foraminiferal assemblages were substantially higher in samples with a fragmentation in excess of 40 %. Note that a small number of shells found in a sediment sample can create a strong bias. Henrich (1986) suggested that dissolution can be recorded using the conventional fragmentation index with *Npl* from high latitudes, but this author also presents a more sensitive SEM-based dissolution index that is based on the most prominent ultrastructural changes of *Npl* during progressive dissolution (Kennett and Srinivasan, 1980; Henrich, 1986, Henrich et al, 1989). Because it is time-consuming, the Henrich (1986) SEM-based approach was not pursued in our study. Foraminifer

whole shells and fragments were bulk sampled and counted for this study with a binocular microscope only.

The ratio of the benthic-to-planktonic foraminifer concentrations (B/P) is:

$$(4) \quad B/P = [\text{benthic foraminifers}]/[\text{planktonic foraminifers}]$$

Deep-sea benthic foraminifers generally have thick, smooth shells that are more resistant to dissolution than those of planktonic foraminifers. As such, B/P increases with increasing calcite dissolution (Thunell, 1976; Kucera, 2007) so high values approaching infinity indicate dissolution. The B/P ratio provides more accurate estimates of changes at a single location than between locations due to regional variations in productivity and water depth, and B/P must be interpreted with caution since B/P has also been used as a non-independent indicator of productivity and water depth (Berger and Diester-Hass, 1988; van der Zwaan et al., 1990; Mleneck-Vautravers, 2018). The B/P ratio can also record very pronounced changes during large scale climatic changes between glacial and interglacial times, primarily because of changes in productivity and secondarily because of calcite dissolution (Henrich, 1986). It also seems possible that the B/P ratio would be impacted by freshwater influx, which modifies productivity in the surface waters.

The concentrations of organic linings (OL) and calcareous shells of benthic foraminifers (B) as a dissolution index (de Vernal et al., 1992) was calculated as follows:

$$(5) \quad \text{Dissolution index} = \log(OL/B)$$

In the absence of calcite dissolution, values of OL/B are close to zero so $\log(OL/B)$ values are negative (de Vernal et al., 1992). Conversely, calcite dissolution yields high OL/B ratios, which means strongly positive $\log(OL/B)$ values, even reaching infinity. In nearby core 007 (Fig. 1; de

Vernal et al., 1992), five out of twenty benthic foraminifer taxa yielded organic linings (de Vernal et al., 1992) and only a few species produce organic linings that fossilize in the sediment (Leduc, 2001). Nevertheless, the log(OL/B) index (de Vernal et al., 1992) with fragmentation and B/P indices (Thunell, 1976; Le and Shackleton, 1992) can provide valuable insight into calcite preservation and dissolution in the northwest Labrador Sea.

4. The stratigraphical context

Core 004 sediments are predominately composed of gray silty clays and interbedded layers of graded mud of low magnetic susceptibility and high carbonate content (Campbell and de Vernal, 2009; Gibb et al., 2014; Nuttin et al., 2015), corresponding to Labrador Sea detrital carbonate events (Andrews and Tedesco, 1992; Andrews et al., 1995). Large variations are observed in the CaCO_3 , IRD, $\delta^{13}\text{C-Npl}$, and $\delta^{18}\text{O-Npl}$ records (Fig. 3). The CaCO_3 percentages are low in the glacial interval except for the Heinrich event 1 (H1) layer, where the percentages jump from nil to up to 60 % (extremely high detrital carbonate) (Gibb et al., 2014; Nuttin et al., 2015). Likewise, the CaCO_3 percentages are low in the deglacial and postglacial sediments (< 20 %), with a slight trend towards higher percentages into the late postglacial. The IRD percentages are highest in the Heinrich layer, reaching nearly 18.2 % in H1, then they fall to near zero and approach ~ 8 % in some samples of the late glacial, followed by percentages < 2 % in both the deglacial and postglacial intervals (Gibb et al., 2014).

The $\delta^{13}\text{C-Npl}$ values are lowest in the glacial interval (-0.23—0.70 ‰) and increase to consistently positive values (0.06—0.94 ‰) in the deglacial and postglacial intervals, which can be interpreted as a change from stratified waters in the glacial to a ventilated subsurface water mass into the postglacial (de Vernal and Hillaire-Marcel, 2006; Ravelo and Hillaire-Marcel,

2007; Gibb et al., 2014). The $\delta^{18}\text{O}$ -Npl values are high in the glacial interval (up to 4.8 ‰), with the exception of excursions to lower values (< 3 ‰) at the base and top of H1 (Gibb et al., 2014). The $\delta^{18}\text{O}$ -Npl values steadily decrease into the deglacial and the postglacial intervals (as low as 2.6 ‰; Fig. 3). The excursions associated with H1 have been recorded in other cores (Hillaire-Marcel et al., 1994a; Hillaire-Marcel and Bilodeau, 2000; Rashid et al., 2011) and are interpreted as the result of brine extrusion during episodes of enhanced sea-ice formation (Hillaire-Marcel and de Vernal, 2008; Gibb et al., 2014). Similarly, Jennings et al. (2015) and Rashid et al. (2017) also document lighter $\delta^{18}\text{O}$ values from both planktonic and benthic foraminifers from the Labrador Shelf that correspond to peaks in detrital carbonate and high IRD percentages.

5. Results

5.1 Coccoliths

Coccoliths (Fig. S1) are easily identifiable in core 004 using the polarized light microscope, even in samples of minimal abundance from the glacial interval (Table S2).

In the glacial interval, older than 12.2 ka, only three out of twenty-seven samples contain coccoliths, with a mean concentration and flux of $9.5 \pm 3.9 \times 10^5$ coccoliths g^{-1} and $9.1 \pm 3.2 \times 10^4$ coccoliths $\text{cm}^{-2} \text{yr}^{-1}$, respectively (Tables S2 and S3). The three samples that contain coccoliths are characterized by low concentrations and the occurrence of only five species, dominated by *Coccolithus pelagicus*, and the second most abundant species is *Emiliania huxleyi*. Coccolith concentrations and fluxes in the deglacial unit (12.2–8.3 ka) average $1.0 \pm 0.3 \times 10^8$ coccoliths g^{-1} and $1.2 \pm 0.7 \times 10^7$ coccoliths $\text{cm}^{-2} \text{yr}^{-1}$, respectively, in the five out of eight

samples containing coccoliths (Tables S2 and S3). In this interval, the non-barren mean percentage of *Emiliania huxleyi* is $49 \pm 3 \%$ (Fig. 4; Table S4). All seventeen samples of the postglacial (< 8.3 ka) contain coccoliths and their mean concentration and flux are $2.1 \pm 0.1 \times 10^8$ coccoliths g^{-1} and $2.1 \pm 0.5 \times 10^6$ coccoliths $\text{cm}^{-2} \text{yr}^{-1}$, respectively. In this interval, *Coccolithus pelagicus* is the most abundant ($61 \pm 1 \%$) species of all coccolithophores identified, followed by *Emiliania huxleyi* ($30 \pm 1 \%$) and *Calcidiscus leptoporus* ($4.8 \pm 0.6 \%$) (Fig. 4). *Emiliania huxleyi* is consistently present among the coccolith assemblages. The other species, *Helicosphaera carteri*, *Gephyrocapsa* spp., *Umbilicosphaera sibogae*, *Pontosphaera discopora*, *Rhabdosphaera clavigera*, *Syracosphaera pulchra*, and *Reticulofenestra producta* occur in low numbers (Table S4).

5.2 Alkenones

Alkenone concentrations show little variation with an overall mean concentration of 77 ± 9 ng g^{-1} and a mean flux of 6.7 ± 1.0 ng $\text{cm}^{-2} \text{yr}^{-1}$ (Fig. 5; Tables S2 and S3). Most samples with the highest alkenone fluxes in this core are from the glacial interval (Fig. 5) with a mean concentration and flux of 72 ± 6 ng g^{-1} and 8.4 ± 1.1 ng $\text{cm}^{-2} \text{yr}^{-1}$, respectively. The mean concentration of alkenones in the deglacial interval is statistically identical to that of the glacial interval, at 82 ± 12 ng g^{-1} , as is the mean flux of 7.8 ± 3.2 ng $\text{cm}^{-2} \text{yr}^{-1}$. Similarly, for the postglacial, the mean alkenone concentration is 84 ± 28 ng g^{-1} . Due to the lower sedimentation rates during the postglacial, the postglacial alkenone flux is only 0.6 ± 0.2 ng $\text{cm}^{-2} \text{yr}^{-1}$, respectively. Alkenones were not detected in the sample at ~ 1.6 ka and in one of the samples dated at 8.2 ka. The highest alkenone concentration in the entire record is found at 3.1 ka (457 ng g^{-1}).

For the entire record (excluding samples with zero *Emiliania huxleyi* and zero alkenones), the alkenone concentrations show no significant relationship with the total concentrations of *Emiliania huxleyi* ($r^2 = 4.3 \times 10^{-4}$, $p = 0.93$), which is the main alkenone producer, and the other abundant coccolithophore species do not produce alkenones (Volkman et al., 1980b; Marlowe et al., 1984b, 1990; Rosell-Melé et al., 2000). A lack of significant relationship also characterizes the deglacial ($r^2 = 0.15$, $p = 0.51$) and postglacial ($r^2 = 0.0096$, $p = 0.73$) intervals when considered separately (Fig. 6). The glacial interval does not have enough samples with coccoliths ($n=3$) for a correlation. The statistics are not significant and nearly identical when *Gephyrocapsa* spp. are added to *Emiliania huxleyi* for the entire record ($r^2 = 3.8 \times 10^{-4}$; $p = 0.93$), for the deglacial ($r^2 = 0.15$; $p = 0.51$), and for the postglacial ($r^2 = 0.011$; $p = 0.71$) intervals.

5.3 Foraminifers and dissolution indices

According to binocular microscope and SEM examinations, foraminifer shells in our samples do not show signs of corrosion. Foraminifer concentrations are much higher in the deglacial and parts of the postglacial intervals (up to 28,700 planktonic foraminifers g^{-1} and 2290 benthic foraminifers g^{-1}) than in the glacial interval (up to 15,900 planktonic foraminifers g^{-1} and 440 benthic foraminifers g^{-1}) (Fig. 7). Benthic foraminifers are absent in seven of the twenty-seven samples from the glacial interval where planktonic foraminifers are found in low concentrations (Table S2). Mean planktonic and benthic foraminifer concentrations (excluding barren samples) from the glacial interval are 2860 ± 580 shells g^{-1} and 150 ± 28 shells g^{-1} , respectively. Three foraminifer dissolution indices indicate good preservation for most samples throughout the core record (Table S5).

Overall, the fragmentation index remains quite low (combined deglacial and postglacial mean of $\sim 4\%$, where foraminifers are in high abundance), but the deglacial fragmentation index does reach up to 8% (Fig. 8; Table S5). A cluster of values up to 16% peak in samples from the Bølling-Allerød of the late glacial (containing moderate-to-high concentrations; $\times 10^3$). One sample from the H1 layer, with a high fragmentation index of 20% and the highest of our record, is not statistically representative because only two fragments and one whole shell were found in that sample. The rest of the samples contain planktonic foraminifers with concentrations on the order of $\times 10^2$ – 10^4 foraminifers g^{-1} , therefore, the other fragmentation percentages are based on representative samples. The fragmentation index of the deglacial interval (up to 8%) is based on high counts of foraminifer shells. In the postglacial interval, fragmentation is low (Fig. 8), ranging from 2.8 to 5.7% , with a mean of $3.9 \pm 0.2\%$.

The B/P values are mostly low and near zero, indicating preservation. B/P values are null for the seven discrete samples of the glacial interval that contain no benthic foraminifers (red circles; Fig. 8). Excluding these samples, the B/P index remains low throughout the glacial interval (mean of 0.07 ± 0.01). Slightly higher B/P values (> 0.15 and < 0.8) characterize the deglacial interval, which may suggest slight or minor calcite dissolution. Despite the extreme climatic differences between the glacial and postglacial intervals, similarly low B/P ratios are recorded, suggesting overall good calcium carbonate preservation (Fig. 8).

The $\log(\text{OL/B})$ values of most samples are negative (whole-core mean of 0.7 with a range from -0.7 to 1.6), which overall indicate good preservation. However, several samples with positive values characterize samples from the Bølling-Allerød and the deglacial interval, which might mean slight dissolution as deduced also from the elevated B/P index (Fig. 8).

6. Discussion

6.1 Calcite preservation and IRD

The three calcium carbonate dissolution indices used in this study reveal that there has been no substantial calcium carbonate dissolution in the northwest Labrador Sea at 2674 m water depth over the past 25,000 years, consistent with findings of Aksu et al. (1989) for ODP site 646 (yellow circle, Fig. 1). Nevertheless, the slightly higher fragmentation index, B/P ratios, and the $\log(\text{OL/B})$ values could imply that marginally more corrosive conditions prevailed during the Bølling-Allerød and the deglacial interval (Fig. 8). Because of independent evidence for high productivity (Fig. 9; Gibb et al., 2014) at least in the deglacial interval, we cannot disregard the possibility of calcite dissolution at that time being related to high organic carbon fluxes to the seafloor leading to organic matter oxidation and accumulation of metabolic carbon dioxide and, thus, a decrease of the calcite saturation state in sediment porewaters (e.g., Mucci et al., 2000; Nesbitt and Mucci, 2020). The higher fragmentation of foraminifers during the Bølling-Allerød, an interval with low foraminifer abundances (Fig. 7) can alternatively be explained by turbulent sedimentary pulses from meltwater and high inputs of IRD causing fragmentation of the planktonic foraminifers in the sediments. The IRD occurrence is moderately high during the Bølling-Allerød, while it is near zero in the deglacial interval; Fig. 3). Thus, while foraminifer fragmentation in the samples of the Bølling-Allerød due to mechanical abrasion from IRD inputs seems possible, slight porewater calcite dissolution is better invoked for the deglacial interval (Table 1).

Due to the proximity of our core site to Davis Strait, we also need to consider the calcite saturation state of nearby water masses and the overall ocean circulation in the region. The

transition from the glacial to the postglacial (the deglacial) was marked by the onset of dissolution in Baffin Bay (Aksu, 1983) and in nearby Davis Strait (de Vernal et al., 1992). Calcite dissolution in Davis Strait since about 13–12 ka has been inferred from the study of core 007 collected in Davis Strait at 823 m water depth (Fig. 1; de Vernal et al., 1992) and of core 009 taken at 1434 m water depth in the northwest Labrador Sea (Fig. 1), in proximity to the shelf edge (Hillaire-Marcel et al., 1994a). Hence, overflow pulses of dense and deep waters from Baffin Bay through Davis Strait during the deglacial interval might have contributed to any undersaturated waters in the northwest Labrador Sea. However, the impact of Davis Strait overflow, if any, was not strong given the subtle indications for some dissolution.

In the sediment accumulated prior to the Bølling-Allerød of the late glacial interval, there is no evidence of dissolution. We observe low concentrations and fluxes from that interval, and therefore, we posit that low foraminifer and coccolith productivity characterize the last glacial interval in the northwest Labrador Sea.

6.2 Productivity

Based on the microfossil and geochemical records of the past 25,000 years, we interpret the overall coccolith and foraminifer abundances in sediments of core 004 as representative of regional biogenic carbonate production. This interpretation is consistent with the results of Gibb et al. (2014), who concluded that most of the northwest Labrador Sea was marked by a strongly stratified upper water column, quasi-perennial sea ice, and extremely low productivity until ~ 11.9 ka (Fig. 9). This is compatible with the low fluxes of all microfossils reported in this study and by Hillaire-Marcel et al. (1994a), Rochon and de Vernal (1994), and Gibb et al. (2014) as well as the low $\delta^{13}\text{C-Npl}$ documented by Ravelo and Hillaire-Marcel (2007) for the glacial

interval. The few planktonic foraminifers found in the glacial interval suggest limited open water and productivity, consistent with dinocyst-based reconstructions of past primary productivity (cf. Radi and de Vernal, 2008; Gibb et al., 2014). Freshening events would have also caused strong stratification over the northern Labrador Shelf, notably at ~ 8.5 ka (Lochte et al., 2019); however, our core is not from the shelf. Barber et al. (1999) indicated that a sudden increase in freshwater flux reduces sea surface salinity as well as LSW formation (Hillaire-Marcel et al., 2001), lowering nutrient availability and thus limiting primary productivity (e.g., Radi and de Vernal, 2008; Gibb et al., 2014, 2015).

Much higher concentrations of foraminifers and coccoliths characterized the deglacial and postglacial intervals compared to the glacial interval, indicating elevated productivity. The locally high concentrations of benthic foraminifers and the high productivity reconstructed from dinocysts (Figs. 7 and 9; Gibb et al., 2014) imply increased organic matter fluxes to the seafloor as time progressed from the glacial into the deglacial interval.

6.3 Decoupling of coccolith and alkenone abundances

Since there is no major or intense calcium carbonate dissolution in the core 004 sediments, the coccolith and alkenone concentrations would be expected to display parallel abundances (e.g., Weaver et al., 1999), particularly in the glacial and postglacial intervals where we infer no dissolution at all. However, relatively high alkenone fluxes are found in the glacial interval despite the coinciding minimal fluxes of coccoliths.

The coccolith assemblages of the sequence are consistent with regional paleoceanographic conditions described in previous studies (e.g., de Vernal et al., 1992, 2005;

Hillaire-Marcel et al., 1994b; Rahman and de Vernal, 1994; Gibb et al., 2014, 2015). In the glacial interval, the low coccolith concentrations and the dispersed yet dominant presence of *Coccolithus pelagicus* can be explained by low coccolithophore productivity due to very cold waters and dense sea-ice cover (cf. Gibb et al., 2014). In the postglacial sediments, the coccolith assemblages dominated by *Coccolithus pelagicus*, *Emiliania huxleyi*, and *Calcidiscus leptoporus* are compatible with similar to modern sea-surface temperatures (SST) and seasonal sea-ice cover at the coring site (Solignac et al., 2008).

The alkenone concentrations in sediments of core 004 are difficult to interpret, especially for the glacial interval. The relatively higher abundance of alkenones despite the rarity of coccoliths and the lack of evidence for calcite dissolution during the last glacial interval imply that the alkenones preserved in the sediments did not originate from the known calcifying and alkenone-producing coccolithophores. This leaves us with the following non-exclusive hypotheses:

- Alkenones derive from distal inputs, possibly including shelf erosion of Cretaceous or Paleogene limestones (e.g., Weaver et al., 1999; Rosell-Melé et al., 2011; Mao et al., 2014; Filippova et al., 2016). These would either have been synthesized by non-calcifying coccolithophores, or the coccoliths would have been altered or dissolved since the time of deposition.
- Alkenones were advected from more open and warmer regions such as the southern Irminger Sea/adjacent North Atlantic, made possible by northward penetration of southern-sourced intermediate waters of the North Atlantic during the deglaciation (Adkins et al., 1998; Rickaby and Elderfield, 2005; Pahnke et al., 2008; Thiagarajan et al., 2014). However, we do not have a mechanism to separate alkenones and coccoliths.

- Alkenones were produced locally and predominantly by haptophytes other than the calcifying *Emiliania huxleyi* during the glacial interval (e.g., Rosell-Melé et al., 2011).

There are a handful of *Emiliania huxleyi* coccoliths found in two glacial samples, so the high fluxes of alkenones throughout the glacial interval must have another source.

Sea-surface temperature estimates derived from alkenones can help us test the above hypotheses. As a tool to investigate the source of the alkenones, the U_{37}^K -based sea-surface temperature estimates (U_{37}^K -SST) in core 004 were compared with summer sea-surface temperatures derived from the modern analogue technique applied to dinocyst assemblages (Gibb et al., 2014). The U_{37}^K -SST equation was based on:

$$(6) \quad U_{37}^K = ([C_{37:2}] - [C_{37:4}]) / ([C_{37:2}] + [C_{37:3}] + [C_{37:4}])$$

where $C_{37:2}$, $C_{37:3}$, and $C_{37:4}$ are the di-, tri-, and tetra-unsaturated C_{37} alkenones, respectively (Brassell et al., 1986). The temperature equation was from Müller et al. (1998):

$$(7) \quad U_{37}^K\text{-SST} = (U_{37}^K - 0.044) / 0.033$$

The U_{37}^K -based sea-surface temperature estimates (U_{37}^K -SSTs) in core 004 (Fig. 10; Table S6) are low in the glacial sediments (mean of -4.4 ± 1.2 °C), especially in the sediments immediately above H1 (as low as -11 °C). Such values are incompatible with ocean waters that freeze at about -1.8 °C. There was a rapid shift to higher temperatures in the deglacial interval (mean of 5.4 ± 1.5 °C) and the temperatures remain relatively high throughout the postglacial interval (mean of 10.4 ± 1.6 °C). These temperature estimates were compared with summer sea-surface temperatures derived from the modern analogue technique applied to dinocyst assemblages (hereafter MAT-SST) (Gibb et al., 2014). The mean MAT-SST in the glacial interval is 1.4 ± 0.3 °C, which is consistent with other regional studies (e.g., de Vernal et al.,

2005, 2006), but $> 5\text{ }^{\circ}\text{C}$ higher than the mean of the temperatures from the $\text{U}^{\text{K}}_{37}\text{-SST}$. The MAT-SST estimates from the deglacial interval (mean of $10.1 \pm 0.6\text{ }^{\circ}\text{C}$) were higher than those of the postglacial interval ($6.5 \pm 0.3\text{ }^{\circ}\text{C}$). The $\text{U}^{\text{K}}_{37}\text{-SST}$ and MAT-SST temperature records are compatible from $\sim 12\text{ ka}$ to the present (Fig. 10).

The colder than possible $\text{U}^{\text{K}}_{37}\text{-SST}$ values from the glacial interval is problematic. If the alkenones in the glacial interval were produced by *Emiliania huxleyi* and *Gephyrocapsa* spp. and represent allochthonous input (Wheeler et al., 1996; Andrews et al., 2012; MacLean et al., 2014), they would have had to come from warmer environments and/or from redeposited sediments containing alkenones that deposited during a warmer time (e.g., Cretaceous sediments deposited during a global greenhouse climate; Zachos et al., 2001, 2008). However, the extremely low (unreasonably cold) $\text{U}^{\text{K}}_{37}\text{-SST}$ estimates rule out such a possibility.

The above considerations lead us to conclude that the alkenones recovered during the glacial interval were most likely produced by non-calcifying haptophytes (cf. Rosell-Melé et al., 2011) that likely have a temperature response different from haptophytes represented in the standard alkenone temperature calibrations. Because cold surface water and dense sea-ice cover have limited primary productivity in the glacial interval, the precise nature and source of the high alkenone fluxes during this interval remain unresolved.

7. Conclusions

The fluxes of biogenic remains to the seafloor at our core site and the preservation of carbonate foraminiferal shells indicate low to nil calcium carbonate dissolution but provide a picture of productivity in the upper water column. They reveal a large contrast in productivity

between glacial and postglacial conditions. Low fluxes of foraminifers and coccoliths during the glacial interval related to low productivity are consistent with nearly year-round persistent and dense sea-ice cover during that time as reconstructed by means of other proxies (see de Vernal et al., 2005; Gibb et al., 2014). Planktonic foraminifer and coccolith fluxes to the seafloor increased sharply during the deglacial interval, paralleled by an increase in benthic foraminifers, which together indicate an increase in primary productivity and export of organic matter to the poorly ventilated seafloor from surface waters, possibly causing minor calcite dissolution in the sediment porewaters. The record shows a minor increase in foraminifer fragmentation during the Bølling-Allerød and the subsequent deglacial interval. There was influence of late glacial age (the Bølling-Allerød) turbulent IRD inputs at our core site, followed by likely calcite undersaturation from high productivity in the deglacial interval. Our evidence suggests good preservation of calcium carbonate shells in the deep northwest Labrador Sea throughout the postglacial interval, which could be related to the establishment of modern-like conditions with penetration of the Irminger/Atlantic waters through the West Greenland Current into the Labrador Sea, as previously described by Gibb et al. (2014, 2015).

Whereas the alkenone record is compatible with the microfossil content of the postglacial interval, the alkenones of the glacial interval are difficult to interpret. Both the higher alkenone fluxes compared to the nearly absent coccoliths as well as the U^{K}_{37} indices that are incompatible with other paleoceanographic data, suggest alkenone production occurred most likely from non-calcifying haptophytes.

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Figures

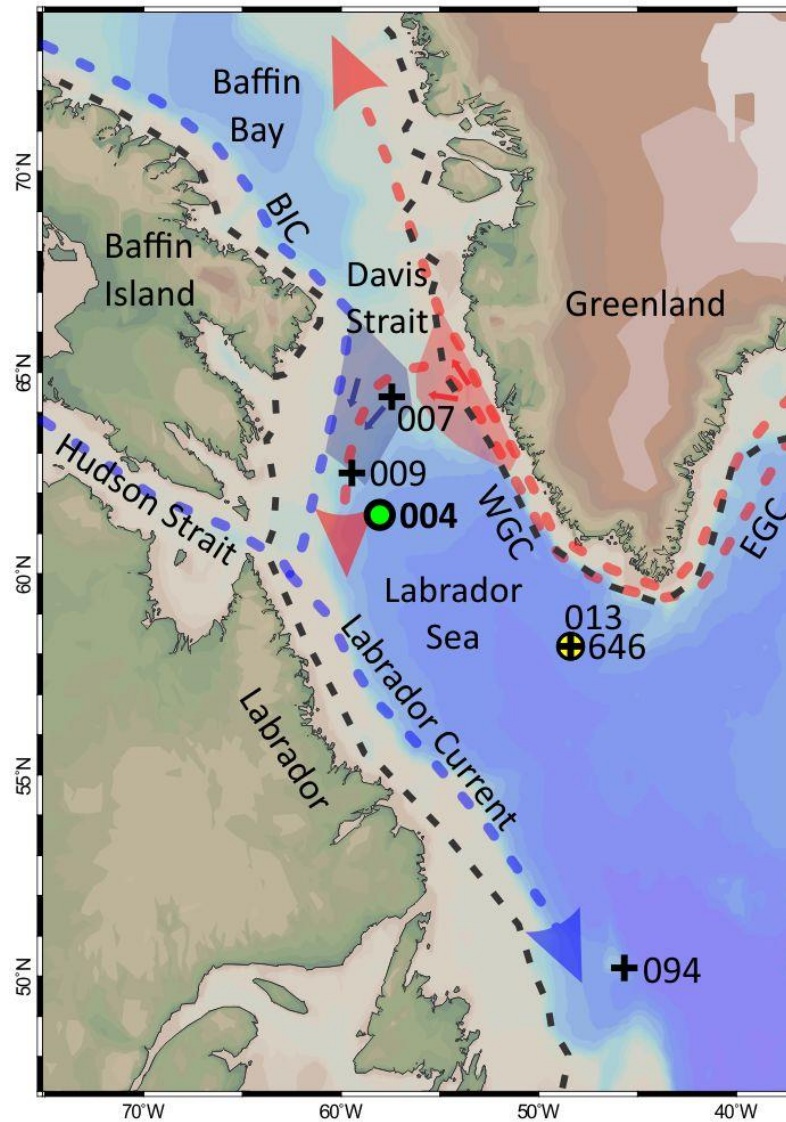


Figure 1. Map of the study area showing the core site, bathymetry, and modern surface currents (warmer in dashed red and colder in dashed blue; adapted from Lewis et al., 2012). The transparent shaded zones show slope waters flowing northward into Baffin Bay (red) and the southward Baffin Bay water overflow (blue) flowing into the Labrador Sea (Azetsu-Scott et al., 2010, 2012). The green circle shows the location of core 004. Crosses are sites of cores referred to in the text (de Vernal et al., 1992; Rahman and de Vernal, 1994; Hillaire-Marcel et al., 1994a, b; Rahman, 1995; and others), and the yellow circle shows the location of Ocean Drilling Program (ODP) core 646 (cf. Aksu et al., 1989). The approximate extent of the ice sheets during the LGM (black dotted lines) are based on estimates from Dyke et al. (2002) and Shaw (2006). Map was created in Ocean Data View (Schlitzer, 2018).

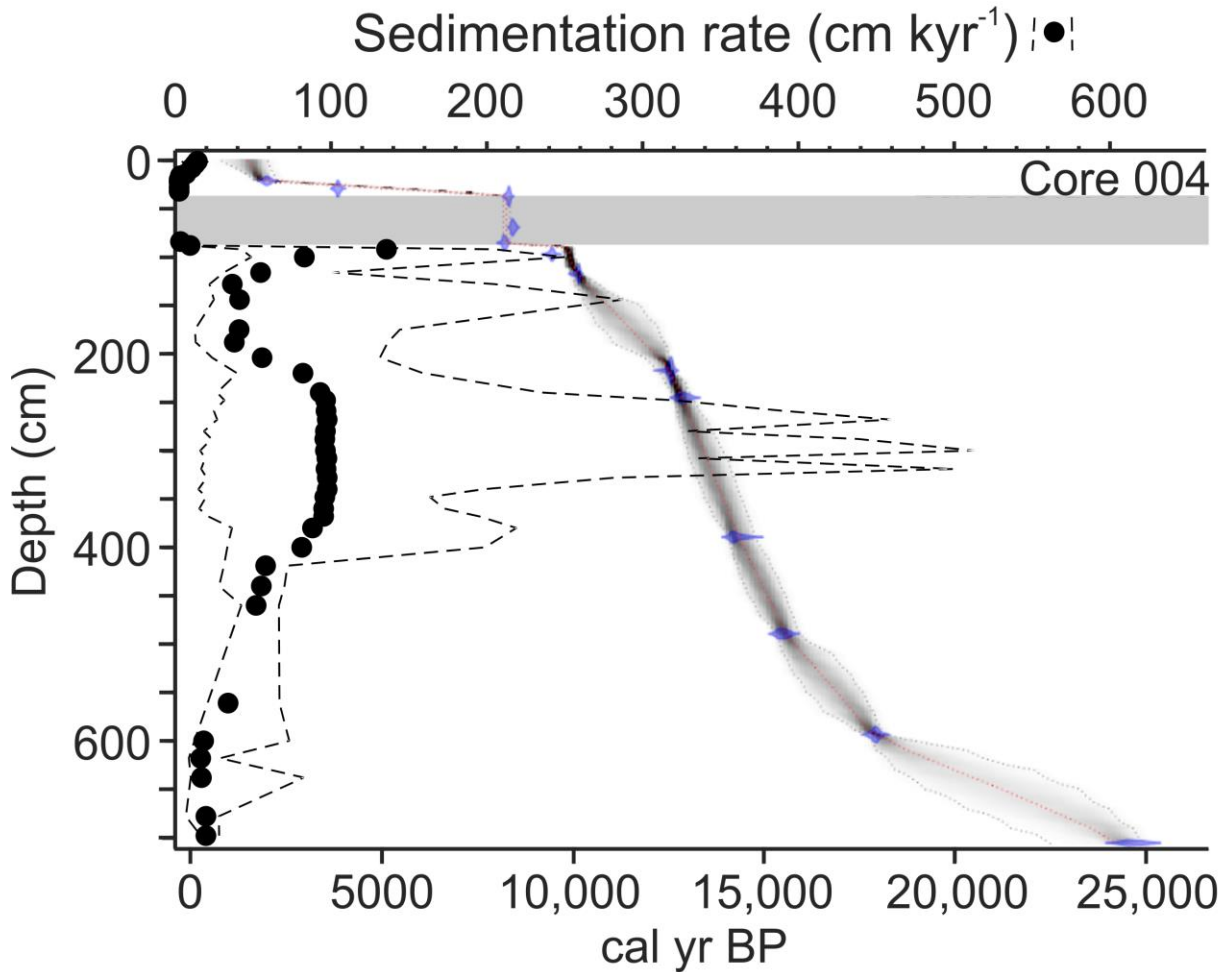


Figure 2. Age-depth relationship for core 004 based on the Bacon software (Blaauw and Christen, 2011). A slump (grey area) is identified between 36 and 85 cm. This age-depth model is modified from Gibb et al. (2014). The mean sedimentation rates (black circles; cm kyr⁻¹) with minimum and maximum sedimentation rates (dashed lines) are plotted on top of the age-depth relationship.

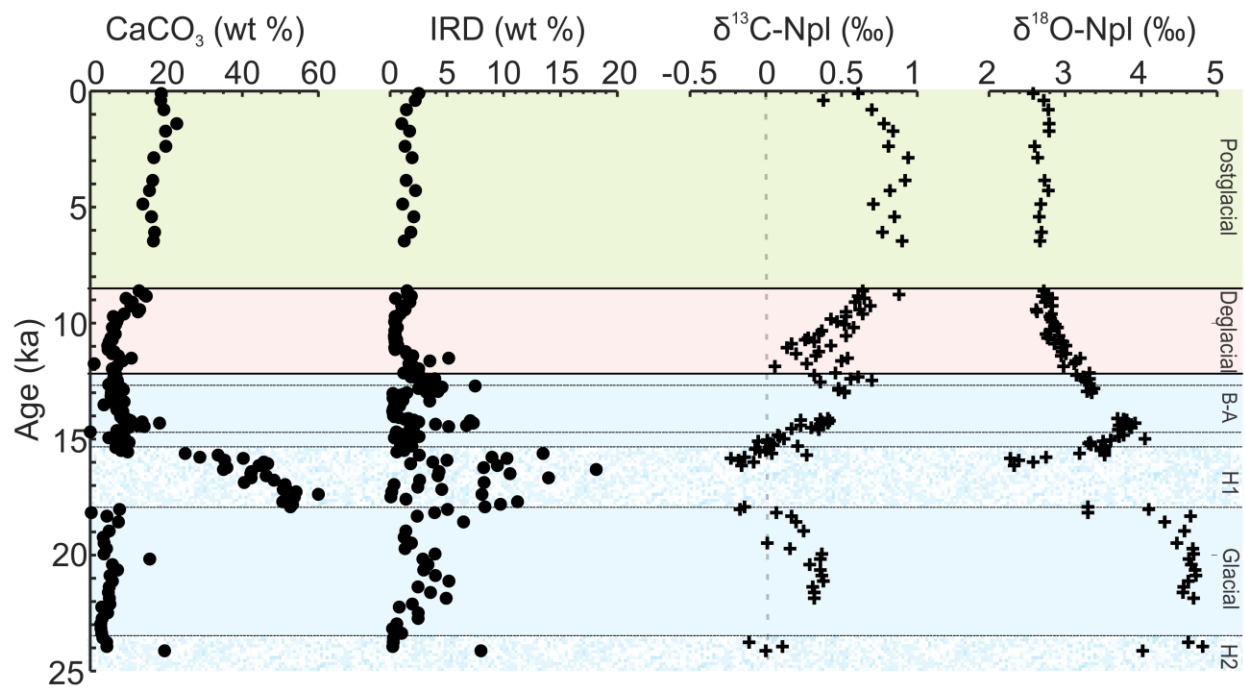


Figure 3. Percentages of CaCO₃ and IRD, and isotope stratigraphy (δ¹³C-Npl and δ¹⁸O-Npl) (Gibb et al., 2014). The glacial (light blue), deglacial (pink), and postglacial (green) intervals are presented, along with the Heinrich (H) layers identified with light blue dots (H1 and the top of H2) (from Gibb et al., 2014) and the Bølling-Allerød (B-A) of the late glacial (Thiagarajan et al., 2014; Dendievel et al., 2019). The color scheme for these time intervals remain the same for all figures that have age on the y axis. The vertical dotted line with the δ¹³C-Npl data is a zero-line to highlight the negative from positive values.

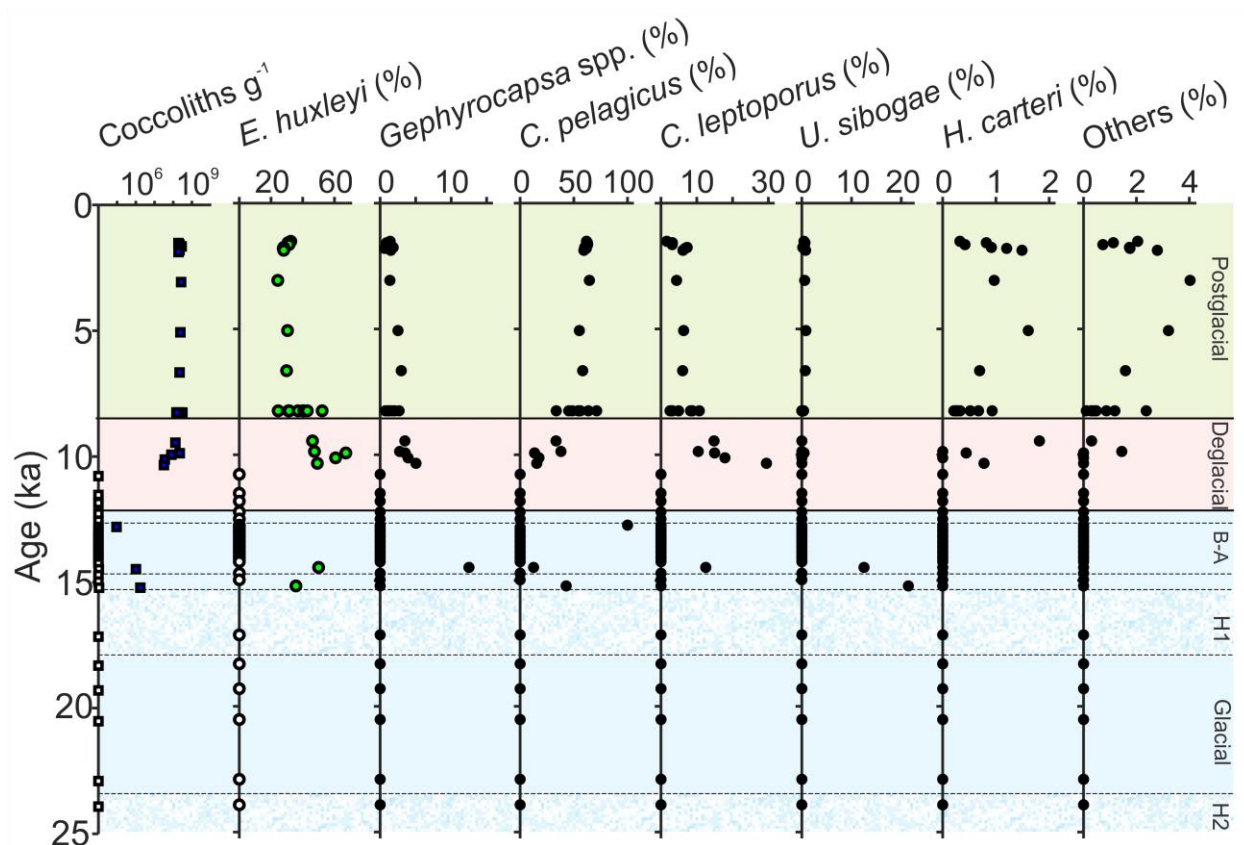


Figure 4. Total coccolith concentrations and percentages of coccolith species. Total coccoliths and *Emiliania huxleyi* are plotted as blue squares and green circles, respectively (see also Fig. 5). No coccoliths are indicated by empty squares and no *Emiliania huxleyi* are indicated by empty circles.

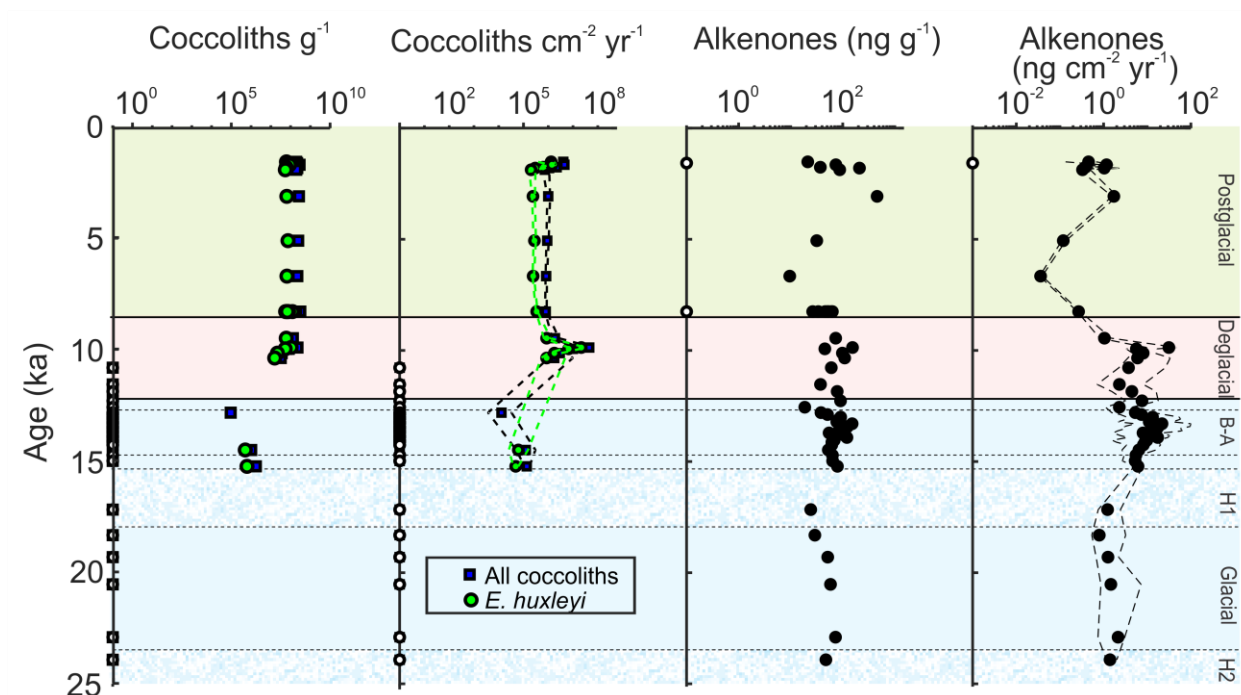


Figure 5. Concentrations and fluxes of total coccoliths (all species; blue squares), *Emiliania huxleyi* (green circles), and alkenones (black circles) in sediments from core 004. Samples with no coccoliths, *Emiliania huxleyi*, or alkenones are indicated by empty symbols.

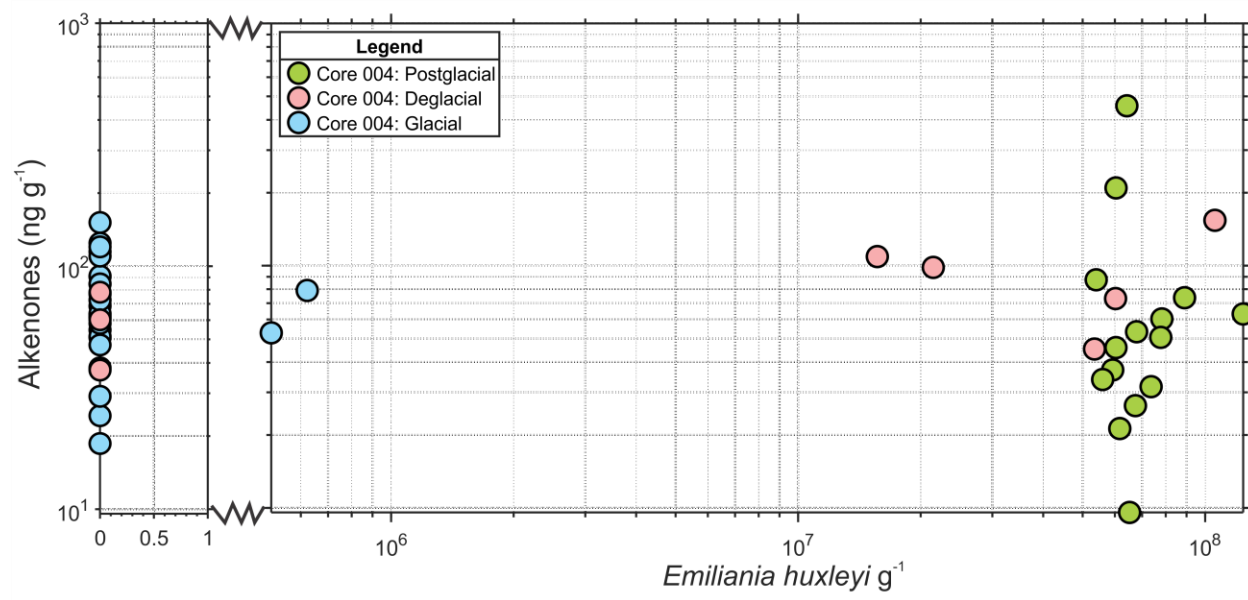


Figure 6. Alkenone versus *Emiliana huxleyi* concentrations in samples from core 004. Data are grouped by intervals: glacial (light blue), deglacial (pink), and postglacial (green).

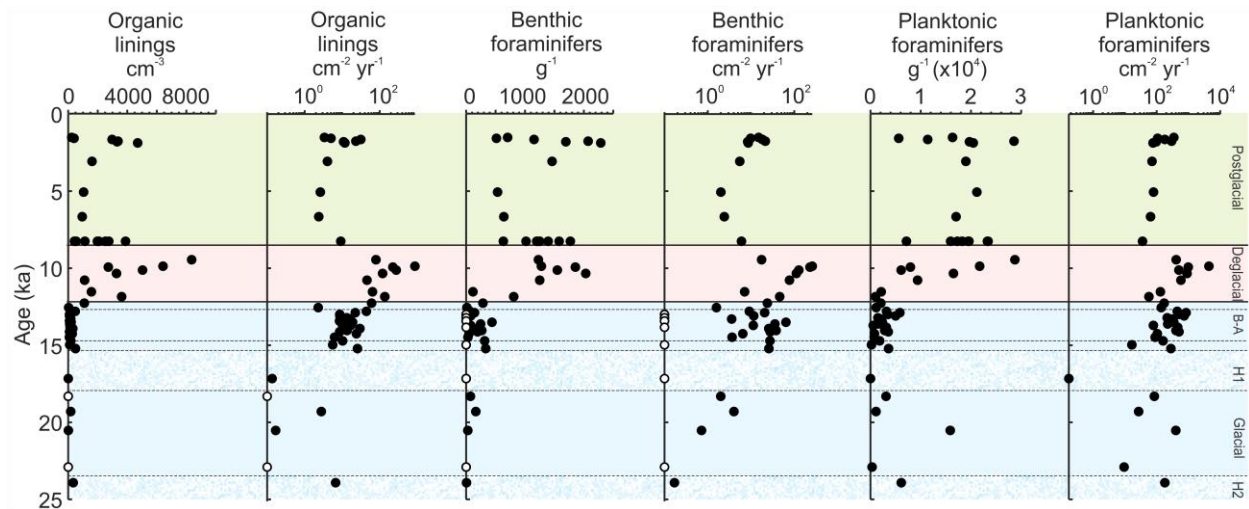


Figure 7. Concentrations and fluxes of organic linings (Gibb et al., 2014) as well as benthic and planktonic foraminifera (this study) in core 004.

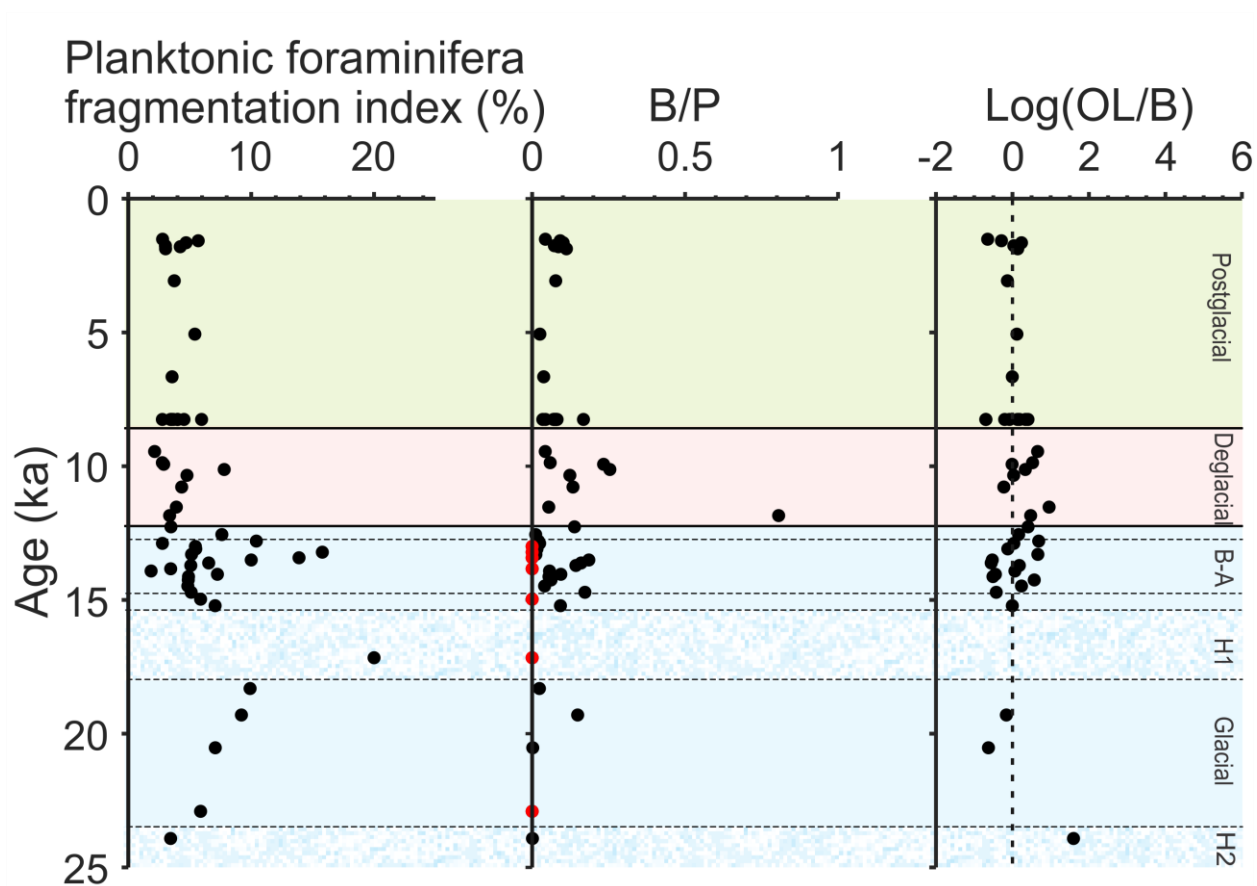


Figure 8. Planktonic foraminifera fragmentation index (%), B/P index, and log(OL/B) in core 004. Values of 0 B/P are identified as the red circles. The grey dotted line is a zero line to easily identify the negative from the positive log(OL/B) values.

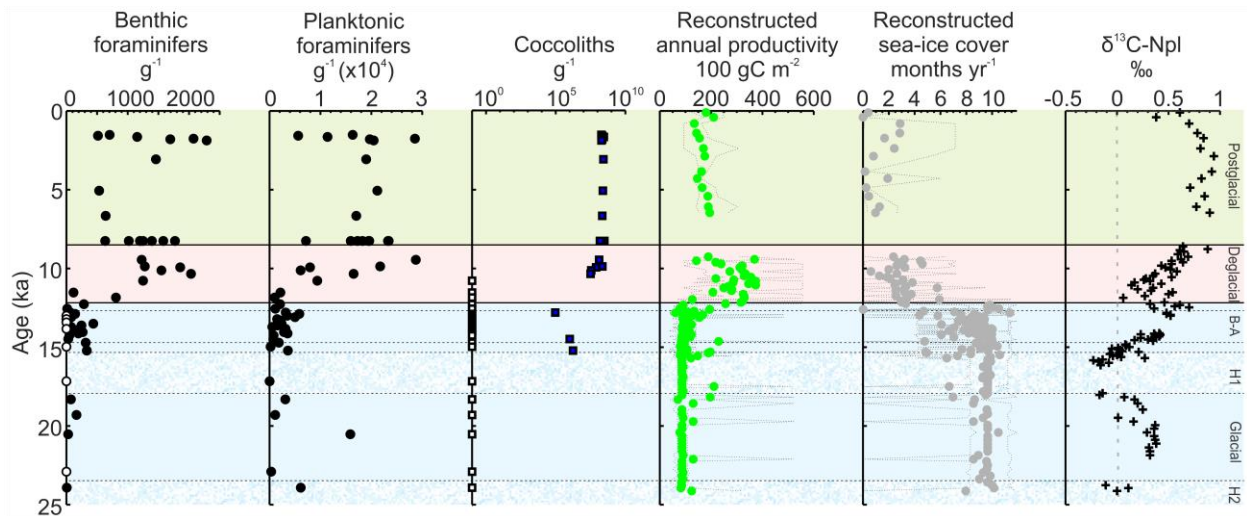


Figure 9. Downcore concentrations of benthic and planktonic foraminifera (black circles) and total coccoliths (red circles), with estimates of annual productivity (green circles) and sea-ice cover (grey circles) derived from dinocyst assemblages and $\delta^{13}\text{C-Npl}$ (black crosses) from Gibb et al. (2014). The grey dotted lines correspond to minimum and maximum estimates of productivity and sea-ice cover based on a set of five analogues (Gibb et al., 2014). The vertical dotted line with $\delta^{13}\text{C-Npl}$ is a zero-line separating the negative from positive values.

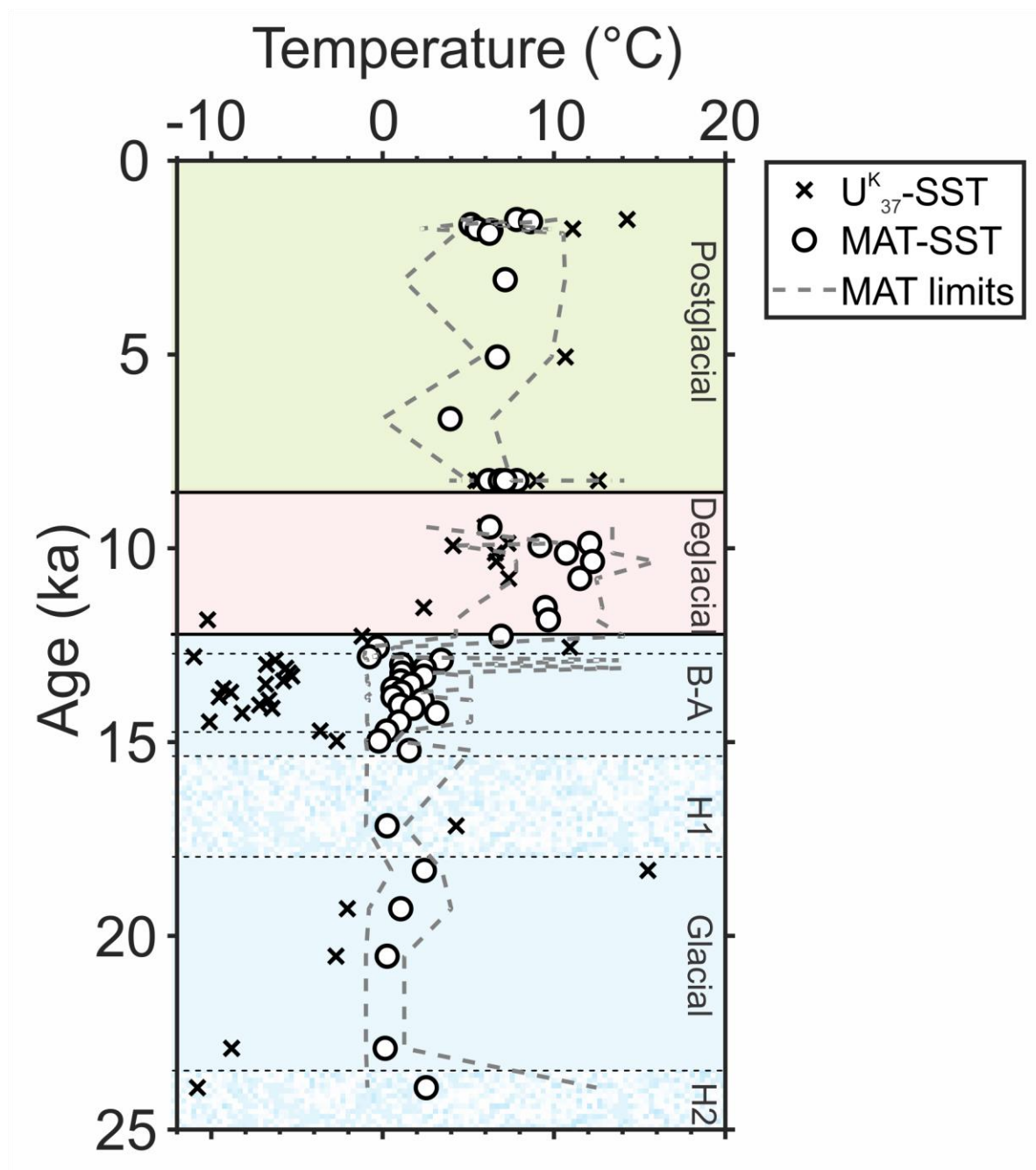


Figure 10. U^{K}_{37} -based sea-surface temperature estimates (U^{K}_{37} -SST; black crosses) and summer sea-surface temperatures calculated from the modern analogue technique (MAT-SST; white circles). The grey dotted lines indicate the lower and upper MAT-SST limits (see Gibb et al., 2014).

Table

Interval (time in ka)	PF Frag. (%)	IRD (%)	PF concentration (PF g ⁻¹)	Coccolith concentration (coccoliths g ⁻¹)	Dinocyst annual productivity from Gibb et al. (2014) (100 gC m ⁻²)	Fragmentation interpretation
Deglacial (12.2 to 8.3)	Lower ↓ < 8; mean: 4	Lower ↓ < 6; mean: 1.5	Elevated ↑ < 28742 mean: 11694	Elevated ↑ < 2×10^8 ; mean: 1×10^8	Elevated ↑ < 373 mean: 291	Elevated productivity and carbon flux to the seafloor likely caused oxic remineralization in sediment porewaters leading to porewater calcite undersaturation.
Bølling- Allerød (14.75–12.7)	Elevated ↑ < 16; mean: 7	Elevated ↑ < 8; mean: 1.6	Lower ↓ < 5847 mean: 2573	Lower ↓ < 1×10^6 mean: 5×10^5	Lower ↓ < 168 Mean: 103	Low productivity during elevated mechanical abrasion from turbulent sedimentary and meltwater pulses.

Table 1. Comparison and interpretation of the Bølling-Allerød and the deglacial interval for core 004 with IRD, foraminifer and coccolith concentrations, and dinocyst productivity (Gibb et al. 2014). The mechanism behind the elevated fragmentation percentages changed from the Bølling-Allerød to the deglacial interval. We interpret that mechanical abrasion characterizes the fragmentation of the Bølling-Allerød, while porewater calcite dissolution best explains the lower, but still elevated fragmentation percentages from the deglacial interval.

Supplemental figure and tables

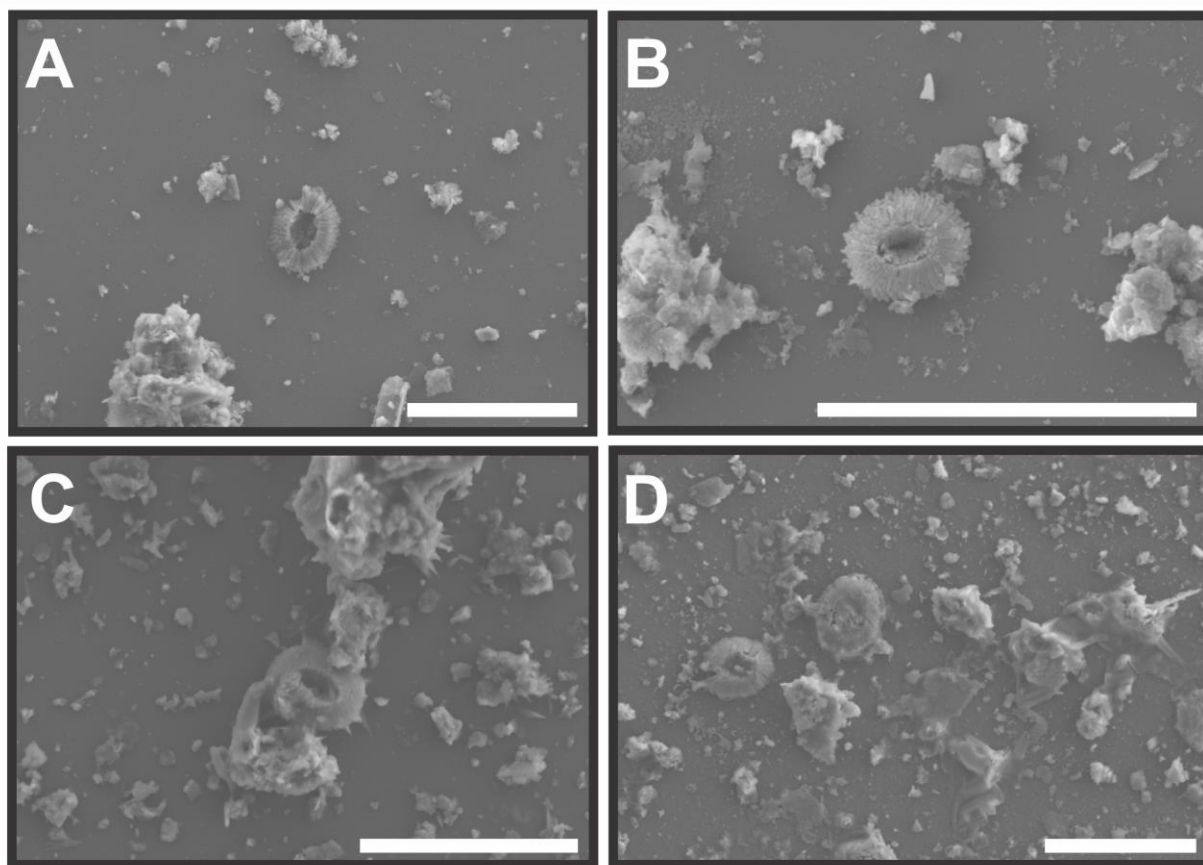


Figure S1. Scanning Electron Microscope (SEM) images of coccoliths from core 004. Images A and B are from the 1–2 cm depth interval (0.1 ka). Images C and D are from the 23–24 cm depth interval (2.9 ka). Scale bars are 20 μm .

Depth interval (cm)	Min. age (ka)	Mean age (ka)	Max. age (ka)	Est. sed. density (g cm ⁻³)	Min. sed. rate (cm kyr ⁻¹)	Mean sed. rate (cm kyr ⁻¹)	Max. sed. rate (cm kyr ⁻¹)
1–2	0.860	1.516	2.028	1.5	4.21	14.18	21.15
4–5	1.007	1.572	2.053	1.5	6.25	12.70	18.72
8–9	1.204	1.647	2.093	1.5	8.05	10.56	15.38
14–15	1.471	1.759	2.186	1.5	6.13	6.88	10.20
16–17	1.544	1.797	2.213	1.5	3.13	3.29	8.19
20–21	1.644	1.872	2.273	1.5	2.11	2.51	3.77
23–24	2.884	3.068	3.393	1.5	2.29	2.51	2.99
28–29	4.939	5.063	5.265	1.5	2.33	2.51	2.87
32–33	6.571	6.658	6.780	1.5	2.38	2.51	2.72
36–37	8.173	8.253	8.350	1.5	-	-	-
40–41	8.173	8.253	8.350	1.5	-	-	-
44–45	8.173	8.253	8.350	1.5	-	-	-
48–49	8.173	8.253	8.350	1.5	-	-	-
68–69	8.173	8.253	8.350	1.5	-	-	-
72–73	8.173	8.253	8.350	1.5	-	-	-
80–81	8.173	8.253	8.350	1.5	-	-	-
84–85	8.173	8.253	8.350	1.5	3.13	3.34	3.64
88–89	9.329	9.450	9.578	1.5	7.39	9.52	13.70
92–93	9.741	9.870	10.005	1.5	42.55	135.59	205.13
100–101	9.794	9.929	10.058	1.5	48.78	82.90	250.00
116–117	9.956	10.122	10.222	1.5	31.17	54.79	100.84
128–129	10.051	10.341	10.700	1.5	22.01	36.61	205.13
144–145	10.280	10.778	11.421	1.5	24.82	41.28	287.04
175–176	10.818	11.529	12.243	1.5	12.65	41.01	144.23
188–189	11.047	11.846	12.418	1.5	13.10	37.91	136.48
204–205	11.880	12.268	12.525	1.5	23.70	55.75	131.39
220–221	12.295	12.555	12.709	1.5	39.68	81.97	159.09
240–241	12.587	12.799	13.045	1.5	26.85	93.02	235.29
248–249	12.651	12.885	13.173	1.5	31.61	96.49	320.00
259–260	12.729	12.999	13.318	1.5	24.79	96.77	390.48
268–269	12.769	13.092	13.508	1.5	26.91	97.56	459.46
280–281	12.863	13.215	13.652	1.5	18.39	96.39	327.87
288–289	12.907	13.298	13.780	1.5	23.26	96.00	437.96
300–301	13.007	13.423	13.925	1.5	16.03	96.39	512.82
308–309	13.045	13.506	14.037	1.5	19.16	97.35	334.88
319–320	13.171	13.619	14.130	1.5	16.64	96.77	500.00
328–329	13.239	13.712	14.224	1.5	20.13	97.56	282.35
340–341	13.379	13.835	14.313	1.5	14.87	97.56	197.50
348–349	13.435	13.917	14.408	1.5	19.77	96.00	163.41
360–361	13.607	14.042	14.508	1.5	15.41	95.24	172.79
368–369	13.683	14.126	14.602	1.5	21.09	95.24	195.12
380–381	13.930	14.252	14.697	1.5	36.43	88.11	218.98
400–401	14.144	14.479	14.913	1.5	33.39	81.20	198.68
419–420	14.299	14.713	15.190	1.5	31.25	58.01	72.04
440–441	14.505	14.971	15.435	1.5	28.17	55.25	70.10
460–461	14.780	15.215	15.643	1.5	42.42	51.90	66.53
561–562	16.431	17.161	17.728	1.5	20.74	33.91	66.90
600–601	17.837	18.311	19.054	1.5	12.30	18.20	73.17
618–619	18.233	19.300	20.809	1.5	8.73	16.33	28.67
638–639	18.868	20.525	22.417	1.5	9.92	16.83	82.47
678–679	20.990	22.902	24.216	1.5	6.83	19.70	28.00
698–699	22.215	23.917	24.777	1.5	16.33	19.70	28.17

Table S1. Depth intervals, ages, sediment densities, and the sedimentation rates. Bulk dry sediment densities were assumed to be 1.5 g cm⁻³ (as per Hillaire-Marcel et al., 1994a; Andrews et al., 2018).

Age (ka)	Total Coccoliths g ⁻¹	<i>C.</i> <i>pelagicus</i> g ⁻¹	<i>E.</i> <i>huxleyi</i> g ⁻¹	<i>C.</i> <i>leptoporus</i> g ⁻¹	<i>Geph.</i> spp. g ⁻¹	<i>U. sibogae</i> g ⁻¹	<i>H.</i> <i>carteri</i> g ⁻¹	<i>P.</i> <i>discopora</i> g ⁻¹	<i>R.</i> <i>clavigera</i> g ⁻¹	<i>S.</i> <i>pulchra</i> g ⁻¹	<i>R.</i> <i>producta</i> g ⁻¹	Alkenones ng g ⁻¹	BF g ⁻¹	PF g ⁻¹	OL cm ⁻³ (Gibb et al., 2014)
1.516	1.90E+08	1.17E+08	6.17E+07	3.07E+06	2.66E+06	8.20E+05	6.15E+05	1.43E+06	0	2.05E+06	4.10E+05	21.26	705	1.63E+04	237
1.572	2.13E+08	1.33E+08	6.54E+07	6.76E+06	1.96E+06	1.31E+06	1.74E+06	1.31E+06	0	8.72E+05	2.18E+05	0	514	5.62E+03	396
1.647	2.84E+08	1.79E+08	8.90E+07	8.90E+06	2.97E+06	1.48E+06	1.19E+06	1.48E+06	0	0	5.93E+05	73.74	1154	1.14E+04	2973
1.759	2.12E+08	1.27E+08	5.93E+07	1.55E+07	3.87E+06	3.87E+05	1.94E+06	1.94E+06	0	1.55E+06	1.94E+05	37.12	2073	2.86E+04	3372
1.797	2.18E+08	1.34E+08	6.04E+07	1.47E+07	1.41E+06	8.05E+05	2.62E+06	2.42E+06	0	1.01E+06	4.03E+05	209.41	1694	1.97E+04	3313
1.872	1.93E+08	1.14E+08	5.40E+07	1.18E+07	2.87E+06	1.43E+06	2.87E+06	3.41E+06	0	1.79E+06	1.79E+05	87.44	2287	2.05E+04	4702
3.068	2.65E+08	1.71E+08	6.42E+07	1.15E+07	3.63E+06	1.49E+06	2.56E+06	4.69E+06	0	5.76E+06	2.13E+05	457.11	1460	1.90E+04	1612
5.063	2.42E+08	1.33E+08	7.37E+07	1.53E+07	6.08E+06	1.94E+06	3.89E+06	3.40E+06	0	3.40E+06	9.72E+05	31.66	534	2.12E+04	1043
6.658	2.19E+08	1.27E+08	6.52E+07	1.32E+07	6.50E+06	1.52E+06	1.52E+06	2.60E+06	0	8.67E+05	0	9.58	640	1.70E+04	948
8.253	2.08E+08	1.15E+08	6.42E+07	1.71E+07	4.28E+06	6.42E+05	1.93E+06	3.00E+06	0	1.71E+06	2.14E+05	0	1580	1.94E+04	1974
8.253	2.29E+08	1.63E+08	5.60E+07	5.79E+06	1.68E+06	7.47E+05	7.47E+05	7.47E+05	0	0	0	33.89	1394	1.96E+04	419
8.253	3.09E+08	1.67E+08	1.24E+08	9.76E+06	4.07E+06	0	8.13E+05	3.25E+06	0	4.07E+05	0	63.19	1018	2.34E+04	3881
8.253	1.92E+08	1.22E+08	6.04E+07	6.02E+06	2.41E+06	6.02E+05	4.01E+05	0	2.01E+05	0	0	45.91	1771	2.33E+04	536
8.253	1.82E+08	1.02E+08	6.74E+07	8.94E+06	2.18E+06	2.18E+05	4.36E+05	8.72E+05	0	0	0	26.44	1205	1.73E+04	1129
8.253	1.92E+08	9.33E+07	7.83E+07	1.68E+07	1.81E+06	0	1.29E+06	7.75E+05	0	0	0	60.28	632	1.82E+04	2077
8.253	1.49E+08	5.01E+07	7.78E+07	1.59E+07	2.57E+06	5.13E+05	7.70E+05	1.28E+06	0	0	0	50.59	1250	1.60E+04	2523
8.253	1.58E+08	7.16E+07	6.78E+07	1.37E+07	4.21E+06	0	4.43E+05	4.43E+05	0	0	0	53.36	1199	7.15E+03	2746
9.450	1.31E+08	4.36E+07	6.02E+07	1.94E+07	4.54E+06	0	2.37E+06	0	0	1.98E+05	1.98E+05	73.16	1228	2.87E+04	8356
9.870	2.23E+08	8.47E+07	1.06E+08	2.32E+07	6.12E+06	0	0	2.26E+06	0	9.67E+05	0	153.93	1278	2.17E+04	6417
9.929	7.96E+07	1.07E+07	5.35E+07	1.19E+07	2.80E+06	3.51E+05	3.51E+05	0	0	0	0	45.28	1856	7.95E+03	2715
10.122	3.54E+07	6.19E+06	2.15E+07	6.32E+06	1.39E+06	0	0	0	0	0	0	98.37	1549	6.10E+03	5032
10.341	3.18E+07	4.93E+06	1.56E+07	9.36E+06	1.60E+06	0	2.46E+05	0	0	0	0	109.04	2032	1.65E+04	3269
10.778	0	0	0	0	0	0	0	0	0	0	0	60.20	1250	9.38E+03	1112
11.529	0	0	0	0	0	0	0	0	0	0	0	37.38	116	2.13E+03	1571
11.846	0	0	0	0	0	0	0	0	0	0	0	78.17	808	1.00E+03	3618
12.268	0	0	0	0	0	0	0	0	0	0	0	90.64	285	2.06E+03	1092
12.555	0	0	0	0	0	0	0	0	0	0	0	18.64	13	1.13E+03	28
12.799	9.29E+04	9.29E+04	0	0	0	0	0	0	0	0	0	38.18	65	3.18E+03	475
12.885	0	0	0	0	0	0	0	0	0	0	0	50.92	142	5.85E+03	232
12.999	0	0	0	0	0	0	0	0	0	0	0	91.16	0	3.47E+03	89
13.092	0	0	0	0	0	0	0	0	0	0	0	84.59	79	5.00E+03	89
13.215	0	0	0	0	0	0	0	0	0	0	0	76.80	0	1.48E+03	138
13.298	0	0	0	0	0	0	0	0	0	0	0	151.31	25	2.05E+03	171
13.423	0	0	0	0	0	0	0	0	0	0	0	124.49	0	2.23E+03	88
13.506	0	0	0	0	0	0	0	0	0	0	0	114.78	439	2.37E+03	195
13.619	0	0	0	0	0	0	0	0	0	0	0	84.53	245	1.53E+03	102
13.712	0	0	0	0	0	0	0	0	0	0	0	54.66	77	5.38E+02	174
13.835	0	0	0	0	0	0	0	0	0	0	0	110.01	0	3.15E+03	125
13.917	0	0	0	0	0	0	0	0	0	0	0	120.16	179	3.15E+03	310
14.042	0	0	0	0	0	0	0	0	0	0	0	68.56	265	2.83E+03	142
14.126	0	0	0	0	0	0	0	0	0	0	0	61.86	195	3.54E+03	90
14.252	0	0	0	0	0	0	0	0	0	0	0	61.54	49	7.93E+02	272
14.479	1.02E+06	1.27E+05	5.08E+05	1.27E+05	1.27E+05	1.27E+05	0	0	0	0	0	53	30	7.35E+02	77
14.713	0	0	0	0	0	0	0	0	0	0	0	63.22	314	1.82E+03	177
14.971	0	0	0	0	0	0	0	0	0	0	0	63.83	0	2.00E+02	101
15.215	1.74E+06	7.47E+05	6.22E+05	0	0	3.73E+05	0	0	0	0	0	79.00	332	3.60E+03	493
17.161	0	0	0	0	0	0	0	0	0	0	0	24.24	0	3.38	4
18.311	0	0	0	0	0	0	0	0	0	0	0	29.15	73	3.09E+03	0
19.300	0	0	0	0	0	0	0	0	0	0	0	51.42	164	1.10E+03	170
20.525	0	0	0	0	0	0	0	0	0	0	0	58.18	28	1.59E+04	10
22.902	0	0	0	0	0	0	0	0	0	0	0	72.58	0	3.19E+02	0
23.917	0	0	0	0	0	0	0	0	0	0	0	47.44	6	6.13E+03	339

Table S2. Concentrations of coccoliths, alkenones, foraminifers, and organic linings. The organic linings are from Gibb et al. (2014).

Age (ka)	Cocco cm ² yr ⁻¹	Cpel cm ² yr ⁻¹	Ehux cm ² yr ⁻¹	Clep cm ² yr ⁻¹	Geph cm ² yr ⁻¹	Usib cm ² yr ⁻¹	Hear cm ² yr ⁻¹	Alk. ng cm ² yr ⁻¹	PF cm ² yr ⁻¹	BF cm ² yr ⁻¹	OL cm ² yr ⁻¹
1.516	4.04E+06	2.50E+06	1.31E+06	6.54E+04	5.67E+04	1.74E+04	1.31E+04	0.45	3.47E+02	14.98	3.36
1.572	4.06E+06	2.54E+06	1.25E+06	1.29E+05	3.74E+04	2.49E+04	3.32E+04	0	1.07E+02	9.80	5.03
1.647	4.50E+06	2.83E+06	1.41E+06	1.41E+05	4.70E+04	2.35E+04	1.88E+04	1.17	1.80E+02	18.27	31.38
1.759	2.19E+06	1.31E+06	6.11E+05	1.60E+05	4.00E+04	4.00E+03	2.00E+04	0.38	2.95E+02	21.37	23.18
1.797	1.08E+06	6.63E+05	2.98E+05	7.26E+04	6.96E+03	3.98E+03	1.29E+04	1.03	9.73E+01	8.36	10.90
1.872	7.25E+05	4.31E+05	2.03E+05	4.45E+04	1.08E+04	5.40E+03	1.08E+04	0.33	7.70E+01	8.61	11.79
3.068	9.95E+05	6.42E+05	2.41E+05	4.33E+04	1.36E+04	5.61E+03	9.62E+03	1.72	7.14E+01	5.49	4.04
5.063	9.11E+05	5.02E+05	2.77E+05	5.76E+04	2.29E+04	7.32E+03	1.46E+04	0.12	7.97E+01	2.01	2.62
6.658	8.23E+05	4.79E+05	2.45E+05	4.97E+04	2.44E+04	5.70E+03	5.70E+03	0.04	6.41E+01	2.41	2.38
8.253	-	-	-	-	-	-	-	-	-	-	-
8.253	-	-	-	-	-	-	-	-	-	-	-
8.253	-	-	-	-	-	-	-	-	-	-	-
8.253	-	-	-	-	-	-	-	-	-	-	-
8.253	-	-	-	-	-	-	-	-	-	-	-
8.253	-	-	-	-	-	-	-	-	-	-	-
8.253	-	-	-	-	-	-	-	-	-	-	-
8.253	-	-	-	-	-	-	-	-	-	-	-
8.253	7.94E+05	3.59E+05	3.40E+05	6.89E+04	2.11E+04	0	2.22E+03	0.27	3.59E+01	6.01	9.18
9.450	1.86E+06	6.24E+05	8.61E+05	2.77E+05	6.49E+04	0	3.39E+04	1.05	4.11E+02	17.54	79.58
9.870	4.53E+07	1.72E+07	2.15E+07	4.72E+06	1.25E+06	0	0	31.31	4.42E+03	259.97	870.10
9.929	9.90E+06	1.33E+06	6.65E+06	1.48E+06	3.49E+05	4.36E+04	4.36E+04	5.63	9.89E+02	230.79	225.08
10.122	2.91E+06	5.09E+05	1.77E+06	5.19E+05	1.14E+05	0	0	8.09	5.02E+02	127.34	275.73
10.341	1.75E+06	2.71E+05	8.59E+05	5.14E+05	8.80E+04	0	1.35E+04	5.99	9.07E+02	111.60	119.69
10.778	0	0	0	0	0	0	0	3.73	5.80E+02	77.40	45.90
11.529	0	0	0	0	0	0	0	2.30	1.31E+02	7.11	64.43
11.846	0	0	0	0	0	0	0	4.45	5.70E+01	45.94	137.18
12.268	0	0	0	0	0	0	0	7.58	1.72E+02	23.84	60.88
12.555	0	0	0	0	0	0	0	2.29	1.39E+02	1.58	2.30
12.799	1.30E+04	1.30E+04	0	0	0	0	0	5.33	4.44E+02	9.12	44.19
12.885	0	0	0	0	0	0	0	7.37	8.46E+02	20.62	22.39
12.999	0	0	0	0	0	0	0	13.23	5.03E+02	0	8.61
13.092	0	0	0	0	0	0	0	12.38	7.32E+02	11.52	8.68
13.215	0	0	0	0	0	0	0	11.10	2.14E+02	0	13.30
13.298	0	0	0	0	0	0	0	21.79	2.96E+02	3.56	16.42
13.423	0	0	0	0	0	0	0	18.00	3.22E+02	0	8.48
13.506	0	0	0	0	0	0	0	16.76	3.46E+02	64.04	18.98
13.619	0	0	0	0	0	0	0	12.27	2.23E+02	35.62	9.87
13.712	0	0	0	0	0	0	0	8.00	7.88E+01	11.26	16.98
13.835	0	0	0	0	0	0	0	16.10	4.61E+02	0	12.20
13.917	0	0	0	0	0	0	0	17.30	4.54E+02	25.71	29.76
14.042	0	0	0	0	0	0	0	9.79	4.05E+02	37.93	13.52
14.126	0	0	0	0	0	0	0	8.84	5.06E+02	27.79	8.57
14.252	0	0	0	0	0	0	0	8.13	1.05E+02	6.45	23.96
14.479	1.24E+05	1.55E+04	6.18E+04	1.55E+04	1.55E+04	1.55E+04	0	6.43	8.95E+01	3.64	6.25
14.713	0	0	0	0	0	0	0	5.50	1.59E+02	27.36	10.27
14.971	0	0	0	0	0	0	0	5.29	1.66E+01	0	5.58
15.215	1.36E+05	5.81E+04	4.84E+04	0	0	2.91E+04	0	6.15	2.80E+02	25.88	25.59
17.161	0	0	0	0	0	0	0	1.23	1.72E-01	0	0.14
18.311	0	0	0	0	0	0	0	0.80	8.45E+01	2.00	0
19.300	0	0	0	0	0	0	0	1.26	2.70E+01	4.02	2.78
20.525	0	0	0	0	0	0	0	1.47	4.01E+02	0.72	0.17
22.902	0	0	0	0	0	0	0	2.15	9.43	0	0
23.917	0	0	0	0	0	0	0	1.40	1.81E+02	0.17	

Table S3. Fluxes of coccoliths, alkenones, foraminifers, and organic linings.

Age (ka)	<i>C. pelagicus</i> (%)	<i>E. huxleyi</i> (%)	<i>C. leptoporus</i> (%)	<i>Gephyrocapsa</i> spp. (%)	<i>U. sibogae</i> (%)	<i>H. carteri</i> (%)	<i>P. discopora</i> (%)	<i>R. clavigera</i> (%)	<i>S. pulchra</i> (%)	<i>R. producta</i> (%)
1.516	61.75	32.44	1.62	1.40	0.43	0.32	0.75	0	1.08	0.22
1.572	62.64	30.71	3.17	0.92	0.61	0.82	0.61	0	0.41	0.10
1.647	62.84	31.32	3.13	1.04	0.52	0.42	0.52	0	0	0.21
1.759	60.05	27.97	7.31	1.83	0.18	0.91	0.91	0	0.73	0.09
1.797	61.59	27.70	6.74	0.65	0.37	1.20	1.11	0	0.46	0.18
1.872	59.35	28.00	6.14	1.49	0.74	1.49	1.77	0	0.93	0.09
3.068	64.46	24.25	4.35	1.37	0.56	0.97	1.77	0	2.18	0.08
5.063	55.12	30.42	6.33	2.51	0.80	1.61	1.41	0	1.41	0.40
6.658	58.22	29.80	6.04	2.97	0.69	0.69	1.19	0	0.40	0
8.253	55.15	30.93	8.25	2.06	0.31	0.93	1.44	0	0.82	0.10
8.253	71.27	24.49	2.53	0.73	0.33	0.33	0.33	0	0	0
8.253	54.01	40.08	3.15	1.31	0	0.26	1.05	0	0.13	0
8.253	63.49	31.49	3.14	1.26	0.31	0.21	0	0.10	0	0
8.253	56.00	37.05	4.92	1.20	0.12	0.24	0.48	0	0	0
8.253	48.52	40.73	8.74	0.94	0	0.67	0.40	0	0	0
8.253	33.62	52.24	10.69	1.72	0.34	0.52	0.86	0	0	0
8.253	45.24	42.86	8.68	2.66	0	0.28	0.28	0	0	0
9.450	33.43	46.14	14.83	3.48	0	1.82	0	0	0.15	0.15
9.870	38.01	47.40	10.40	2.75	0	0	1.01	0	0.43	0
9.929	13.44	67.18	14.98	3.52	0.44	0.44	0	0	0	0
10.122	17.50	60.71	17.86	3.93	0	0	0	0	0	0
10.341	15.50	49.22	29.46	5.04	0	0.78	0	0	0	0
10.778	0	0	0	0	0	0	0	0	0	0
11.529	0	0	0	0	0	0	0	0	0	0
11.846	0	0	0	0	0	0	0	0	0	0
12.268	0	0	0	0	0	0	0	0	0	0
12.555	0	0	0	0	0	0	0	0	0	0
12.799	100.00	0	0	0	0	0	0	0	0	0
12.885	0	0	0	0	0	0	0	0	0	0
12.999	0	0	0	0	0	0	0	0	0	0
13.092	0	0	0	0	0	0	0	0	0	0
13.215	0	0	0	0	0	0	0	0	0	0
13.298	0	0	0	0	0	0	0	0	0	0
13.423	0	0	0	0	0	0	0	0	0	0
13.506	0	0	0	0	0	0	0	0	0	0
13.619	0	0	0	0	0	0	0	0	0	0
13.712	0	0	0	0	0	0	0	0	0	0
13.835	0	0	0	0	0	0	0	0	0	0
13.917	0	0	0	0	0	0	0	0	0	0
14.042	0	0	0	0	0	0	0	0	0	0
14.126	0	0	0	0	0	0	0	0	0	0
14.252	0	0	0	0	0	0	0	0	0	0
14.479	12.50	50.00	12.50	12.50	12.50	0	0	0	0	0
14.713	0	0	0	0	0	0	0	0	0	0
14.971	0	0	0	0	0	0	0	0	0	0
15.215	42.86	35.71	0	0	21.43	0	0	0	0	0
17.161	0	0	0	0	0	0	0	0	0	0
18.311	0	0	0	0	0	0	0	0	0	0
19.300	0	0	0	0	0	0	0	0	0	0
20.525	0	0	0	0	0	0	0	0	0	0
22.902	0	0	0	0	0	0	0	0	0	0
23.917	0	0	0	0	0	0	0	0	0	0

Table S4. Percentages of coccolith species in the samples.

Age (ka)	Frag idx (/8) (%)	B/P Ratio	Log (OL/B)
1.516	2.79	0.04	-0.936
1.572	5.69	0.09	-0.456
1.647	4.70	0.10	-0.080
1.759	3.02	0.07	0.132
1.797	4.22	0.09	-0.070
1.872	3.03	0.11	-0.049
3.068	3.74	0.08	-0.513
5.063	5.41	0.03	-0.089
6.658	3.56	0.04	-0.261
8.253	3.67	0.08	-0.366
8.253	4.00	0.07	-0.844
8.253	3.52	0.04	0.183
8.253	3.43	0.08	-0.899
8.253	4.53	0.07	-0.460
8.253	4.02	0.03	-0.015
8.253	2.77	0.08	-0.263
8.253	5.96	0.17	-0.102
9.450	2.14	0.04	0.342
9.870	2.77	0.06	0.269
9.929	2.88	0.23	-0.366
10.122	7.80	0.25	-0.057
10.341	4.77	0.12	-0.155
10.778	4.34	0.13	-0.595
11.529	3.91	0.05	0.702
11.846	3.36	0.81	0.289
12.268	3.47	0.14	0.203
12.555	7.61	0.01	-0.005
12.799	10.42	0.02	0.519
12.885	2.77	0.02	-0.401
12.999	5.45	0.00	-
13.092	5.49	0.02	-0.491
13.215	15.79	0.00	-
13.298	5.14	0.01	0.334
13.423	13.89	0.00	-
13.506	10.00	0.19	-0.857
13.619	6.54	0.16	-0.858
13.712	5.08	0.14	-0.269
13.835	3.45	0.00	-
13.917	1.85	0.06	-0.192
14.042	7.25	0.09	-0.652
14.126	4.90	0.05	-0.697
14.252	4.88	0.06	0.366
14.479	4.84	0.04	0.030
14.713	5.11	0.17	-0.818
14.971	5.88	0.00	-
15.215	7.06	0.09	-0.334
17.161	20.00	0.00	-
18.311	9.91	0.02	-
19.300	9.20	0.15	-0.416
20.525	7.08	0.00	-0.900
22.902	5.88	0.00	-
23.917	3.44	0.00	1.204

Table S5. The calcite dissolution indices: fragmentation index, B/P, and log(OL/B).

Age (ka)	U ^K ₃₇	U ^K ₃₇ - SST (°C)	MAT- SST (°C) lower limit	MAT- SST (°C)	MAT- SST (°C) upper limit
1.516	0.515	14.3	4.54	7.82	10.19
1.572	-	-	7.27	8.63	9.21
1.647	-	-	4.29	5.15	6.24
1.759	0.410	11.1	2.17	5.51	9.86
1.797	-	-	4.29	6.32	7.98
1.872	-	-	4.29	6.22	10.55
3.068	-	-	1.21	7.15	10.64
5.063	0.396	10.7	5.73	6.69	9.86
6.658	-	-	-0.04	3.94	6.40
8.253	-	-	4.97	6.17	7.51
8.253	-	-	3.87	6.87	10.96
8.253	0.224	5.4	5.56	7.83	14.34
8.253	-	-	3.87	7.15	13.34
8.253	0.459	12.6	-	-	-
8.253	0.340	9.0	-	-	-
8.253	0.307	8.0	-	-	-
8.253	0.230	5.6	-	-	-
9.450	0.241	6.0	2.55	6.26	13.40
9.870	0.285	7.3	10.42	12.08	13.40
9.929	0.179	4.1	4.29	9.18	13.40
10.122	0.261	6.6	7.38	10.72	13.40
10.341	0.263	6.6	7.79	12.24	15.95
10.778	0.287	7.4	7.79	11.50	12.46
11.529	0.123	2.4	5.18	9.49	12.82
11.846	-0.293	-10.2	4.27	9.67	12.46
12.268	0.004	-1.2	4.27	6.91	14.09
12.555	0.405	10.9	-0.98	-0.31	0.72
12.799	-0.319	-11.0	-1.08	-0.78	-0.58
12.885	-0.163	-6.3	-0.89	3.41	13.94
12.999	-0.180	-6.8	-0.91	1.11	5.16
13.092	-0.143	-5.7	-0.98	2.44	13.94
13.215	-0.136	-5.5	-0.98	1.12	2.92
13.298	-0.131	-5.3	-0.91	2.40	5.16
13.423	-0.147	-5.8	-0.91	1.05	5.16
13.506	-0.181	-6.8	-0.80	1.67	5.16
13.619	-0.263	-9.3	-0.98	0.59	5.16
13.712	-0.249	-8.9	-0.60	1.09	2.30
13.835	-0.271	-9.5	-0.98	0.60	2.92
13.917	-0.175	-6.6	-0.80	2.30	5.16
14.042	-0.193	-7.2	-0.98	1.02	5.16
14.126	-0.169	-6.5	-0.91	1.77	5.16
14.252	-0.227	-8.2	-0.91	3.15	5.16
14.479	-0.290	-10.1	-0.91	0.96	5.16
14.713	-0.076	-3.6	-0.80	0.24	1.26
14.971	-0.044	-2.7	-0.98	-0.23	0.72
15.215	0.092	1.4	-0.91	1.54	5.16
17.161	0.186	4.3	-0.98	0.26	1.26
18.311	0.555	15.5	0.51	2.43	3.46
19.300	-0.024	-2.1	-0.80	1.05	4.00
20.525	-0.046	-2.7	-0.98	0.26	1.26
22.902	-0.247	-8.8	-0.98	0.14	1.26
23.917	-0.313	-10.8	-0.89	2.54	12.48

Table S6. The U^K₃₇, U^K₃₇-SST (Filippova, 2016), and the MAT-SST (Gibb et al., 2014).

CHAPTER THREE

Coccolith and alkenone records from the northwest North Atlantic illustrate contrasted productivity from glacial to interglacial

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Abstract

We evaluate coccolithophore productivity since the last glaciation based on temporal variations of coccolith and alkenone concentrations and fluxes in sediment cores from the Baffin Bay (two cores), the Labrador Sea (two cores), and the Charlie-Gibbs Fracture Zone (CGFZ; one core). Coccoliths are not preserved in the Holocene sediments of Baffin Bay, a basin in which modern calcite dissolution occurs. Few coccoliths were recovered in Baffin Bay sediments younger than 18 ka but older than 10 ka and low but detectable alkenone concentrations were consistently found throughout the Baffin Bay sediment records since the last glaciation, suggesting haptophyte productivity. Coccolith fluxes from the northwest and southeast Labrador Sea records are low during the last glaciation but rise simultaneously into the early Holocene. The alkenone fluxes largely follow coccolith fluxes in the CGFZ and to a lower extent in the southeast Labrador Sea, but this is not the case in the northwest Labrador Sea where alkenone fluxes peaked from 12 to 10 ka. Whereas there is a clear relationship between coccolith and alkenone fluxes in the distal oceanic CGFZ, there is no correlation in both Labrador Sea records. With no evidence of intense calcium carbonate dissolution throughout the Labrador Sea, the lack of correlation suggests that non-calcifying haptophytes or other organisms might account for part of the alkenone fluxes. Given the regional paleoceanographic context, we hypothesize that non-calcifying alkenone-producing haptophytes might have thrived in the presence of sea ice in the subpolar Labrador Sea, for at least part of the accumulation histories.

Highlights

- Coccolith fluxes concurrently rose in the early Holocene across the NW North Atlantic.
- Far offshore in the NW North Atlantic, there is a positive correlation between *E. huxleyi* and alkenone abundances.
- Alkenones and *E. huxleyi* abundances are uncorrelated in areas influenced by sea-ice cover.

Keywords

Coccoliths, alkenones, concentrations, fluxes, productivity, Labrador Sea, Baffin Bay

1. Introduction

Coccolithophores, among the most important pelagic calcifying organisms in the ocean (Baumann et al., 2004; Hay 2004), play a leading role in the marine ecosystem as primary producers (Bown, 1998). Coccolithophores have a unique function in the marine carbon cycle with combined effects on biological carbon uptake through photosynthesis and carbonate production (Baumann et al., 2005). Their coccoliths are found in sedimentary records as far back as the Mesozoic (Bown et al., 2004) and they are valuable indicators of coccolithophore productivity and environmental changes in the surface ocean (Baumann et al., 2005). However, the preservation of coccoliths, which are composed of calcium carbonate, is impacted by dissolution (McIntyre and McIntyre, 1971; Honjo, 1975; Morse and Mackenzie, 1990).

In the marine realm, coccolithophores (haptophytes) *Emiliana huxleyi* and *Gephyrocapsa oceanica* produce biomarkers that are long-chain unsaturated ketones called alkenones (Volkman et al., 1980a, b; Marlowe et al., 1984a, b, 1990). Hence, alkenones and coccoliths are used concomitantly in this study to investigate changes in haptophyte productivity. Alkenones are most commonly used to reconstruct past sea-surface temperatures using the $U^{K_{37}}$ index (Müller et al., 1998) based on the relative abundance of di-, tri-, and tetra-unsaturated C_{37} alkenones (Brassell et al., 1986). The $U^{K_{37}}$ index is larger when there is a smaller degree of alkenone unsaturation (when there are fewer double bonds; alkenones that are synthesized at warmer temperatures; Brassell et al., 1986). The concentrations and fluxes of alkenones (with coccoliths) can help deduce variations in coccolithophore productivity, a method that has been previously applied to water column samples from offshore California (Malinverno et al., 2008). Reconstructing temporal variations of coccolithophore productivity and disentangling the effect of calcium carbonate dissolution since the last glaciation across the northwest North Atlantic was

the main goal of this present study. This study area was selected for two reasons: 1) the large regional oceanographic and hydrologic changes that took place since the last ice age (Ruddiman and McIntyre, 1981; Hillaire-Marcel et al., 1994a, b; Gibb et al., 2014; Jackson et al., 2017) and 2) the central sub-basin in this area, the Labrador Sea, is a critical area of deep-water formation for global ocean circulation. The impacts of changes in both the climate and oceanographic conditions since the last ice age on the marine carbon cycle, especially coccolithophore productivity and preservation, in the region are not well understood. One way of evaluating changes in past productivity is to investigate these over intervals characterized by extreme changes in climate conditions. The study area, currently characterized by large hydrographic gradients but high overall primary productivity (Yool et al., 2015), deserves some attention as it might have served either as a carbon sink or source, depending on the type and extent of productivity, through glacial-interglacial transitions (cf. Hillaire-Marcel et al., 1994a, b).

In this study, sediment from five cores were analyzed to determine the coccolith assemblages and abundances as well as alkenone concentrations: two cores from Baffin Bay, two from the Labrador Sea, and one from the Charlie-Gibbs Fracture Zone (CGFZ) of the central North Atlantic (Fig. 1; Table 1).

2. Regional setting

2.1 Oceanographic context

Along the southern Greenland margin, the polar and low salinity East Greenland Current and warm and saline Irminger Current (a side branch of the North Atlantic Drift) join to form the

West Greenland Current (Buch 2000; Cuny et al., 2002; Fig. 1). The West Greenland Current travels northwards along western Greenland and enters Baffin Bay via eastern Davis Strait. In northern Baffin Bay, this flow turns southwards along western Baffin Bay, incorporating Arctic waters to form the Baffin Island Current (Curry et al., 2011; Aksenov et al., 2010; Serreze et al., 2006). There is little shelf in western Baffin Bay, so the Baffin Island Current flows over the upper continental slope (Tang et al., 2004). The Baffin Island Current flows southwards into the Labrador Sea via western Davis Strait. Once in the northwest Labrador Sea, this current mixes with colder and fresher waters from the Hudson Strait to become the Labrador Current (Myers et al., 1990; Percy and Fife, 1993; Lazier and Wright, 1993) that flows through the southeast Labrador Sea and along the continental shelf of eastern Canada. Further south and east, far from shore and in the open North Atlantic, is the CGFZ. This area is a natural corridor that coincides with the northern edge of the surface subpolar front (Priede et al., 2013) and it is also where both the warm and cold limbs of the Atlantic Meridional Overturning Circulation (AMOC) cross the Mid-Atlantic Ridge (Furey et al., 2014). The surface flow of the North Atlantic Drift circulates above the CGFZ region (Breckenfelder et al., 2017; Fig. 1).

Deep-to-intermediate water formation in the Labrador Sea (Labrador Sea Water), directly influenced by the Labrador Current and the North Atlantic Drift, contributes to the North Atlantic Deep Water (NADW) of the AMOC (Yashayaev, 2007; Jones and Anderson, 2008; Yashayaev and Loder, 2009). Labrador Sea Water is formed by deep convection (overturning) following strong surface cooling during severe winters (Clarke and Gascard, 1983). It spreads throughout the deep North Atlantic (Yashayaev et al., 2007) transferring water, heat, and biogeochemical properties (e.g., oxygen and carbon) between the atmosphere and the deep ocean (Yashayaev et al., 2007).

The calcite saturation horizon ($\Omega_{\text{cal}} = 1$; the depth below which calcite begins to dissolve) in Baffin Bay is relatively shallow (1,000–1,500 m; Azetsu-Scott et al., 2010). Sharp gradients in calcite saturation (Ω_{cal}) characterize the upper continental slopes with $\Omega_{\text{cal}} > 2$ down to more than 400 m deep along the Greenland margin for the shelf-slope waters entering Baffin Bay in the east and $\Omega_{\text{cal}} < 2$ at 50 m depth for the waters exiting Baffin Bay in the west (Azetsu-Scott et al., 2010). Holocene sediments of Baffin Bay display extremely poor preservation of calcite (Aksu, 1983) and, thus, rules out a radiocarbon chronology for the Holocene (Jackson et al., 2017). The calcite saturation horizon is not reached in the Labrador Sea (even at a depth of 3,500 m; Azetsu-Scott et al., 2010) or in the deep North Atlantic until about 4,300 m (Chung et al., 2003). According to three foraminifer-based calcite dissolution indices, there has been overall good calcite preservation in the northwest Labrador Sea since the last glaciation (*Chapter Two*, Marshall et al., *Subm.*).

3. Materials and Methods

3.1 Core location and stratigraphy

We analyzed five sediment cores (Fig. 1; Table 1). SL 174 (core 174) and SL 170 (core 170) were recovered on the *RV Maria S. Merian* (MSM09/02) in Baffin Bay (Kucera et al. 2014). Though from similar latitudes, cores 174 and 170 were from western and eastern Baffin Bay, respectively (Fig. 1). Core 004 (Gibb et al., 2014, 2015; Nuttin et al., 2015; Filippova, 2016; *Chapter Two*, Marshall et al., *Subm.*) was retrieved from the northwest Labrador Sea during a *CCGS Hudson* cruise (2008-029; Campbell and de Vernal, 2009). Two other cores were

retrieved during the *CCGS Hudson* cruise 91-045: core 094 from the southeast Labrador Sea and core 080 from the Charlie-Gibbs Fracture Zone (CGFZ).

At our study sites, modern sea-surface conditions (salinity, temperature, sea-ice cover, annual productivity, and spring productivity; Table 2) help define the different environments across our study area. Spring blooms, which occur in May through June in the Labrador Sea (Balaguru et al., 2018), in response to the seasonal increases in irradiance and stratification, track poleward into summer and are one of the most characteristic features of seasonal primary productivity in the North Atlantic (Yool et al., 2015). The magnitude and timing of blooms play a fundamental role in the marine food webs and in carbon export (Cushing, 1990; Briggs et al., 2011; Cole et al., 2012). In western Baffin Bay, close to core site 174, the mean annual productivity is $67 \text{ gC m}^{-2} \text{ yr}^{-1}$ whereas the mean spring productivity is $280 \text{ mgC m}^{-2} \text{ day}^{-1}$ (Table 2). In eastern Baffin Bay, close to core site 170, the annual and spring productivity are higher, with means of $89 \text{ gC m}^{-2} \text{ yr}^{-1}$ and $336 \text{ mgC m}^{-2} \text{ day}^{-1}$, respectively. At our core site in the northwest Labrador Sea (core 004), the mean annual productivity is $137 \text{ gC m}^{-2} \text{ yr}^{-1}$ while the spring productivity is $624 \text{ mgC m}^{-2} \text{ day}^{-1}$. Mean annual productivity near our core sites in the southeast Labrador Sea (core 094) and the CGFZ (core 080) are similar to each other ($242 \text{ gC m}^{-2} \text{ yr}^{-1}$ and $225 \text{ gC m}^{-2} \text{ yr}^{-1}$, respectively), but the spring productivity is on average higher at the southeast Labrador Sea core site ($1,117 \text{ mgC m}^{-2} \text{ day}^{-1}$) than of that at the CGFZ site ($837 \text{ mgC m}^{-2} \text{ day}^{-1}$). Note that the primary productivity values incorporate all primary productivity, so values are not directly comparable to the sedimentary contents of coccoliths and alkenones.

The chronologies of the Baffin Bay cores were each based on twenty-eight Acceleration Mass Spectrometry (AMS) ^{14}C dates of mixed planktonic/benthic foraminifer assemblages (Jackson et al., 2017). However, no sediment younger than 10.6 ka for core 174 and 10.3 ka for

core 170 are dated due to the absence of foraminifers in those sediments. Accordingly, we present data from five subsamples without ages from the uppermost portions of both Baffin Bay cores, but we reasonably assume that they are of Holocene age. For core 174 (> 10.6 ka), sedimentation rates range from 3 to 260 cm kyr⁻¹. Subsample intervals (1 cm) from the top 20 cm were taken every 3 to 5 cm, and every 20 to 40 cm between 20–400 cm. For core 170 (> 10.3 ka), sedimentation rates range from 20 to 250 cm kyr⁻¹. This core was subsampled (1 cm thick) at 3 to 6 cm intervals over the top 16 cm and every 30 to 40 cm from 40 to 637 cm. We analyzed a total of nineteen subsamples from core 174 and twenty-one subsamples from core 170. The extremely variable sediment accumulation rates in Baffin Bay imply significant changes in sedimentary regimes, including mass-wasting events (cf. Jackson et al., 2017), thus preventing us from deriving accurate fluxes of coccoliths or alkenones from the estimated sediment accumulation rates.

The chronologies of cores 004, 094, and 080 were constructed with ¹⁴C dates from foraminifers using the Bacon software (Blaauw and Christen, 2011). A marine reservoir correction of 400 years and no additional correction (ΔR) for the regional air-sea reservoir difference was applied to those cores (Fig. S1; Table S1). The chronologies of Baffin Bay cores 174 (Table S2) and 170 (Table S3) were from Jackson et al. (2017) for which they applied a ΔR of 140 ± 35 years.

In core 004, thirteen ¹⁴C dates of *Neogloboquadrina pachyderma* left-coiled (Npl) populations (cf. Gibb et al., 2014) were used to set the age-depth relationship (Table S4; Fig. S1). The sedimentation rates for this core range from 3 to 136 cm kyr⁻¹, with overall higher sedimentation rates during the glacial interval and lower sedimentation rates during the Holocene. In addition, there is a noticeable slump or rapid sedimentation event around 8.5–8.2

ka (Gibb et al., 2014; Nuttin et al., 2015), which coincides with the final drainage of Lake Agassiz through Hudson Strait (Barber et al., 1999; Hillaire-Marcel et al., 2007; Lewis et al., 2012), although the slump sediments do not contain more detrital carbonate (Gibb et al., 2014; Nuttin et al., 2015) than what was observed throughout the core (e.g., Hillaire-Marcel et al., 2007; Nicholl et al., 2012). Subsamples (fifty-two, 1-cm intervals) were taken every 8–40 cm from 699 to 100 cm (23.9 to 9.9 ka) and every 4–20 cm from 100 to 0 cm (9.9 to 1.5 ka), which provides a temporal resolution of approximately two samples per thousand years.

In core 094, fifteen ^{14}C ages of planktonic foraminifers (Hillaire-Marcel et al., 1994a) define the age-depth relationship (Table S5, Figure S2). Sedimentation rates range from 6 to 28 cm kyr^{-1} for the past 25,000 years. The 1-cm thick subsamples were taken at approximately 15-cm intervals for alkenone measurements (thirty-four subsamples) and at 10-cm intervals for coccoliths (forty subsamples), which provide a temporal resolution of 1–2 samples per thousand years.

In core 080, seven ^{14}C ages of planktonic foraminifers (Levesque, 1995; Marmen, 2000) were used to develop the age-depth relationship (Table S6; Fig. S3). Sedimentation rates vary from 6 to 35 cm kyr^{-1} with maximum values recorded between 12 and 9 ka. The sampling was done at about 10-cm intervals and provides a temporal resolution of approximately one sample per thousand years.

3.2 Coccolith analyses

Subsamples were analyzed for coccoliths in cores 174 and 170 using a Scanning Electron Microscope (SEM) at the University of Bremen (Germany). In the preparation for SEM slide

examination, a total of 0.1–0.2 g of dry bulk sediment was suspended in buffered water (a 25 % ammonia solution, pH of ~ 8.5; slightly alkaline water to prevent coccolith dissolution, e.g., Weiser et al., 2016) and disaggregated in an ultrasonic bath for 30 seconds. After dilution (/100) with a rotary splitter, the suspension was filtered onto a polycarbonate membrane filter (50-mm diameter, 0.4- μm pore size). Filters were dried overnight and 1 cm^2 was cut out and mounted on an aluminium stub. The filter section was scanned at a magnification of 3000x to count and identify coccoliths over a total filter area of 0.324 mm^2 for each subsample. Coccolith identifications were made based on the Nannotax3 online database of SEM images (Young et al., 2017). Coccoliths were recorded in counts per gram (coccoliths g^{-1}) of dry sediment:

$$(1) \quad C_{\text{coccolith_SEM}} = (F \times C \times S)/(A \times W)$$

where F is the filtration area (mm^2), C is the number of coccoliths, S is the split factor, A is the counted area (mm^2), and W is the sediment weight (g).

For subsamples from the other three cores, the coccolith counts for the other cores were carried out by polarized light microscopy at UQAM/GEOTOP (Canada), as described in detail by Marshall et al. (*Subm., Chapter Two*) for core 004, Rahman and de Vernal (1994) for core 094, and Levesque (1995) for core 080. Slides were made from ca. 0.01 grams of dry sediments dispersed in petri dishes. Using a polarizing optical transmitted-light microscope at a high magnification (1250x) (de Vernal et al., 2010), up to 100 fields of view, with a diameter of 160 μm , were examined. A minimum of 300 coccoliths of *Emiliana huxleyi* per sample were counted. In the case of rare occurrences, all coccoliths in 100 fields of view were counted. The equation to calculate coccolith concentrations from polarized light microscope counting ($C_{\text{coccolith_PLM}}$; coccoliths g^{-1}) is as follows:

$$(2) \quad C_{\text{coccolith_PLM}} = [(n \times (rp)^2)/(nc \times (rc)^2)]/p_{\text{se}}$$

where n is the total number of counted coccoliths, rp is the radius of the petri dish (cm), nc is the number of optical fields (no units), rc is the radius of the optical field (cm), and p_{se} is the weight of the subsample (g) (de Vernal et al., 2010). Coccolith fluxes ($F_{\text{coccolith}}$; coccoliths $\text{cm}^{-2} \text{yr}^{-1}$), regardless of counting methodology, were computed according to:

$$(3) \quad F_{\text{coccolith}} (\text{coccoliths cm}^{-2} \text{yr}^{-1}) = C_{\text{coccolith}} \times \rho \times \text{SR}$$

where ρ is the dry sediment bulk density (g cm^{-3}) and SR is the sedimentation rate (cm yr^{-1}).

3.3 Alkenones

Alkenone concentrations (C_{alkenone}) were measured by gas chromatography (GC) with flame ionization detection (GC-FID). The Baffin Bay subsamples were analyzed at MARUM (Germany) and the Labrador Sea subsamples (cores 004 and 094) were analyzed at Dalhousie University (Canada) (Filippova, 2016; *Chapter Two*, Marshall et al., *Subm.*). Marmen (2000) determined the alkenone concentrations in core 080 using an early alkenone analytical procedure at UQAM (Canada). For the Baffin Bay and Labrador Sea subsamples, total lipids were extracted from 2 to 4 grams of dried and homogenized sediments using an Accelerated Solvent Extraction (ASE) system. Extracts were saponified and purified by elution through a silica column. The purified extracts were then analyzed by flame ionization detection (GC-FID) (limit of detection < 10 ng g^{-1}) to obtain total alkenone concentrations ($C_{37:2}$, $C_{37:3}$, and $C_{37:4}$ alkenones; ng g^{-1} ; Kienast et al., 2006, 2012). Peak areas of each alkenone component were defined based on retention times of an *Emiliania huxleyi* extract, with the limit of quantification defined as 10 times the background noise analyzed by GC-FID (Filippova et al., 2016; Filippova, 2016).

Limits of alkenone detection with GC-FID are $< 10 \text{ ng g}^{-1}$ (e.g., Kienast et al., 2006, 2012).

Alkenone fluxes for all five cores were computed according to:

$$(4) \quad F_{\text{alkenone}} = C_{\text{alkenone}} \times \rho \times \text{SR}$$

where ρ is the dry sediment bulk density (g cm^{-3}) and SR is the sedimentation rate (cm yr^{-1}).

4. Results

4.1 Coccolith concentrations and fluxes

Coccoliths are scarce in the Baffin Bay sediment subsamples (Fig. 2). In core 174, there are only a few coccoliths in four out of nineteen subsamples (Table S2). In core 170, five subsamples contain a few coccoliths while the other sixteen are barren (Table S3). The species identified in the Baffin Bay sediments include *Emiliana huxleyi*, *Calcidiscus leptoporus*, *Reticulofenestra sessilis*, *Reticulofenestra* spp., *Umbilicosphaera foliosa*, and *Umbilicosphaera sibogae*.

Sediment subsamples from core 004 of the northwest Labrador Sea contain variable concentrations of coccoliths, ranging from 0 to 3.1×10^8 coccoliths g^{-1} (Fig. 3A; Table S4). In the interval spanning 24 to 10 ka, most subsamples are barren with only three subsamples containing coccoliths with concentrations $< 1.7 \times 10^6$ coccoliths g^{-1} (Fig. 3A). The mean coccolith concentrations are $1.9 \pm 0.1 \times 10^8$ coccoliths g^{-1} in the subsamples younger than 10 ka with maximum values reached in the early Holocene. *Emiliana huxleyi* is a common species of the assemblages with percentages ranging from 24 to 52 % (Table S4). The other taxa include *Calcidiscus leptoporus*, *Coccolithus pelagicus*, *Gephyrocapsa* spp., *Helicosphaera carteri*,

Pontosphaera discopora, *Reticulofenestra producta*, *Rhabdosphaera clavigera*, *Syracosphaera pulchra*, and *Umbilicosphaera sibogae*.

In core 094, from the southeast Labrador Sea, most samples contain coccoliths (Table S5; Fig. 3B). In the interval older than 17 ka, coccolith concentrations are low and range from 0 to 1.7×10^6 coccoliths g^{-1} . They increase in younger sediments and reach maximum values $> 1.3 \times 10^7$ coccoliths g^{-1} after ~ 6.7 ka (Fig. 3B; cf. also Rahman and de Vernal, 1994; Hillaire-Marcel et al., 1994a, b). As the sedimentation rates are relatively uniform throughout this core, the fluxes fluctuate in parallel with concentrations. For the purposes of this study, we compiled the total coccoliths and *Emiliana huxleyi* counts for this core (Rahman and de Vernal, 1994). Using these data, the percentages of *Emiliana huxleyi* vary from 0 to 69 % with a mean of 16 ± 3 %.

In core 080 from the CGFZ, the most oceanic site of our study, coccolith concentrations are consistently higher than in the other study cores (Table S6; Fig. 3C). In the interval spanning 25 to 16.3 ka, concentrations are about 7.9×10^8 coccoliths g^{-1} but range from 5.5×10^9 coccoliths g^{-1} to 1.3×10^{10} coccoliths g^{-1} in the Holocene. *Emiliana huxleyi* is by far the most abundant species recorded in this core (mean of 61 ± 5 %). The concentrations and fluxes of *Emiliana huxleyi* display maxima after ~ 12 ka.

4.2 Alkenone concentrations and fluxes

In cores 174 and 170 from Baffin Bay, alkenone concentrations are all < 10 ng g^{-1} in sediments older than 10 ka. In subsamples younger than 10 ka, alkenone concentrations range up to 11 ng g^{-1} and 29 ng g^{-1} in the upper parts of cores 174 and 170, respectively (Fig. 2).

In core 004, the alkenone concentrations are generally low with a mean of $77 \pm 9 \text{ ng g}^{-1}$ but values $> 200 \text{ ng g}^{-1}$, reaching up to 457 ng g^{-1} , are recorded around 3–2 ka (Fig. 3A). The alkenone fluxes (Fig. 5) are relatively high in sediments older than 10 ka. The mean alkenone flux prior to 10 ka is $7.9 \pm 1.0 \text{ ng cm}^{-2} \text{ yr}^{-1}$ whereas it is $3.6 \pm 2.4 \text{ ng cm}^{-2} \text{ yr}^{-1}$ after 10 ka.

In core 094, alkenone concentrations increase from the base to the top of the study interval (Fig. 3B). From 25 to 12 ka, concentrations are low with some samples containing undetectable alkenones. From the early-to-late Holocene, the alkenone concentrations mostly vary from 50 to 100 ng g^{-1} . The alkenone flux prior to 10 ka averages $0.51 \pm 0.17 \text{ ng cm}^{-2} \text{ yr}^{-1}$, while after 10 ka it is $3.16 \pm 0.70 \text{ ng cm}^{-2} \text{ yr}^{-1}$. The alkenone fluxes vary concomitantly with the coccolith fluxes (Figs. 4 and 5).

In core 080, the alkenone concentrations show large amplitude variations from 0 to 1425 ng g^{-1} . In sediments older than 14 ka, concentrations vary from 0 to 934 ng g^{-1} . They increase after 14 ka with a peak of $> 1000 \text{ ng g}^{-1}$ between 9 and 6 ka. The alkenone fluxes are low in the interval older than 17 ka with values $< 0.1 \text{ ng cm}^{-2} \text{ yr}^{-1}$ and high, from 9 to 6 ka, with values up to $50 \text{ ng cm}^{-2} \text{ yr}^{-1}$. Overall, the alkenone and coccolith fluxes trend together throughout core 080 (Figs. 4 and 5).

4.3 Correlations of *Emiliania huxleyi* and alkenones

Since *Emiliania huxleyi* is the main alkenone producer in the marine realm, we attempted to correlate concentrations of *Emiliania huxleyi* and alkenones (Fig. 6). The results from core 004 do not show a significant relationship between total alkenone and *Emiliania huxleyi* coccolith concentrations ($r^2 = 4.29 \times 10^{-4}$, $p = 0.927$). Using age-depth windows to account for

the offset of subsampling for alkenones and coccoliths for core 094 (Table S7), we find that the *Emiliana huxleyi* coccolith and alkenone concentrations do not display a significant correlation either ($r^2 = 0.067$; $p = 0.256$) for core 094 (Fig. 6). Nevertheless, a statistically significant but weak positive correlation ($p < 0.05$) between total concentrations of *Emiliana huxleyi* and alkenones (Fig. 6) characterizes the subsamples from core 080 ($r^2 = 0.276$; $p = 0.007$) from the CGFZ. Correlations were not computed for the Baffin Bay cores due to the dearth of *Emiliana huxleyi*.

5. Discussion

5.1 Coccoliths and alkenones in Baffin Bay

Coccolith and alkenone concentrations are extremely low in Baffin Bay sediments, especially in western Baffin Bay. Nonetheless, the presence of rare coccoliths and alkenones in Baffin Bay sediments from the last glaciation may hint at some coccolithophore productivity in the overlying surface waters and the occurrence of short-lived sea-ice-free episodes. Lateral transport of both coccoliths and alkenones cannot, however, be disregarded. During the last glaciation, the northward subsurface flow of warmer water from the North Atlantic may have also contributed to the ventilation of the deep waters of Baffin Bay, deepening the calcite saturation horizon in Baffin Bay (e.g., de Vernal et al., 1992) and allowing for the preservation of coccoliths either produced locally or advected with the Atlantic waters. A modern analogue to the glacial Baffin Bay could be the central Arctic Ocean (e.g., Stein et al., 2015; Stein, 2019), which is characterized by perennial sea ice (Johannessen et al., 2004), very low productivity, but

good preservation of biogenic calcium carbonates (Bergsten, 1994; Jutterström and Anderson, 2005).

Baffin Bay is characterized by poor preservation of biogenic carbonates (Azetsu-Scott et al., 2010), which explains the absence of coccoliths in modern Baffin Bay sediments (Fig. 2). Baffin Bay is a nearly isolated basin where the organic carbon produced in surface waters accumulates and remineralizes at depth (Top et al., 1980; Azetsu-Scott et al., 2010). The accumulation of the resulting metabolic carbon dioxide in deep Baffin Bay decreases both the pH and carbonate ion concentrations, resulting in a shallow calcite saturation horizon (Azetsu-Scott et al., 2010). Because of the relatively shallow Davis Strait sill (~ 700 m water depth; Baker and Friedman, 1973; McLaughlin et al., 2006), only upper waters from the North Atlantic and the Labrador Sea penetrate into Baffin Bay and their lower densities prevent them from mixing with the relatively dense Baffin Bay bottom waters (Tang et al., 2004). The presence of poorly-ventilated deep waters in Baffin Bay maintains the low calcite saturation state in Baffin Bay. However, this has not always been the case because (rare) coccoliths and calcitic foraminifers are preserved in sediments that date back from the last glaciation in Baffin Bay (Aksu, 1983; Jackson et al., 2017) and in the adjacent Davis Strait (de Vernal et al., 1992), corroborating our Baffin Bay results. Persistent quasi-perennial sea-ice cover in Baffin Bay occurred since the last glaciation until about 10 ka (Gibb et al., 2015), minimizing primary productivity and organic carbon synthesis in Baffin Bay (de Vernal et al., 1992, 2005), consistent with results from Jackson et al. (2017) and Jennings et al. (2018).

The transition from ventilated waters to poorly ventilated and calcite-corrosive waters in deep Baffin Bay occurred between 14 and 10.4–9 ka (de Vernal et al., 1992; Levac et al., 2001; Sheldon et al., 2016), which corresponds also to the deglaciation of the Canadian Arctic

Archipelago (cf. Pieńkowski et al., 2012; Stokes, 2017; Dalton et al., 2020), rapid sea-level rise (Gregoire et al., 2012), and decreased regional sea-ice cover from quasi-perennial to seasonal (de Vernal et al., 2013; Gibb et al., 2015; Jennings et al., 2018).

5.2 Coccolith and alkenone production across the northwest North Atlantic

Sediments that accumulated in the northwest Labrador Sea do not appear to have been subjected to significant calcite dissolution over the last 25,000 years, as demonstrated by multi-proxy analyses of core 004 (*Chapter Two*, Marshall et al., *Subm.*). Hence, we are confident that variations in coccolith fluxes recorded in core 004 are a reliable proxy of surface coccolithophore productivity. We also assume that calcite preservation was good at core sites 094 (southeast Labrador Sea) and 080 (CGFZ) based on results of other studies of carbonate microfossils. For example, Hillaire-Marcel et al. (1994b) observed a slight but insignificant increase in calcite dissolution, based on the organic linings and benthic foraminifers dissolution index of de Vernal et al. (1992), for the early-to-mid Holocene section of core 094. While we do not have calcite dissolution indicator data for core 080 of the CGFZ, Aksu et al. (1989) reported a record of a benthic-to-planktonic foraminifer ratio (B/P) of $\ll 1$ in ODP core 646 from the northeast Labrador Sea, implying that extensive calcite dissolution did not take place in the Labrador Sea during the last 1 Ma. Planktonic foraminifer fragmentation percentages in the same core displayed a broad cyclicity from $< 10\%$ to 60% , with the highest fragmentation intervals corresponding to those of low total planktonic foraminifer abundance. Combined, these data suggest that the fluctuations in the total planktonic abundances cannot be ascribed to dissolution (Aksu et al., 1989). Whereas this information would be desirable for the CGFZ, the numerous

records from the Labrador Sea suggest that calcite preservation likely extended further out in the distal oceanic northwest North Atlantic.

Using our transect of cores, in the presumed absence of dissolution, we document evidence for an increase in coccolithophore productivity since the last glaciation through to the Holocene. Other studies have also documented a rise in coccolith fluxes over the same time period from other locations across the northwest North Atlantic. These studies include those of Rahman and de Vernal (1994), Solignac et al. (2008), Giraudeau et al. (2000), and Hillaire-Marcel et al. (1994b). Likewise, Radi and de Vernal (2008), who reconstructed past productivity using dinocyst abundances, also reported higher productivity during the early Holocene than during the last glacial maximum.

During the last glaciation, ice sheets were present on the shelves of Baffin Bay and the Labrador Sea (Dyke et al., 2002; Shaw, 2006; Lewis et al., 2012), sea-surface temperatures were very low, and sea-ice cover was close to perennial (de Vernal et al., 2005). These conditions resulted in low primary productivity (Radi and de Vernal, 2008) and are similar to those that characterized our core sites in Baffin Bay and the Labrador Sea that were close enough to the shelves to be impacted by the proximal ice sheets and dense sea-ice cover. In the CGFZ, however, core 080 was retrieved far from any proximal ice sheet influence (Fig. 1). The sea-surface reconstructions for the last glacial maximum at this site indicate close to nil sea-ice cover (de Vernal et al., 2005; Table 3).

The core sites in the Labrador Sea were characterized by dense sea-ice cover and low surface salinity during the last glacial interval and the early deglaciation (de Vernal et al., 2005; Gibb et al., 2014; Table 3). These conditions are encountered in modern, high latitude neritic environments impacted by freshwater discharge as well as polar regions, areas that are not prone

to sustain high coccolithophore productivity (O'Brien et al., 2016), but where non-calcifying haptophytes may be abundant (Liu et al., 2009; Endo et al., 2018). In contrast, the core 080 site is located offshore and remained in a fully oceanic context throughout the last 25,000 years. The lack of sea ice may explain why this is the only core in our study that displays a significant, positive correlation between *Emiliania huxleyi* and alkenone concentrations in the sediments.

Also keep in mind that alkenones from freshwater haptophytes (Schulz et al., 2000; Zink et al., 2001; Chu et al., 2005; Hou et al., 2016; Warden et al., 2016; Longo et al., 2016, 2018; Araie et al., 2018), are notably used for paleo-salinity reconstructions, as in the Baltic Sea (Warden et al., 2016), the Okhotsk Sea (Harada et al., 2008), and offshore West Greenland (Moros et al., 2016) in relation to freshwater or meltwater inputs. Terrestrial and freshwater inputs from the coast are where freshwater alkenones to the sediments can be linked to fluvial inputs (e.g., the St. Lawrence Estuary and the Laurentian Channel). Terrestrial material, including freshwater alkenones produced by non-calcifying haptophytes, may have been transported to the sea, thus impacting alkenone-derived temperature estimates, in addition to contributing to the overall alkenone contents in nearshore marine sediments.

5.3 Contrasting environmental conditions

LGM temperatures were low in the northwest and southeast Labrador Sea with mean temperatures of 1.0 ± 1.0 °C and 2.5 ± 3.5 °C, respectively (Table 3). In the LGM, among the core sites in the Labrador Sea and in the CGFZ, the mean sea-surface temperature was by far the highest at the CGFZ at 12.9 ± 2.8 °C. The mean modern sea-surface temperature for the CGFZ is nearly identical (Table 2). The salinities at the Labrador Sea and CGFZ core sites in the LGM (31 to 33) are less saline than recorded from modern historical oceanographic data (~ 34).

Differences in sea-ice cover between the LGM and the present appear to have had the biggest impact on coccolithophore productivity. Sea-ice cover for the northwest Labrador Sea was 9.2 months yr^{-1} in the LGM but the modern sea-ice cover is 0.72 months yr^{-1} . For the southeast Labrador Sea and the CGFZ, the sea-ice cover during the LGM was 8.4 and 0.5 months yr^{-1} , respectively. In contrast, both of these sites exhibit 0 months yr^{-1} of sea-ice cover today. The presence of sea ice appears to be a governing factor in inhibiting the proliferation of coccolithophores, especially in the northwest Labrador Sea.

Coccolith assemblages, just like in this study, have been used to infer past ocean conditions as their distribution in sediments is linked to the properties of surface water masses, in the Norwegian-Greenland Seas (e.g., McIntyre and Bé, 1967; Eide, 1990; Samtleben and Schröder, 1992; Baumann and Matthiessen, 1992; Levesque, 1995; Baumann et al., 1999; Boeckel and Baumann, 2004). Our results showing high abundances of *Coccolithus pelagicus* and *Emiliana huxleyi* throughout most samples in our study area are also comparable with the coccolith abundance results from the above studies from the Norwegian-Greenland Seas. Combined, these studies can inform of changes in the surface water mass properties since the last glacial period for the North Atlantic.

6. Conclusions

The complementary records of coccolith and alkenone concentrations presented in this study serve as a basis for the interpretation of past coccolithophore productivity in the northwest North Atlantic. The coccolith and alkenone abundances reveal large changes in coccolithophore productivity when the former are well preserved, i.e., not subjected to significant calcium carbonate dissolution. At the core 080 site, the fluxes of alkenones and coccoliths to the

sediment, which both increased by more than three orders of magnitude from the glacial to interglacial stages, highlight a particularly important change in productivity. In contrast, in the subpolar Baffin Bay and the Labrador Sea, where large changes in coccolith and alkenone abundances are also recorded, there is a general lack of clear relationship between their respective concentrations. In the Baffin Bay, decoupling of the two tracers likely reflect pre- or post-depositional calcium carbonate dissolution, at least for the present interglacial. In contrast, there is no evidence of significant calcium carbonate dissolution in the Labrador Sea over the last 25,000 years (Hillaire-Marcel et al., 1994b; *Chapter Two*, Marshall et al., *Subm.*). Hence, whereas coccolith data show a significant increase of coccolithophore productivity from the last glacial to the interglacial, the alkenone data indicate little change (core 094) or even a decrease in fluxes from the glacial to the interglacial (core 004). Intervals with high alkenone but low coccolith fluxes suggest that alkenone production was most likely by non-calcifying haptophytes, especially for core 004 (e.g., *Chapter Two*, Marshall et al., *Subm.*). In this study, most sediment samples containing alkenones but low or no coccoliths occur along continental margins in a polar or glacio-marine setting marked by low salinity and extensive sea-ice cover. Ruling out carbonate dissolution, we speculate that in such a hydrographic setting, alkenone fluxes might, at least in part, be accounted for by non-calcifying haptophytes or other organisms.

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Figures

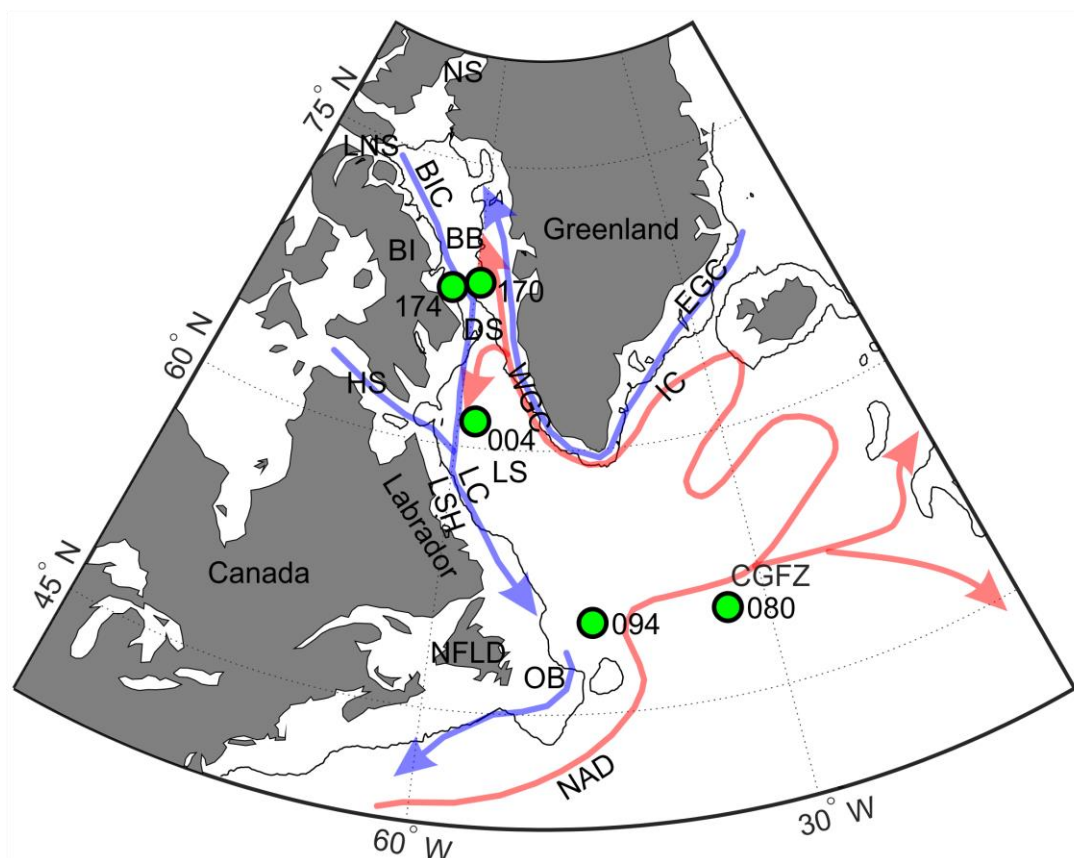


Figure 1. Map of the five core site locations and modern surface currents (blue = colder, red = warmer) of the study area. The green dots are the locations of cores for which data are presented in this study (Table 1). The solid bathymetric contour marks the 500 m isobath. Acronyms on the map include the Irminger Current (IC), East Greenland Current (EGC), West Greenland Current (WGC), Baffin Bay (BB), Baffin Island (BI), Baffin Island Current (BIC), Davis Strait (DS), Hudson Strait (HS), Labrador Sea (LS), Labrador Current (LC), Labrador Shelf (LSH), Newfoundland (NFLD), North Atlantic Drift (NAD), and Charlie Gibbs Fracture Zone (CGFZ).

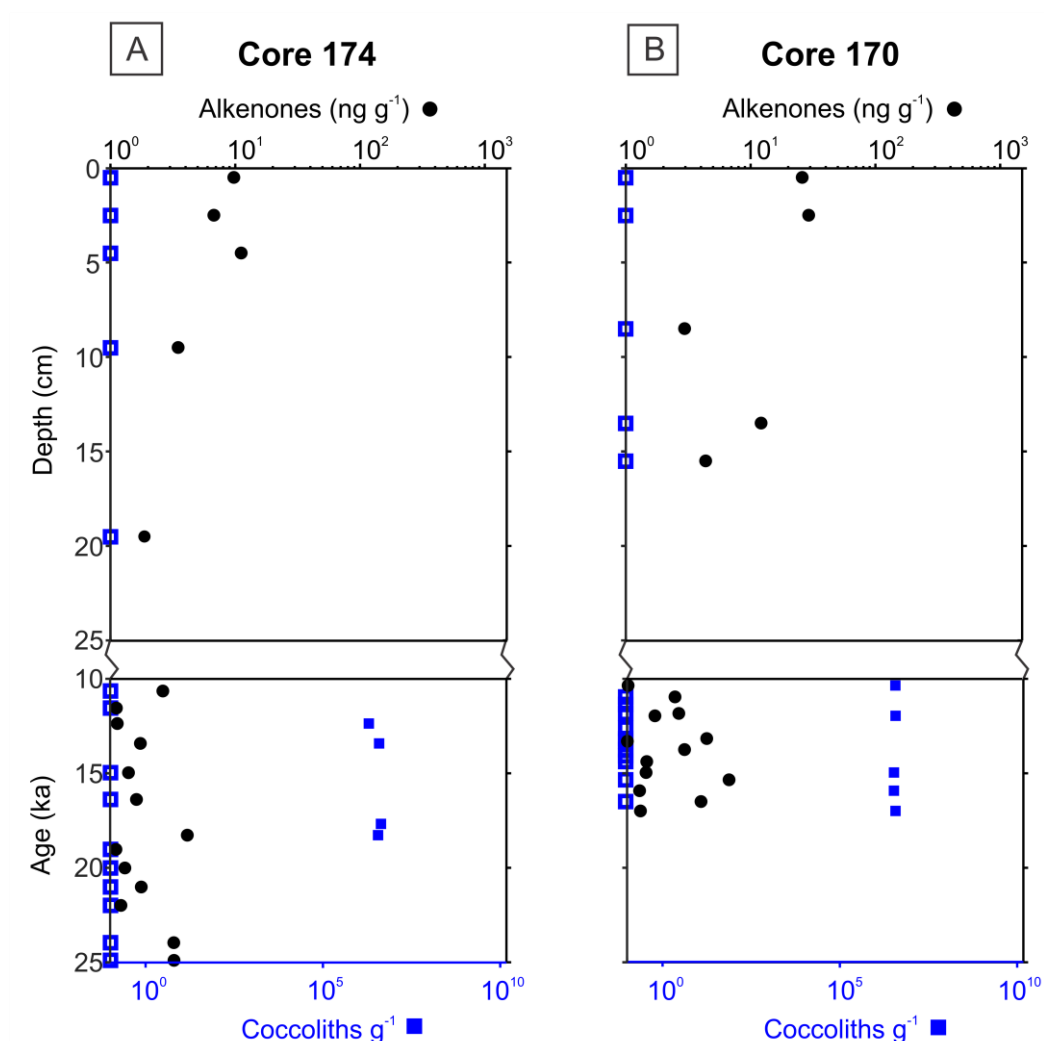


Figure 2. Concentrations of alkenones (black circles) and coccoliths (blue squares) from cores 174 (A) and 170 (B). The data from the upper five subsamples in both cores are reported on a depth scale (0–25 cm), and the data from other subsamples are reported with an age (10–25 ka; Jackson et al., 2017). Empty squares indicate subsamples without coccoliths.

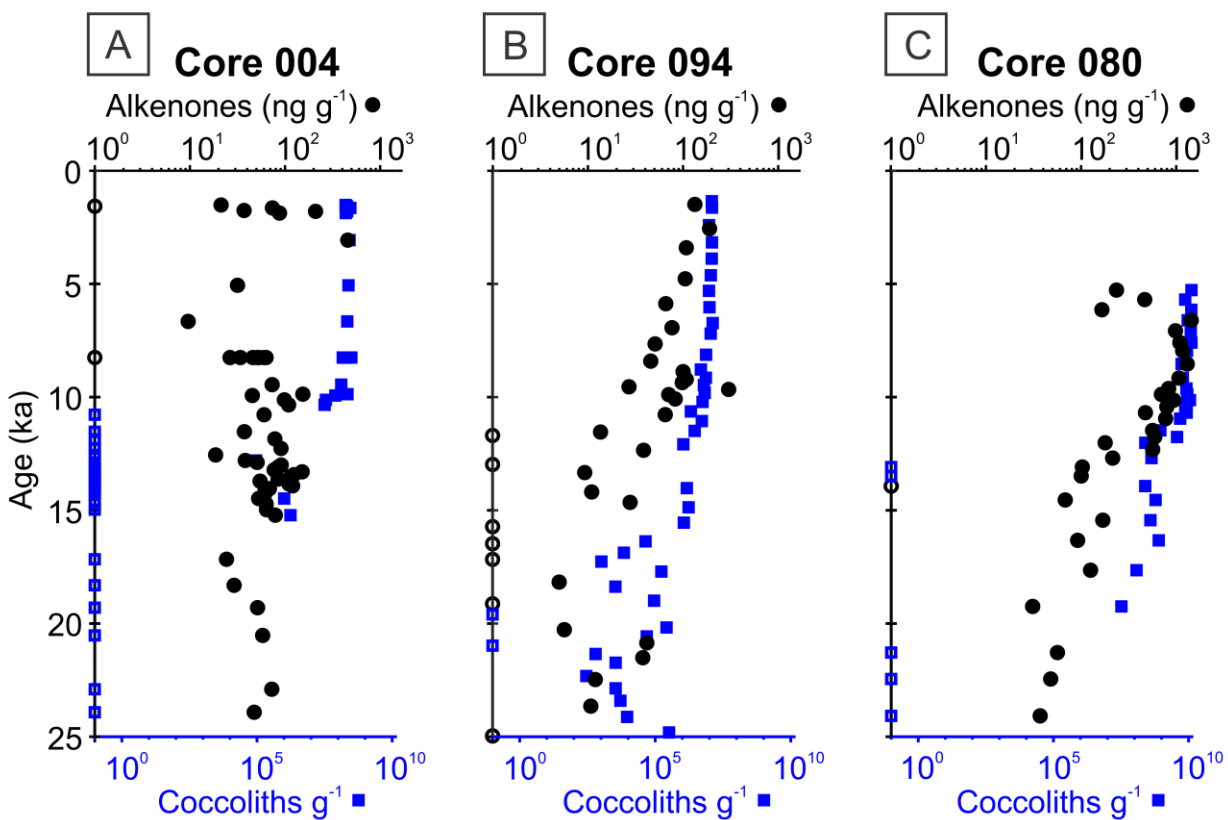


Figure 3. Concentrations of alkenones (black circles) and coccoliths (blue squares) from cores 004 (A), 094 (B), and 080 (C). Empty circles and squares indicate subsamples without alkenones and coccoliths, respectively.

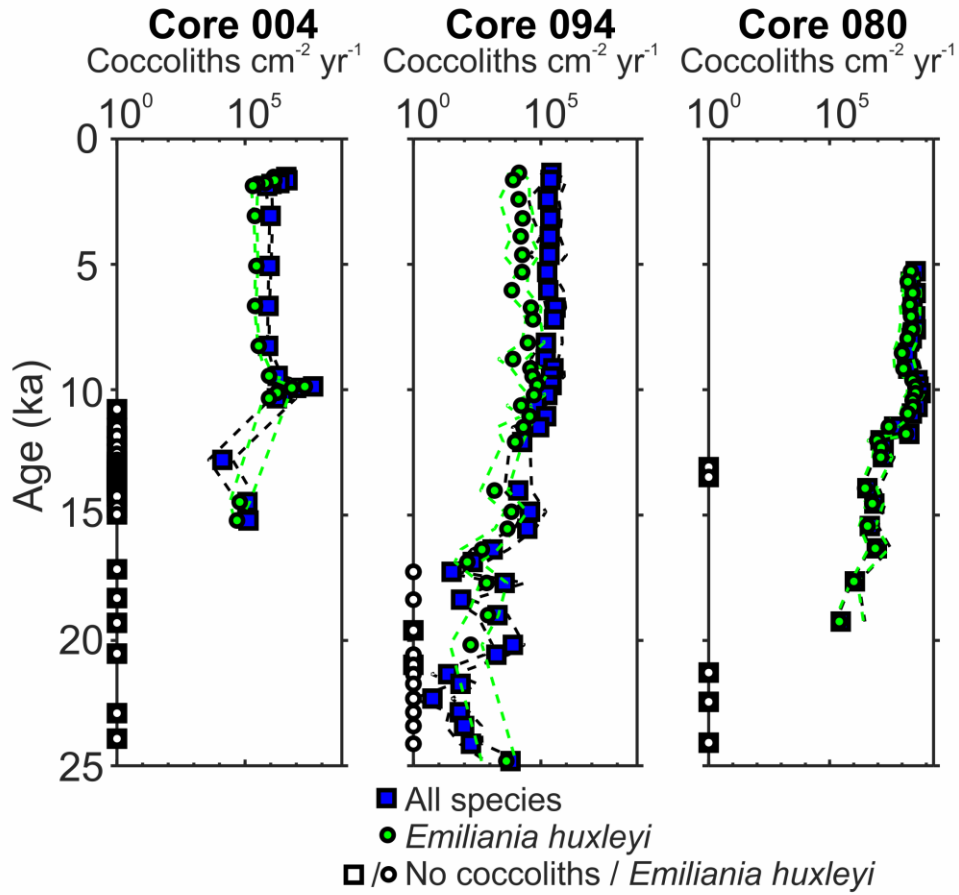


Figure 4. Fluxes (coccoliths cm⁻² yr⁻¹) of all coccoliths (blue squares) and *Emiliana huxleyi* (green circles) in cores 004, 094, and 080. Subsamples with no coccoliths or no *Emiliana huxleyi* are plotted as empty squares or circles, respectively. The black and green dotted lines indicate the range of uncertainties for, respectively, the total coccolith and *Emiliana huxleyi* fluxes.

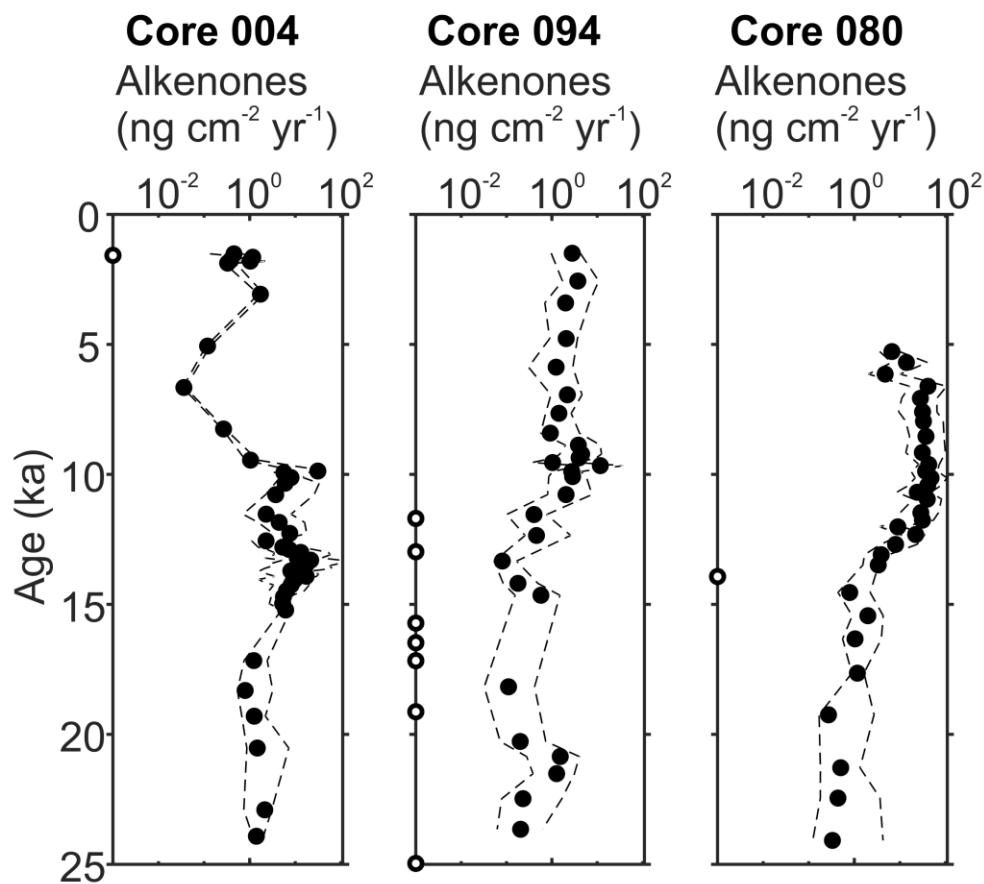


Figure 5. Alkenone fluxes (ng cm⁻² yr⁻¹) for cores 004, 094, and 080. Empty circles indicate undetectable alkenones. The range of uncertainties for the alkenone fluxes are indicated by the dashed lines.

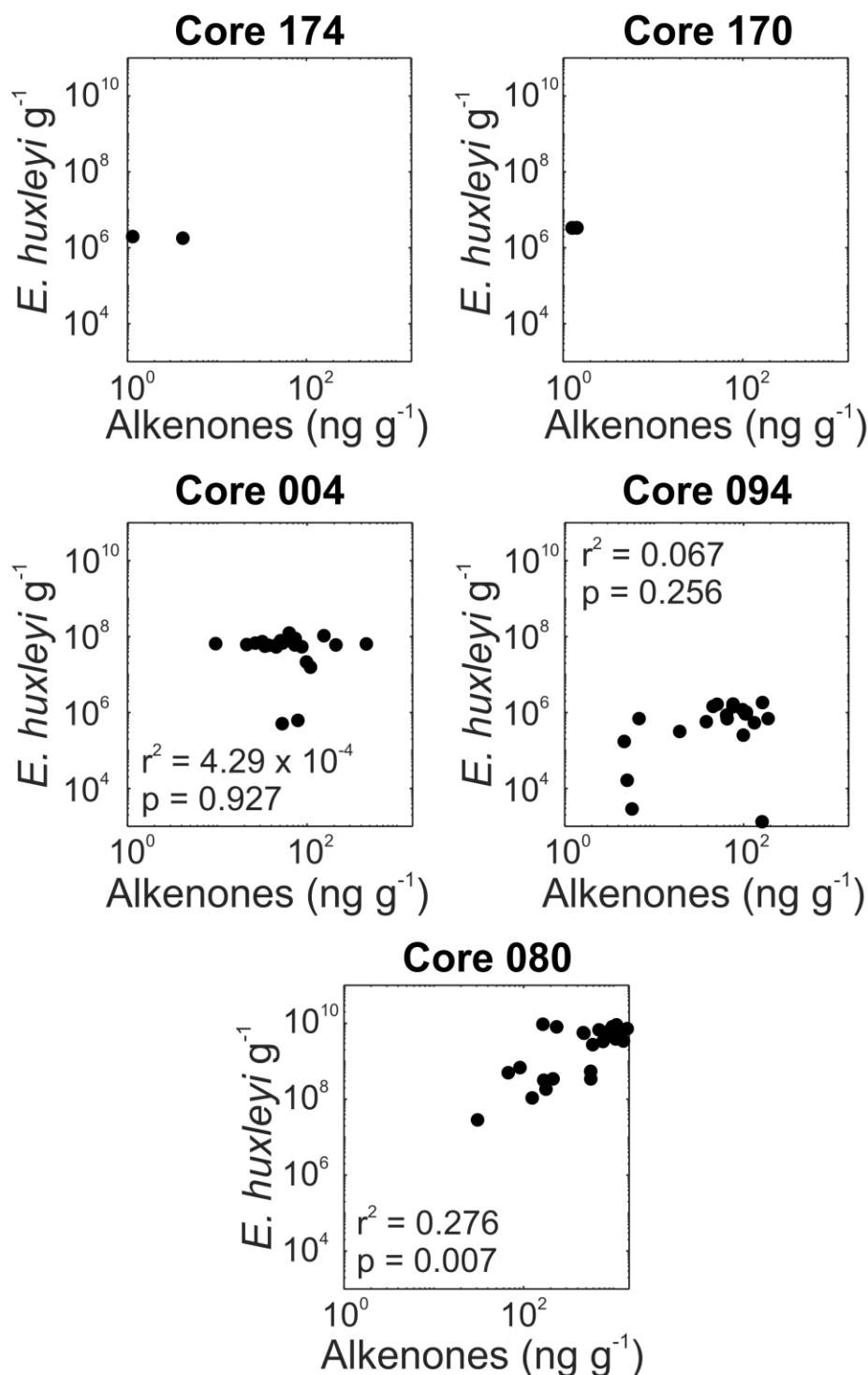


Figure 6. *Emiliania huxleyi* versus alkenone concentrations for all five cores. Core 094 was included here using the depth windows of the data (Table S7) since the sediment depths analyzed for alkenones and coccoliths were not the same. All axes are identical and on log scales (thus, only non-zero data are presented). The r^2 and p -values are based on non-zero data only. Due to a limited number of non-barren samples, r^2 and p -values were not computed for cores 174 and 170.

Tables

Core #	Cruise	Latitude (N)	Longitude (W)	Water Depth (m)	Region	Sources of the Alkenones (A) Coccoliths (C)
174	MSM09/02	68.53	-63.33	1559	Baffin Bay	This study ^{A,C}
170	MSM09/02	68.97	-59.39	1078	Baffin Bay	This study ^{A,C}
004	HU2008-029	61.46	-58.04	2674	northwest Labrador Sea	<i>Chapter Two</i> , Marshall et al., <i>Subm.</i> ^{A,C} , Filippova (2016) ^A , Filippova (2016) ^A
094	HU91-045	50.20	-45.69	3448	southeast Labrador Sea	Hillaire-Marcel et al. (1994a, b) ^C Rahman and de Vernal (1994) ^C
080	HU91-045	50.06	-33.53	3024	CGFZ/ North Atlantic	Marmen (2000) ^A Levesque (1995) ^C

Table 1. List of cores analyzed in this study, ordered by longitude (west to east).

Region	Site Latitude	Site Longitude	Summer Salinity	Summer SST	Winter SST	Sea-ice cover	Annual productivity	Spring Productivity
	°N	°W	-	°C	°C	months yr ⁻¹	gC m ⁻² yr ⁻¹	mgC m ⁻² day ⁻¹
Western Baffin Bay	68.31	-63.80	33.35	4.10	-	9.10	66.95	280.00
Eastern Baffin Bay	68.23	-57.62	32.58	3.56	-1.68	6.18	88.76	336.00
Northwest Labrador Sea	61.46	-58.04	34.39	6.20	3.73	0.72	136.78	623.67
Southeast Labrador Sea	50.21	-45.69	34.91	12.78	4.71	0.00	241.84	1117.33
CGFZ	53.06	-33.53	34.68	12.45	6.73	0.00	224.93	837.33

Table 2. Modern sea-surface conditions at/near the location of the study sites. The mean of winter (January-February-March) and summer (July-August-September) sea-surface salinities (SSS) and temperatures (SST; °C) were compiled from the 1955–2012 1/4° dataset of the World Ocean Atlas (WOA 2013 V2; cf. Locarnini et al., 2013; Zweng et al., 2013; <https://www.nodc.noaa.gov/OC5/woa13/woa13data.html>). Sea-ice cover (months yr⁻¹ with greater than 50 % sea-ice concentration) was compiled from the 1955–2012 gridded dataset of the National Snow and Ice Data Center (NSIDC V1; Walsh et al., 2015; <http://nsidc.org/data/G10010>). Annual (gC m⁻² yr⁻¹) and spring (mgC m⁻² day⁻¹) primary productivity (2002–2017) was compiled from the monthly files of Moderate Resolution Imaging Spectroradiometer (MODIS) R2018 (Behrenfeld and Falkowski, 1997; <http://orca.science.oregonstate.edu/2160.by.4320.monthly.xyz.vgpm.m.chl.m.sst.php>).

Core Site	Summer SST (°C)		Winter SST (°C)		Summer Salinity		Sea-ice cover (months yr ⁻¹)	
	Mean	std	Mean	std	Mean	std	Mean	std
Core 004	1.0	1.0	-1.3	0.5	30.9	0.3	9.2	0.7
Core 094	2.5	3.5	-0.7	1.2	31.0	1.8	8.4	3.1
Core 080	12.9	2.8	5.2	2.5	33.2	1.4	0.5	0.9

Table 3. LGM sea-surface conditions at the study sites (core 004, 094, and 080 only). The core 004 LGM data are from Gibb et al. (2014) and the LGM data for cores 094 and 080 are from de Vernal et al. (2005).

Supplemental figures and tables

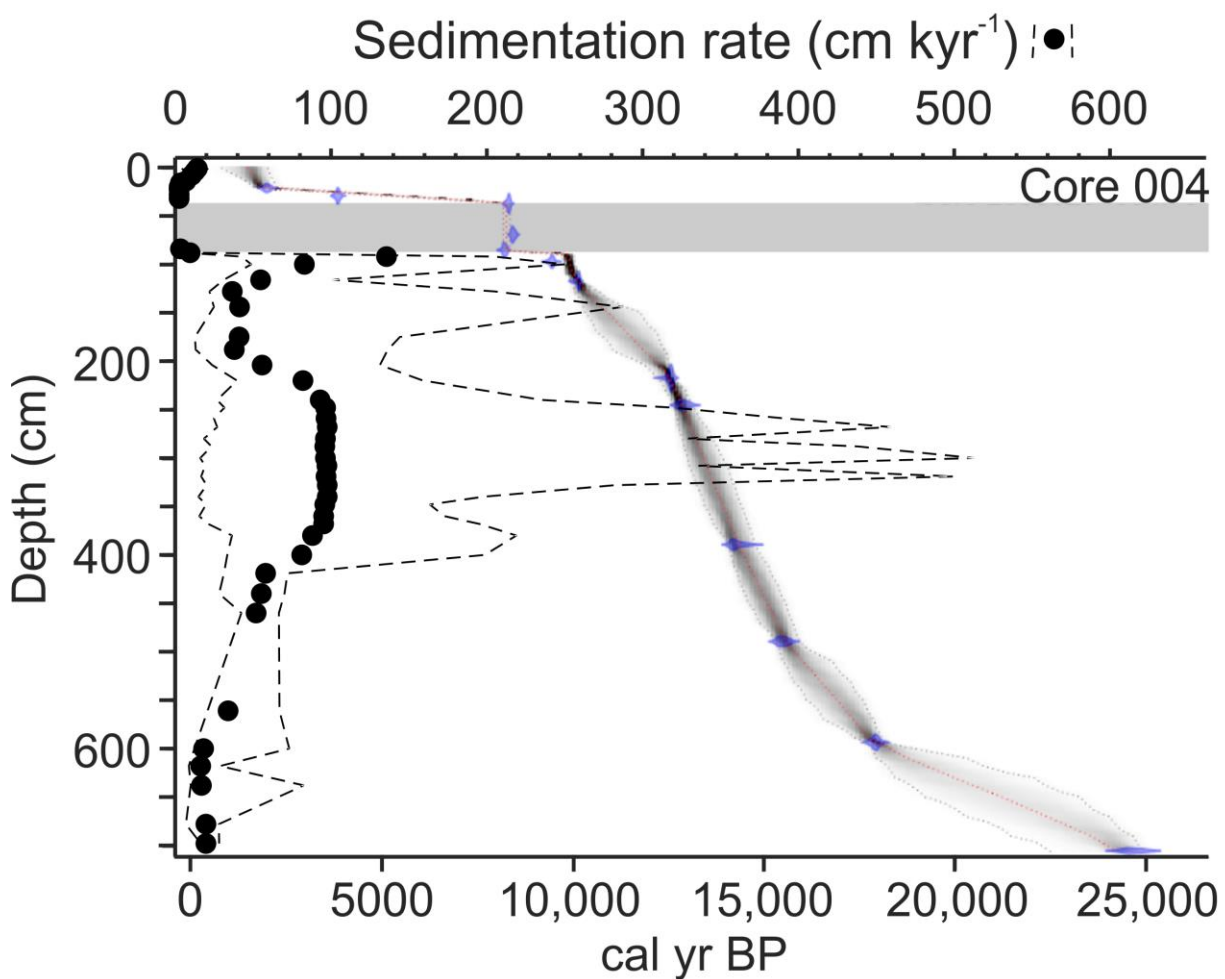


Figure S1. Age-depth relationship for core 004 based on the Bacon software (Blaauw and Christen, 2011). A slump (grey area) is identified between 36 and 85 cm. This age-depth model is modified from Gibb et al. (2014). The mean sedimentation rates (black circles; cm kyr⁻¹) with minimum and maximum sedimentation rates (dashed lines) are plotted on top of the age-depth relationship.

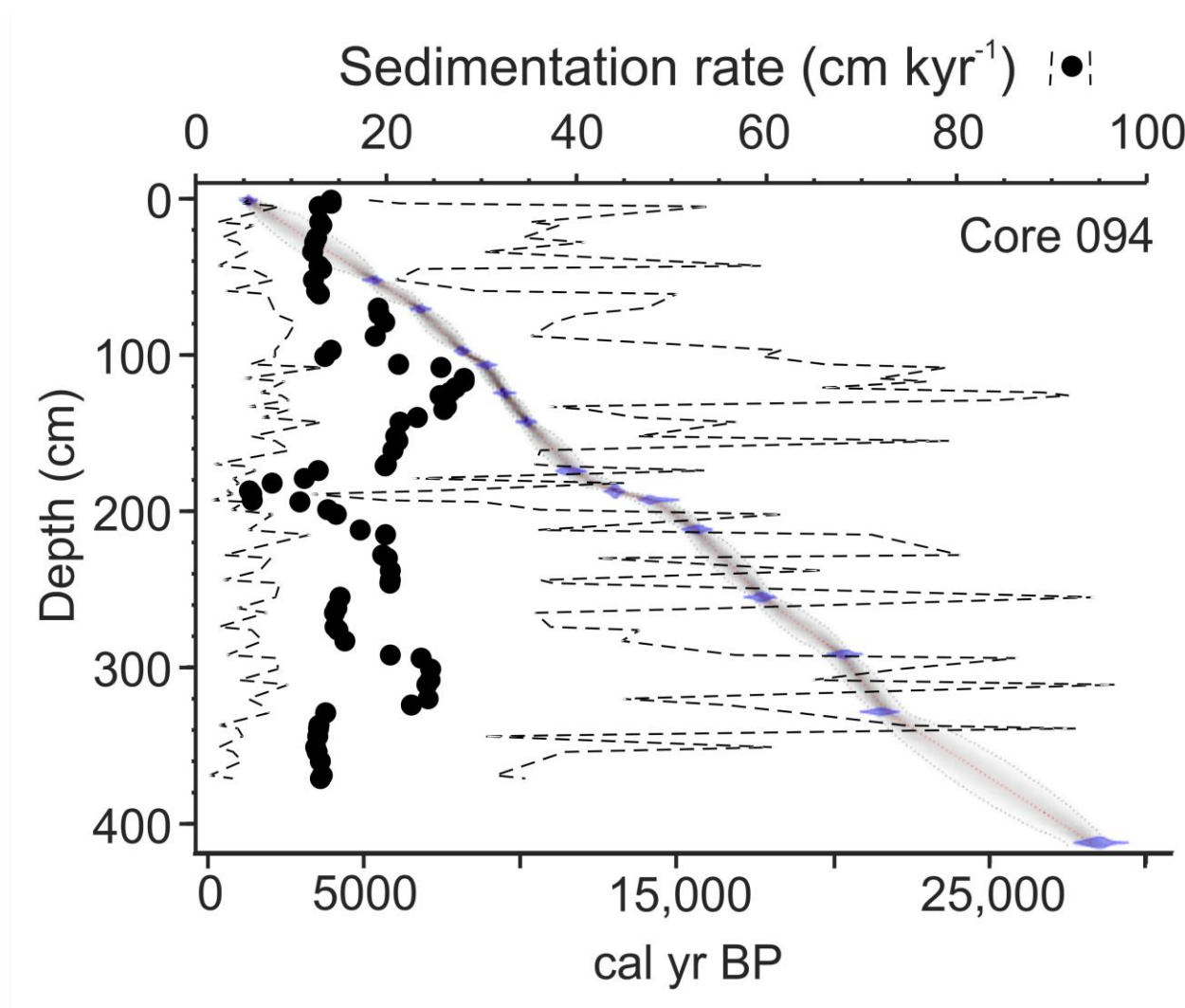


Figure S2. Age-depth relationship for core 094 based on the Bacon software (Blaauw and Christen, 2011). The mean sedimentation rates (black circles; cm kyr⁻¹) with minimum and maximum sedimentation rates (dashed lines) are plotted on top of the age-depth relationship.

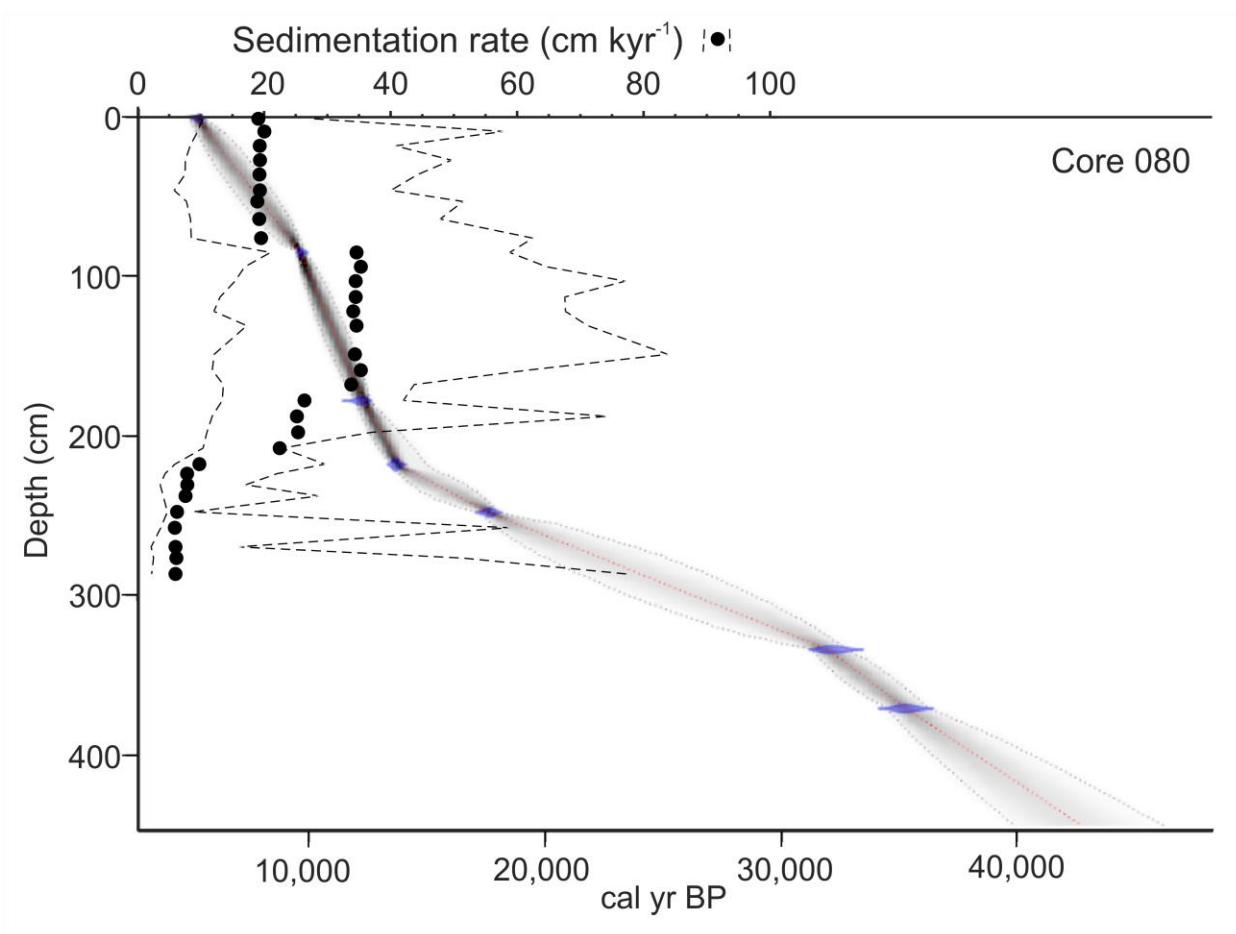


Figure S3. Age-depth relationship for core 080 based on the Bacon software (Blaauw and Christen, 2011). The mean sedimentation rates (black circles; cm kyr⁻¹) with minimum and maximum sedimentation rates (dashed lines) are plotted on top of the age-depth relationship. Only the top 300 cm were analyzed in this study (< 25,000 years) so sedimentation rates are shown only for the top 300 cm of core 080.

Core #	Depth interval (cm)	AMS- ¹⁴ C Ages (years)	Laboratory identifier	Reference
004	20–21	2370 ± 60	CAMS-153491	1
	28–29	3880 ± 25	OS-96931	1
	36–37	7845 ± 30	CAMS-155459	1
	68–69	7975 ± 35	CAMS-155461	1
	84–85	7735 ± 30	CAMS-155462	1
	96–97	8780 ± 60	CAMS-153492	1
	116–117	9290 ± 30	CAMS-155463	1
	216–217	10990 ± 45	CAMS-153493	1
	244–245	11400 ± 120	CAMS-153494	1
	388–389	12700 ± 60	CAMS-153495	1
	488–489	13360 ± 60	CAMS-153496	1
	592–593	15120 ± 60	CAMS-153497	1
	704–705	20860 ± 170	CAMS-153498	1
094	0–2	1760 ± 70	TO-3435	2, 3
	51–53	4970 ± 70	TO-8808	4
	69.5–71.5	6330 ± 80	TO-3439	2, 3
	96.5–98.5	7660 ± 70	TO-3551	2, 3
	105.5–107.5	8350 ± 60	TO-5030	4
	123.5–125.5	8810 ± 80	TO-3436	2, 3
	142–144	9360 ± 70	TO-3552	2, 3
	174–175	10420 ± 90	TO-4490	4
	187–188	11540 ± 70	TO-4146	4
	192–194	12650 ± 100	TO-3438	2, 3
	211–213	13460 ± 90	TO-3440	2, 3
	254.5–256.5	14920 ± 110	TO-3434	2, 3
	291–293	17270 ± 130	TO-3437	2, 3
	328–330	18160 ± 140	TO-3555	2, 3
	412–414	24820 ± 230	TO-3553	2, 3
080	0–2	4990 ± 70	TO-4488	4, 5, 6
	84–86	8985 ± 50	LLNL-92742	4
	177–179	10750 ± 100	TO-4337	4, 5, 6
	217–219	12210 ± 90	TO-4489	4, 5, 6
	247–249	14880 ± 130	TO-4339	4, 5, 6
	333–335	28690 ± 270	TO-4340	4, 5, 6
	370–372	31770 ± 380	TO-4338	4, 5, 6

Table S1. AMS-¹⁴C ages for cores 004, 094, and 080. For the references: 1 = Gibb et al. (2014), 2 = Hillaire-Marcel et al. (1994a), 3 = Rahman and de Vernal (1994), 4 = GEOTOP, 5 = Levesque (1995), and 6 = Marmen (2000).

Mid-depth (cm)	Min. age (ka)	Mean age (ka)	Max. age (ka)	Min. sed. rate (cm kyr ⁻¹)	Mean sed. rate (cm kyr ⁻¹)	Max. Sed. rate (cm kyr ⁻¹)	Density (g cm ⁻³)	Alkenone conc. (ng g ⁻¹)	Coccoliths g ⁻¹	<i>E. huxleyi</i> g ⁻¹
0.5	nd	nd	nd	nd	nd	nd	nd	9.74	0	0
2.5	nd	nd	nd	nd	nd	nd	nd	6.74	0	0
4.5	nd	nd	nd	nd	nd	nd	nd	11.16	0	0
9.5	nd	nd	nd	nd	nd	nd	nd	3.49	0	0
19.5	nd	nd	nd	nd	nd	nd	nd	1.87	0	0
100.5	10.314	10.645	11.003	24.29	33.21	55.01	0.586	2.64	0	0
130.5	11.223	11.549	12.020	34.97	48.89	115.24	0.586	1.12	0	0
170.5	12.097	12.367	12.567	37.84	47.55	58.75	0.586	1.14	1.98E+06	1.98E+06
220.5	13.208	13.419	13.625	17.04	19.35	22.33	0.586	1.75	3.85E+06	0
250.5	14.070	14.969	16.307	8.64	14.13	261.78	0.586	1.40	0	0
270.5	15.352	16.384	17.120	10.33	18.59	43.21	0.586	1.63	0	0
294.5	17.334	17.675	17.978	8.51	13.36	27.05	0.586	0.81	4.34E+06	0
302.5	17.554	18.274	19.634	5.43	10.63	47.95	0.586	4.15	3.58E+06	1.79E+06
310.5	17.786	19.027	21.178	4.50	10.18	36.95	0.586	1.11	0	0
320.5	18.187	20.009	22.539	3.54	9.94	28.09	0.586	1.31	0	0
330.5	18.721	21.015	23.629	3.06	10.26	90.12	0.586	1.78	0	0
340.5	19.402	21.990	24.517	4.39	10.14	78.86	0.586	1.22	0	0
360.5	21.217	23.962	25.814	2.72	10.70	56.95	0.586	3.23	0	0
370.5	22.579	24.897	26.226	3.83	10.46	25.80	0.586	3.25	0	0

Table S2. Core 174 data. The ages are from Jackson et al. (2017). Bulk dry sediment densities were not available for core 174 (Jackson et al., 2017; Jackson, 2017). Therefore, the densities were estimated to be the average of the densities from the other Baffin Bay core (core 170).

Mid-depth (cm)	Min. age (ka)	Mean age (ka)	Max. age (ka)	Min. sed. rate (cm kyr ⁻¹)	Mean sed. rate (cm kyr ⁻¹)	Max. sed. rate (cm kyr ⁻¹)	Density (g cm ⁻³)	Alkenone conc. (ng g ⁻¹)	Coccoliths g ⁻¹	<i>E. huxleyi</i> g ⁻¹
0.5	nd	nd	nd	nd	nd	nd	nd	25.90	0	0
2.5	nd	nd	nd	nd	nd	nd	nd	29.19	0	0
8.5	nd	nd	nd	nd	nd	nd	nd	2.96	0	0
13.5	nd	nd	nd	nd	nd	nd	nd	12.13	0	0
15.5	nd	nd	nd	nd	nd	nd	nd	4.36	0	0
40.5	10.127	10.353	10.561	60.19	82.78	126.01	0.625	1.02	3.68E+06	0
90.5	10.766	10.957	11.131	67.73	100.23	177.07	0.577	2.42	0	0
130.5	11.123	11.357	11.673	56.50	84.33	253.49	0.504	0.39	0	0
170.5	11.506	11.831	12.071	44.16	156.25	223.13	0.650	2.60	0	0
190.5	11.653	11.959	12.208	104.66	153.82	249.69	0.666	1.67	3.72E+06	0
290.5	12.431	12.609	12.743	136.97	181.09	239.12	0.465	0.57	0	0
390.5	12.965	13.161	13.332	58.84	139.28	145.03	0.668	4.35	0	0
410.5	13.109	13.305	13.544	62.86	90.79	198.81	0.369	1.00	0	0
450.5	13.410	13.745	14.137	45.19	91.21	206.44	0.362	2.89	0	0
480.5	13.767	14.074	14.510	32.66	65.53	111.46	0.538	0.69	0	0
500.5	14.023	14.379	14.955	32.05	51.79	128.63	0.623	1.44	0	0
530.5	14.349	14.959	15.721	20.10	51.96	249.88	0.580	1.42	3.36E+06	3.36E+06
550.5	14.625	15.344	16.136	23.14	51.93	167.13	0.601	6.56	0	0
580.5	15.141	15.921	16.657	22.16	52.32	167.21	0.596	1.26	3.35E+06	3.35E+06
610.5	15.755	16.495	17.127	21.03	52.33	167.21	0.895	3.91	0	0
636.5	16.439	16.992	17.463	21.03	37.46	167.21	0.663	1.28	3.71E+06	0

Table S3. Core 170 data. The ages and bulk dry sediment densities are from Jackson et al. (2017).

Mid-depth (cm)	Min. age (ka)	Mean age (ka)	Max age (ka)	Min. sed. rate (cm kyr ⁻¹)	Mean sed. rate (cm kyr ⁻¹)	Max. sed. rate (cm kyr ⁻¹)	Density (g cm ⁻³)	Alkenones (ng g ⁻¹)	Coccoliths g ⁻¹	<i>E. huxleyi</i> g ⁻¹	<i>E. huxleyi</i> %	Alk. flux (ng cm ⁻² yr ⁻¹)	Cocco flux (coccoliths cm ⁻² yr ⁻¹)	<i>E. huxleyi</i> flux (coccoliths cm ⁻² yr ⁻¹)
1.5	0.860	1.516	2.028	4.21	14.18	21.15	1.5	21.26	1.90E+08	6.17E+07	32.44	0.45	4.04E+06	1.31E+06
4.5	1.007	1.572	2.053	6.25	12.70	18.72	1.5	0.00	2.13E+08	6.54E+07	30.71	0.00	4.06E+06	1.25E+06
8.5	1.204	1.647	2.093	8.05	10.56	15.38	1.5	73.74	2.84E+08	8.90E+07	31.32	1.17	4.50E+06	1.41E+06
14.5	1.471	1.759	2.186	6.13	6.88	10.20	1.5	37.12	2.12E+08	5.93E+07	27.97	0.38	2.19E+06	6.11E+05
16.5	1.544	1.797	2.213	3.13	3.29	8.19	1.5	209.41	2.18E+08	6.04E+07	27.70	1.03	1.08E+06	2.98E+05
20.5	1.644	1.872	2.273	2.11	2.51	3.77	1.5	87.44	1.93E+08	5.40E+07	28.00	0.33	7.25E+05	2.03E+05
23.5	2.884	3.068	3.393	2.29	2.51	2.99	1.5	457.11	2.65E+08	6.42E+07	24.25	1.72	9.95E+05	2.41E+05
28.5	4.939	5.063	5.265	2.33	2.51	2.87	1.5	31.66	2.42E+08	7.37E+07	30.42	0.12	9.11E+05	2.77E+05
32.5	6.571	6.658	6.780	2.38	2.51	2.72	1.5	9.58	2.19E+08	6.52E+07	29.80	0.04	8.23E+05	2.45E+05
36.5	8.173	8.253	8.350	-	-	-	1.5	0.00	2.08E+08	6.42E+07	30.93	-	-	-
40.5	8.173	8.253	8.350	-	-	-	1.5	33.89	2.29E+08	5.60E+07	24.49	-	-	-
44.5	8.173	8.253	8.350	-	-	-	1.5	63.19	3.09E+08	1.24E+08	40.08	-	-	-
48.5	8.173	8.253	8.350	-	-	-	1.5	45.91	1.92E+08	6.04E+07	31.49	-	-	-
68.5	8.173	8.253	8.350	-	-	-	1.5	26.44	1.82E+08	6.74E+07	37.05	-	-	-
72.5	8.173	8.253	8.350	-	-	-	1.5	60.28	1.92E+08	7.83E+07	40.73	-	-	-
80.5	8.173	8.253	8.350	-	-	-	1.5	50.59	1.49E+08	7.78E+07	52.24	-	-	-
84.5	8.173	8.253	8.350	3.13	3.34	3.64	1.5	53.36	1.58E+08	6.78E+07	42.86	0.27	7.94E+05	3.40E+05
88.5	9.329	9.450	9.578	7.39	9.52	13.70	1.5	73.16	1.31E+08	6.02E+07	46.14	1.05	1.86E+06	8.61E+05
92.5	9.741	9.870	10.005	42.55	135.59	205.13	1.5	153.93	2.23E+08	1.06E+08	47.40	31.31	4.53E+07	2.15E+07
100.5	9.794	9.929	10.058	48.78	82.90	250.00	1.5	45.28	7.96E+07	5.35E+07	67.18	5.63	9.90E+06	6.65E+06
116.5	9.956	10.122	10.222	31.17	54.79	100.84	1.5	98.37	3.54E+07	2.15E+07	60.71	8.09	2.91E+06	1.77E+06
128.5	10.051	10.341	10.700	22.01	36.61	205.13	1.5	109.04	3.18E+07	1.56E+07	49.22	5.99	1.75E+06	8.59E+05
144.5	10.280	10.778	11.421	24.82	41.28	287.04	1.5	60.20	0.00	0.00	0.00	3.73	0.00	0.00
175.5	10.818	11.529	12.243	12.65	41.01	144.23	1.5	37.38	0.00	0.00	0.00	2.30	0.00	0.00
188.5	11.047	11.846	12.418	13.10	37.91	136.48	1.5	78.17	0.00	0.00	0.00	4.45	0.00	0.00
204.5	11.880	12.268	12.525	23.70	55.75	131.39	1.5	90.64	0.00	0.00	0.00	7.58	0.00	0.00
220.5	12.295	12.555	12.709	39.68	81.97	159.09	1.5	18.64	0.00	0.00	0.00	2.29	0.00	0.00
240.5	12.587	12.799	13.045	26.85	93.02	235.29	1.5	38.18	9.29E+04	0.00	0.00	5.33	1.30E+04	0.00
248.5	12.651	12.885	13.173	31.61	96.49	320.00	1.5	50.92	0.00	0.00	0.00	7.37	0.00	0.00
259.5	12.729	12.999	13.318	24.79	96.77	390.48	1.5	91.16	0.00	0.00	0.00	13.23	0.00	0.00
268.5	12.769	13.092	13.508	26.91	97.56	459.46	1.5	84.59	0.00	0.00	0.00	12.38	0.00	0.00
280.5	12.863	13.215	13.652	18.39	96.39	327.87	1.5	76.80	0.00	0.00	0.00	11.10	0.00	0.00
288.5	12.907	13.298	13.780	23.26	96.00	437.96	1.5	151.31	0.00	0.00	0.00	21.79	0.00	0.00
300.5	13.007	13.423	13.925	16.03	96.39	512.82	1.5	124.49	0.00	0.00	0.00	18.00	0.00	0.00
308.5	13.045	13.506	14.037	19.16	97.35	334.88	1.5	114.78	0.00	0.00	0.00	16.76	0.00	0.00
319.5	13.171	13.619	14.130	16.64	96.77	500.00	1.5	84.53	0.00	0.00	0.00	12.27	0.00	0.00
328.5	13.239	13.712	14.224	20.13	97.56	282.35	1.5	54.66	0.00	0.00	0.00	8.00	0.00	0.00
340.5	13.379	13.835	14.313	14.87	97.56	197.50	1.5	110.01	0.00	0.00	0.00	16.10	0.00	0.00
348.5	13.435	13.917	14.408	19.77	96.00	163.41	1.5	120.16	0.00	0.00	0.00	17.30	0.00	0.00
360.5	13.607	14.042	14.508	15.41	95.24	172.79	1.5	68.56	0.00	0.00	0.00	9.79	0.00	0.00
368.5	13.683	14.126	14.602	21.09	95.24	195.12	1.5	61.86	0.00	0.00	0.00	8.84	0.00	0.00
380.5	13.930	14.252	14.697	36.43	88.11	218.98	1.5	61.54	0.00	0.00	0.00	8.13	0.00	0.00
400.5	14.144	14.479	14.913	33.39	81.20	198.68	1.5	53	1.02E+06	5.08E+05	50.00	6.43	1.24E+05	6.18E+04
419.5	14.299	14.713	15.190	31.25	58.01	72.04	1.5	63.22	0.00	0.00	0.00	5.50	0.00	0.00
440.5	14.505	14.971	15.435	28.17	55.25	70.10	1.5	63.83	0.00	0.00	0.00	5.29	0.00	0.00
460.5	14.780	15.215	15.643	42.42	51.90	66.53	1.5	79.00	1.74E+06	6.22E+05	35.71	6.15	1.36E+05	4.84E+04
561.5	16.431	17.161	17.728	20.74	33.91	66.90	1.5	24.24	0.00	0.00	0.00	1.23	0.00	0.00
600.5	17.837	18.311	19.054	12.30	18.20	73.17	1.5	29.15	0.00	0.00	0.00	0.80	0.00	0.00
618.5	18.233	19.300	20.809	8.73	16.33	28.67	1.5	51.42	0.00	0.00	0.00	1.26	0.00	0.00
638.5	18.868	20.525	22.417	9.92	16.83	82.47	1.5	58.18	0.00	0.00	0.00	1.47	0.00	0.00
678.5	20.990	22.902	24.216	6.83	19.70	28.00	1.5	72.58	0.00	0.00	0.00	2.15	0.00	0.00
698.5	22.215	23.917	24.777	16.33	19.70	28.17	1.5	47.44	0.00	0.00	0.00	1.40	0.00	0.00

Table S4. Core 004 data. Bulk dry sediment densities were assumed to be 1.5 g cm⁻³ (as per Hillaire-Marcel et al., 1994a; Andrews et al., 2018).

Mid- depth (cm)	Min. Age (ka)	Mean age (ka)	Max. age (ka)	Min. sed. rate (cm kyr ⁻¹)	Mean sed. rate (cm kyr ⁻¹)	Max. sed. rate (cm kyr ⁻¹)	Density (g cm ⁻³)	Alkenones (ng g ⁻¹)	Coccoliths g ⁻¹	<i>E. huxleyi</i> g ⁻¹	Alk. flux (ng cm ⁻² yr ⁻¹)	Coccolith flux (coccoliths cm ⁻² yr ⁻¹)	<i>E. huxleyi</i> flux (coccoliths cm ⁻² yr ⁻¹)
1	1.126	1.351	1.636	5.46	14.18	18.16	1.5		1.22E+07	6.53E+05		2.60E+05	1.39E+04
3.5	1.224	1.492	1.846	4.89	14.18	21.39	1.5	132.00			2.81		
5.5	1.266	1.633	2.222	8.76	12.92	54.05	1.5		1.24E+07	4.22E+05		2.40E+05	8.19E+03
15.5	1.681	2.407	3.344	2.27	12.99	35.06	1.5		9.66E+06	6.91E+05		1.88E+05	1.35E+04
17.5	1.845	2.561	3.441	6.05	13.20	38.20	1.5	188.20			3.73		
25	2.272	3.167	4.097	2.65	12.66	34.35	1.5		1.24E+07	1.02E+06		2.35E+05	1.93E+04
28.5	2.505	3.404	4.255	4.34	12.45	40.98	1.5	107.39			2.01		
34.5	2.942	3.886	4.711	5.36	12.24	30.20	1.5		1.21E+07	8.83E+05		2.23E+05	1.62E+04
43	3.915	4.621	5.156	2.32	12.90	59.60	1.5		1.12E+07	9.65E+05		2.18E+05	1.87E+04
45.5	4.077	4.776	5.275	5.69	13.18	23.33	1.5	104.53			2.07		
52	5.015	5.307	5.541	8.14	12.32	20.96	1.5		9.60E+06	9.98E+05		1.77E+05	1.85E+04
59.5	5.390	5.875	6.355	3.11	12.66	29.49	1.5	65.44			1.24		
61	5.516	6.033	6.550	7.43	12.95	50.56	1.5		1.01E+07	3.74E+05		1.95E+05	7.27E+03
70.5	6.443	6.728	7.007	8.10	19.14	47.37	1.5		1.32E+07	1.42E+06		3.79E+05	4.08E+04
74.5	6.598	6.937	7.305	8.35	19.23	40.46	1.5				2.19		
79	6.789	7.197	7.651	10.44	19.82	37.66	1.5	76.02				3.30E+05	4.91E+04
88.5	7.192	7.651	8.042	9.61	18.83	35.23	1.5	50.56			1.43		
97.5	7.928	8.129	8.346	8.28	14.18	61.54	1.5		7.49E+06	1.45E+06		1.59E+05	3.09E+04
101.5	8.177	8.411	8.697	8.28	13.51	59.52	1.5	45.53			0.92		
106.5	8.522	8.781	9.011	5.67	21.28	66.18	1.5		4.84E+06	2.53E+05		1.54E+05	8.09E+03
108.5	8.624	8.875	9.104	13.38	25.74	78.95	1.5	99.29			3.83		
115.5	8.821	9.147	9.467	5.04	28.17	71.79	1.5		7.37E+06	9.22E+05		3.12E+05	3.89E+04
117.5	8.929	9.218	9.506	9.28	28.17	76.92	1.5	107.16			4.53		
121.5	9.093	9.360	9.632	7.96	27.27	65.57	1.5	97.39			3.98		
124.5	9.251	9.470	9.714	6.80	26.67	89.11	1.5		6.24E+06	1.19E+06		2.50E+05	4.75E+04
126.5	9.328	9.545	9.793	8.98	25.64	91.84	1.5	26.92			1.04		
129.5	9.405	9.662	9.952	9.76	26.14	83.97	1.5	298.48			11.71		
133.5	9.528	9.815	10.119	5.51	26.32	37.04	1.5		6.68E+06	1.83E+06		2.64E+05	7.22E+04
135.5	9.572	9.891	10.219	9.78	26.04	41.16	1.5	70.22			2.74		
140.5	9.779	10.083	10.373	6.93	23.26	46.33	1.5	82.16			2.87		
143	9.951	10.212	10.474	13.22	21.43	56.96	1.5		5.58E+06	1.67E+06		1.79E+05	5.37E+04
152	10.255	10.632	11.105	5.77	20.98	46.39	1.5		2.07E+06	5.38E+05		6.51E+04	1.69E+04
155.5	10.336	10.775	11.304	8.31	21.20	79.37	1.5	64.58			2.05		
161	10.581	11.058	11.587	9.87	20.69	36.22	1.5		5.23E+06	1.17E+06		1.62E+05	3.63E+04
170	10.992	11.493	12.011	1.81	20.00	35.61	1.5		2.86E+06	6.91E+05		8.57E+04	2.07E+04
171.5	11.088	11.543	12.057	4.95	19.87	37.80	1.5	13.54			0.40		
174.5	11.287	11.694	12.199	6.27	12.82	53.69	1.5	0.00			0.00		
179	11.578	12.084	12.621	3.90	11.36	22.60	1.5		1.09E+06	5.74E+05		1.86E+04	9.78E+03
182.5	11.856	12.348	12.864	4.47	7.97	45.05	1.5	38.28			0.46		
187.5	12.720	12.975	13.251	3.26	5.59	24.39	1.5	0.00			0.00		
189.5	12.980	13.333	13.687	3.85	5.82	12.01	1.5	9.25			0.08		
193	13.592	14.020	14.348	1.67	5.88	19.93	1.5		1.47E+06	1.73E+05		1.29E+04	1.52E+03
194.5	13.675	14.190	14.599	5.13	10.89	29.96	1.5	10.94			0.18		
199.5	14.060	14.649	15.185	3.72	13.82	36.11	1.5	27.65			0.57		
202.5	14.300	14.866	15.383	8.03	14.73	61.73	1.5		1.70E+06	3.17E+05		3.75E+04	7.00E+03
212	15.172	15.545	15.926	5.48	17.24	35.87	1.5		1.14E+06	1.89E+05		2.94E+04	4.89E+03
215.5	15.285	15.719	16.189	11.96	19.91	71.04	1.5	0.00			0.00		
228.5	15.790	16.372	16.967	2.92	19.61	80.40	1.5		4.38E+04	1.65E+04		1.29E+03	4.85E+02
231.5	15.851	16.474	17.112	7.84	20.10	42.09	1.5	0.00			0.00		
238	16.267	16.872	17.448	6.67	20.41	65.89	1.5		6.97E+03	4.24E+03		2.13E+02	1.30E+02
244.5	16.552	17.166	17.671	2.81	20.41	36.14	1.5	0.00			0.00		
246.5	16.681	17.264	17.743	8.78	20.36	37.56	1.5		1.03E+03	0.00		3.16E+01	0.00
255.5	17.329	17.706	18.095	8.33	15.12	94.59	1.5		1.68E+05	3.28E+04		3.81E+03	7.44E+02
262.5	17.677	18.169	18.775	4.32	14.78	55.56	1.5	4.99			0.11		
265	17.781	18.372	19.093	7.44	14.54	35.64	1.5		3.40E+03	0.00		7.42E+01	0.00
274	18.270	18.991	19.691	2.33	14.60	37.27	1.5		9.10E+04	3.85E+04		1.99E+03	8.44E+02
276.5	18.397	19.128	19.831	5.83	14.89	46.65	1.5	0.00			0.00		
283	18.884	19.598	20.166	6.98	15.63	44.78	1.5		0.00	0.00		0.00	0.00
292	19.650	20.174	20.570	3.22	20.41	56.74	1.5		2.61E+05	5.76E+03		7.99E+03	1.76E+02
294.5	19.750	20.272	20.690	8.56	23.65	86.42	1.5	5.65			0.20		
301.5	20.034	20.568	21.088	8.56	24.65	73.93	1.5		4.86E+04	0.00		1.80E+03	0.00
308.5	20.302	20.852	21.406	4.46	24.59	64.81	1.5	41.43			1.53		
311	20.414	20.974	21.544	9.67	24.26	96.77	1.5		0.00	0.00		0.00	0.00
320	20.775	21.345	21.938	5.45	24.39	44.74	1.5		6.30E+02	0.00		2.30E+01	0.00
324.5	20.997	21.509	22.085	6.82	22.62	55.79	1.5	37.57			1.28		
329	21.269	21.730	22.315	7.63	13.61	63.29	1.5		3.46E+03	0.00		7.05E+01	0.00
337	21.633	22.318	23.222	2.38	12.90	74.47	1.5		2.88E+02	0.00		5.57E+00	0.00
339.5	21.717	22.473	23.487	4.37	12.85	92.59	1.5	11.94			0.23		
344	21.972	22.862	23.981	4.87	12.77	30.12	1.5		3.46E+03	0.00		6.62E+01	0.00
351	22.380	23.410	24.633	2.36	12.55	60.79	1.5		5.18E+03	0.00		9.76E+01	0.00
354.5	22.571	23.649	24.879	3.87	12.71	38.77	1.5	10.71			0.20		
360	22.944	24.121	25.445	4.82	13.04	35.86	1.5		9.22E+03	0.00		1.80E+02	0.00
369	23.550	24.811	26.091	1.42	13.25	31.43	1.5		3.24E+05	2.23E+05		6.43E+03	4.43E+03
371.5	23.681	24.962	26.231	3.85	13.06	34.62	1.5	0.00			0.00		

Table S5. Core 094 data. Bulk dry sediment densities were assumed to be 1.5 g cm⁻³ (as per Hillaire-Marcel et al., 1994a; Andrews et al., 2018).

Mid-depth (cm)	Min. age (ka)	Mean age (ka)	Max. age (ka)	Min. sed. rate (cm kyr ⁻¹)	Mean sed. rate (cm kyr ⁻¹)	Max. sed. rate (cm kyr ⁻¹)	Density (g cm ⁻³)	Alkenones (ng g ⁻¹)	Coccoliths g ⁻¹	<i>E. huxleyi</i> g ⁻¹	Alkenone flux (ng cm ⁻² yr ⁻¹)	Coccolith flux (coccoliths cm ⁻² yr ⁻¹)	<i>E. huxleyi</i> flux (coccoliths cm ⁻² yr ⁻¹)
1	4.917	5.275	5.534	10.30	19.09	27.91	1.5	233.6	1.28E+10	8.05E+09	6.690	3.67E+08	2.31E+08
9	5.195	5.694	6.298	9.49	20.04	57.88	1.5	462.5	7.51E+09	5.69E+09	13.906	2.26E+08	1.71E+08
18	5.526	6.143	6.906	8.31	19.31	40.76	1.5	164.1	1.26E+10	9.45E+09	4.754	3.65E+08	2.74E+08
27	5.883	6.609	7.431	7.56	19.35	49.62	1.5	1425.7	9.29E+09	7.23E+09	41.391	2.70E+08	2.10E+08
36	6.261	7.074	7.904	7.51	19.27	44.30	1.5	974.5	1.21E+10	8.08E+09	28.165	3.50E+08	2.34E+08
46	6.746	7.593	8.411	5.79	19.34	40.05	1.5	1082.9	1.28E+10	9.09E+09	31.410	3.71E+08	2.64E+08
53	7.113	7.955	8.713	7.73	18.93	51.45	1.5	1158.9	8.80E+09	5.91E+09	32.912	2.50E+08	1.68E+08
64	7.725	8.536	9.182	8.36	19.23	47.84	1.5	1293.9	5.54E+09	3.42E+09	37.324	1.60E+08	9.86E+07
76	8.561	9.160	9.593	8.49	19.52	62.50	1.5	1060.8	6.13E+09	3.87E+09	31.065	1.80E+08	1.13E+08
85	9.453	9.621	9.830	21.03	34.62	58.82	1.5	825.7	8.34E+09	5.18E+09	42.873	4.33E+08	2.69E+08
94	9.605	9.881	10.254	16.95	35.29	64.67	1.5	692.8	8.99E+09	6.74E+09	36.678	4.76E+08	3.57E+08
103	9.777	10.136	10.588	15.41	34.48	77.13	1.5	933.7	1.13E+10	6.93E+09	48.295	5.84E+08	3.58E+08
113	9.997	10.426	10.938	13.04	34.48	67.54	1.5	789.6	7.57E+09	5.28E+09	40.841	3.92E+08	2.73E+08
122	10.205	10.687	11.215	12.06	34.09	67.64	1.5	470.3	8.52E+09	5.48E+09	24.049	4.36E+08	2.80E+08
131	10.429	10.951	11.497	17.27	34.62	71.15	1.5	767.1	4.90E+09	3.32E+09	39.830	2.54E+08	1.72E+08
149	10.936	11.471	11.967	12.11	34.36	83.82	1.5	559.3	9.05E+08	5.44E+08	28.830	4.66E+07	2.80E+07
159	11.255	11.762	12.221	11.81	35.29	61.57	1.5	589.1	3.75E+09	2.74E+09	31.188	1.99E+08	1.45E+08
168	11.576	12.017	12.403	13.57	33.78	43.73	1.5	177.6	2.52E+08	1.84E+08	9.000	1.28E+07	9.32E+06
178	11.945	12.313	12.613	13.39	26.39	42.02	1.5	560.6	4.36E+08	3.43E+08	22.187	1.73E+07	1.36E+07
188	12.241	12.692	13.214	11.79	25.19	74.35	1.5	212.9	4.25E+08	3.44E+08	8.044	1.61E+07	1.30E+07
198	12.571	13.089	13.839	10.96	25.38	36.98	1.5	102.1	0.00	0.00	3.887	0.00	0.00
208	12.961	13.483	14.428	10.34	22.47	22.82	1.5	99.4	0.00	0.00	3.351	0.00	0.00
218	13.529	13.928	14.997	5.92	9.77	29.61	1.5	0.0	2.50E+08	2.01E+08	0.000	3.66E+06	2.95E+06
224	13.823	14.542	15.691	4.34	7.83	22.05	1.5	67.5	6.03E+08	4.99E+08	0.793	7.08E+06	5.86E+06
231	14.370	15.436	16.650	3.58	7.87	17.09	1.5	167.7	3.89E+08	3.16E+08	1.978	4.59E+06	3.73E+06
238	15.175	16.326	17.296	4.05	7.58	28.65	1.5	91.2	7.89E+08	6.81E+08	1.037	8.97E+06	7.74E+06
248	17.124	17.645	18.101	4.71	6.25	8.73	1.5	124.7	1.19E+08	1.08E+08	1.168	1.11E+06	1.01E+06
258	18.008	19.246	21.078	3.67	5.89	58.82	1.5	30.7	3.28E+07	2.86E+07	0.271	2.90E+05	2.53E+05
270	19.193	21.282	23.946	2.15	6.00	15.92	1.5	55.9	0.00	0.00	0.503	0.00	0.00
277	20.116	22.449	25.209	2.52	6.14	51.35	1.5	47.5	0.00	0.00	0.437	0.00	0.00
287	21.539	24.078	26.873	2.23	6.00	77.25	1.5	36.8	0.00	0.00	0.331	0.00	0.00

Table S6. Core 080 data. Bulk dry sediment densities were assumed to be 1.5 g cm⁻³ (as per Hillaire-Marcel et al., 1994a; Andrews et al., 2018).

Mid-depth (cm)	Mean age (ka)	Alkenones (ng g ⁻¹)	<i>E. huxleyi</i> g ⁻¹	Mean alkenone conc. (ng g ⁻¹)	Mean <i>E. huxleyi</i> g ⁻¹	Depth-window (cm)	Alkenone conc. (ng g ⁻¹) used for x-y plot	<i>E. huxleyi</i> g ⁻¹ used for x-y plot
1	1.351		6.53E+05		5.38E+05	1-5.5	132.00	5.38E+05
3.5	1.492	132.00				15.5-17.5	188.20	6.91E+05
5.5	1.633		4.22E+05			25-28.5	107.39	1.02E+06
15.5	2.407		6.91E+05			34.5-45.5	104.53	9.24E+05
17.5	2.561	188.20				52-61	65.44	6.86E+05
25	3.167		1.02E+06			70.5-74.5	76.02	1.42E+06
28.5	3.404	107.39				79-88.5	50.56	1.65E+06
34.5	3.886		8.83E+05		9.24E+05	97.5-101.5	45.53	1.45E+06
43	4.621		9.65E+05			106.5-108.5	99.29	2.53E+05
45.5	4.776	104.53				115.5-117.5	107.16	9.22E+05
52	5.307		9.98E+05		6.86E+05	121.5-124.5	97.39	1.19E+06
59.5	5.875	65.44				126.5-133.5	162.70	1.83E+06
61	6.033		3.74E+05			135.5-143	76.19	1.67E+06
70.5	6.728		1.42E+06			152-161	64.58	8.54E+05
74.5	6.937	76.02				170-174.5	6.77	6.91E+05
79	7.197		1.65E+06			179-182.5	38.28	5.74E+05
88.5	7.651	50.56				187.5-193	4.62	1.73E+05
97.5	8.129		1.45E+06			194.5-202.5	19.30	3.17E+05
101.5	8.411	45.53				212-215.5	0.00	1.89E+05
106.5	8.781		2.53E+05			228.5-231.5	0.00	1.65E+04
108.5	8.875	99.29				238-244.5	0.00	4.24E+03
115.5	9.147		9.22E+05			246.5-262.5	4.99	1.64E+04
117.5	9.218	107.16				265-276.5	0.00	1.93E+04
121.5	9.360	97.39				283-294.5	5.65	2.88E+03
124.5	9.470		1.19E+06			301.5-311	41.43	0.00E+00
126.5	9.545	26.92		162.6987777		320-329	37.57	0.00E+00
129.5	9.662	298.48				337-344	11.94	0.00E+00
133.5	9.815		1.83E+06			351-360	10.71	0.00E+00
135.5	9.891	70.22		76.1914937		369-371.5	0.00	2.23E+05
140.5	10.083	82.16						
143	10.212		1.67E+06					
152	10.632		5.38E+05		8.54E+05			
155.5	10.775	64.58						
161	11.058		1.17E+06					
170	11.493		6.91E+05					
171.5	11.543	13.54		6.771380612				
174.5	11.694	0.00						
179	12.084		5.74E+05					
182.5	12.348	38.28						
187.5	12.975	0.00		4.622983051				
189.5	13.333	9.25						
193	14.020		1.73E+05					
194.5	14.190	10.94		19.29523401				
199.5	14.649	27.65						
202.5	14.866		3.17E+05					
212	15.545		1.89E+05					
215.5	15.719	0.00						
228.5	16.372		1.65E+04					
231.5	16.474	0.00						
238	16.872		4.24E+03					
244.5	17.166	0.00						
246.5	17.264		0.00		1.64E+04			
255.5	17.706		3.28E+04					
262.5	18.169	4.99						
265	18.372		0.00		1.93E+04			
274	18.991		3.85E+04					
276.5	19.128	0.00						
283	19.598		0.00		2.88E+03			
292	20.174		5.76E+03					
294.5	20.272	5.65						
301.5	20.568		0.00		0.00E+00			
308.5	20.852	41.43						
311	20.974		0.00					
320	21.345		0.00		0.00E+00			
324.5	21.509	37.57						
329	21.730		0.00					
337	22.318		0.00		0.00E+00			
339.5	22.473	11.94						
344	22.862		0.00					
351	23.410		0.00		0.00E+00			
354.5	23.649	10.71						
360	24.121		0.00					
369	24.811		2.23E+05					
371.5	24.962	0.00						

Table S7. Age-depth windows for core 094 that were used to create the x-y plot of total concentrations of alkenones versus total concentrations of *Emiliania huxleyi*.

SUMMARY & CONCLUSIONS

This thesis focused on the reconstruction of coccolith and alkenone productivity through time from the analyses of sediment cores collected in the northwest North Atlantic under a wide range of environmental conditions, from glacial to interglacial, and from cool temperate to Arctic settings. The sediments analyzed were from as far north as the Baffin Bay, as far south as the Scotian Slope offshore Nova Scotia, and as far east as the Charlie-Gibbs Fracture Zone. This thesis focused on biogenic carbonate production and preservation in an area where biogenic remains from haptophytes (coccoliths and alkenones) have rarely been used for paleoceanographic reconstructions of the last glacial stage (e.g., Waelbroeck et al., 2009) and the Holocene (e.g., Marcott et al., 2013; Kaufman et al., 2020) despite their potential as proxies of primary productivity (e.g., Rosell-Melé and McClymont, 2007; Giraudeau and Beaufort, 2007). The application of coccolith proxies to high-latitude sediments has often been overlooked because the species diversity is typically low and preservation (i.e., calcite dissolution) can be an issue (Gard and Backman, 1990; Baumann and Matthiessen, 1992; Baumann, 1995; Andruleit, 1997; Jackson et al., 2017). Moreover, whereas alkenones are one of the most commonly used sea-surface temperature proxies, their interpretation in high-latitude environments, where low salinity and seasonal sea-ice cover are common features, is often equivocal (e.g., Rosell-Melé, 1998; Moros et al., 2016). From this point of view, this thesis project was a challenge.

Results from this thesis expand upon previously published studies of alkenones and coccoliths in mid-high latitude sediments of the North Atlantic. Among the main findings of this thesis, I complemented the areal distribution of coccoliths in surface sediment samples from previous studies (McIntyre and Bé, 1967; Baumann and Matthiessen, 1992; Baumann, 1995; Baumann et al., 2005; Solignac et al., 2008) and demonstrated that coccolith concentrations and

fluxes from coastal to offshore settings as well as the species distribution are closely related to sea-surface temperatures. Thus, the coccolith assemblage data may serve as an excellent sea-surface temperature proxy, even in offshore settings of subpolar seas. However, I did not explore this possibility further in this thesis.

Within this thesis, I have also shown that whereas biogenic carbonate dissolution impedes the interpretation of the sediment record on the shelves (Mucci et al., 2000) and in the semi-enclosed Baffin Bay (Aksu, 1983; Jackson et al., 2017), biogenic carbonates are overall well preserved in the offshore Labrador Sea since the last glacial period, except perhaps during short episodes between 12.2 and 8.3 ka, when biogenic productivity increased rapidly (e.g., Hillaire-Marcel et al., 1994a, b; Gibb et al., 2014), fragmentation was slightly elevated, and ice-rafted debris was lower compared to the glacial interval. Hence, in the Labrador Sea and the northwest North Atlantic, the data served to show that coccolithophore productivity was much lower during the last glacial stage than during the present interglacial, by at least two orders of magnitude. This may reflect lower primary productivity in general during the last glacial stage, as suggested by the dinocyst (Radi and de Vernal, 2008a, b; Gibb et al., 2014) and foraminifer (Hillaire-Marcel et al., 1994a, b) sediment records. By combining results of both alkenone and coccolith abundance measurements, I was led to propose that, in the Labrador Sea and Baffin Bay, different primary producers may have dominated in the glacial and interglacial period. More specifically, non-calcifying alkenone-producing haptophytes may have accounted for an important component of the biogenic productivity during the last glacial stage, a hypothesis that is consistent with the abundance of haptophyte pigments in modern subpolar waters (cf. Liu et al., 2009). Therefore, this may imply that calcifying haptophytes dominate over non-calcifying haptophytes during interglacials, at least in the northwest Labrador Sea. Hence, the magnitude

and species composition of haptophyte productivity as well as the quantitative carbon fluxes have drastically changed from glacial to interglacial. This deserves more attention, especially for Earth system modelling in the context of global climate change.

In this study, I have shown that alkenones are found at detectable concentrations in sediments at nearly all the study sites, at the surface and downcore. In the most offshore and temperate settings, concentrations are high ($> 100 \text{ ng g}^{-1}$) and vary concomitantly with coccoliths, highlighting the close association between these by-products of haptophyte production. However, there is a strong decoupling between coccolith and alkenone abundances in sediments recovered at more northern locations, sites that are characterized by the presence of seasonal sea ice. My results show that in sea-ice environments and nearshore settings, the equations commonly used to relate sea-surface temperatures to alkenone unsaturation do not readily apply. The discontinuity in the alkenone-temperature relationship at high-latitudes (Conte et al., 2006) and my results suggest that alkenones in sediments that accumulated in Baffin Bay and the northwest Labrador Sea during the last glaciation may not have been produced by calcifying *Emiliania huxleyi*, but by non-calcifying haptophytes, although this hypothesis does remain speculative.

Whereas the use of alkenones as a temperature proxy may be equivocal in subpolar basins, as I have shown and as suggested by Sikes et al. (1997), Bendle and Rosell-Melé (2004), and Conte et al. (2006), their concentrations and fluxes remain useful indicators of productivity and they deserve more attention. The presence of alkenones in the absence of coccoliths in an environment not subjected to calcite dissolution can still provide useful information about productivity as haptophytes other than calcifying coccolithophores produce alkenones. Based on satellite observations and plankton studies in the modern ocean, Liu et al. (2009) and Endo et al.

(2018) have also concluded that haptophytes in the marine realm, even in the subarctic open ocean, are dominated by non-calcifying haptophytes. Likewise, studies conducted in nearshore environments have shown that some freshwater non-calcifying haptophytes produce alkenones (Schulz et al., 2000; Zink et al., 2001; Chu et al., 2005; Randlett et al., 2014; Hou et al., 2016; Warden et al., 2016).

Beyond uncertainties inherent in the use of marine sediment proxies (dating, vertical vs. lateral transport, preservation), this thesis provides clues to the usefulness and limitations of coccoliths and alkenones in sediments from the northwest North Atlantic, including the subpolar Baffin Bay. The combined analysis of coccoliths and alkenones is useful to decipher taphonomic biases related to poor calcite preservation and alkenone fluxes not resulting from calcifying haptophyte fluxes. This thesis also contributes to the understanding of the distribution of coccoliths and alkenones in the northwest North Atlantic, but many questions remain. I focused on coccolithophore productivity in this thesis, but I made no attempt to quantify the relative contribution of coccolithophores to the overall primary productivity. This is a topic that could be explored further through the study of the recent biogenic components of the sediments including biomarkers, species assemblages, and fluxes in relation to modern primary productivity and that of haptophytes specifically.

Non-calcifying haptophytes are an important component of the marine ecosystem. Their presence and relationship to calcifying haptophytes need to be examined thoroughly in future research. Non-calcifying haptophytes produce organic matter, including alkenones, that contributes to the pool of the marine organic carbon in the water column, some of which gets preserved in the sediments. Coccoliths themselves contribute significantly to the marine carbon budget (e.g., Baumann et al., 2004a, b; 2005), as they remove carbon from the ocean when

preserved in the sediments. Accordingly, deciphering the relative contribution of the calcifying coccolithophores from the non-calcifying haptophytes to the carbon flux to the sediment, over time and space, is important to improve our understanding of the marine carbon budget. Their quantification deserve special attention in future studies.

Another important aspect to consider is the sources of errors and uncertainties in the primary productivity data used in this thesis, along with the difference between export productivity and primary productivity. Export productivity is the amount of biomass falling out of the euphotic zone into the deep sea, and it is different again from the fraction of export productivity that gets buried in the sediments (buried export productivity). Primary productivity data are readily available (although imperfectly, with their sources of errors and uncertainties) and rely on global chlorophyll estimates derived from algorithms that transform remote sensing data. For example, the primary productivity estimates that I used in this thesis were generated using a model that relies on MODIS chlorophyll data. However, I am aware of another source, the GlobColour project, which merges chlorophyll data from different sensors in missions managed by NASA and the European Space Agency (ESA), which is considered to be the current state-of-the-art data on surface ocean chlorophyll concentration derived from remote sensing (<http://globcolour.info>; d'Andon et al., 2009). Estimating export productivity, and export haptophyte productivity, requires further steps in modelling than what I did in this thesis. I, therefore, suggest further steps with modelling to capture buried export haptophyte productivity, along with considering using data from the GlobColour project. Future studies could investigate how alkenone concentrations scale with the buried export productivity, and regardless of whether there is a significant relationship, that type of future work can shine a light on an issue that needs more discussion in future research.

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