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U, Th and Pa insights into sedimentological and paleoceanographic changes off Hudson Strait (Labrador Sea) during the last ~37 ka with special attention to methodological issues

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A B S T R A C T

A ~9 m-long sediment core spanning the last ~37 ka has been raised from the lower Labrador continental slope, off the Hudson Strait shelf edge. It has been analyzed for its U, Th and Pa isotope contents, along with current sedimentological parameters, as a means to retrieve information about sedimentological changes in response to northeastern Laurentide Ice Sheet (LIS) margin instabilities. The sequence yielded a high-resolution record of subglacial detrital carbonate pulses from Hudson Strait assigned to "Heinrich events" H2 and H1, whereas H0 was missing. Large variations in bulk sediment U- and Th-contents as well as in ^{234}U/^{238}U activity ratio are observed throughout the sequence, leading to large uncertainties when calculating excesses in ^{231}Pa and ^{230}Th ($^{231}Pa_{xs}$ and $^{230}Th_{xs}$) over their supported and in-growth fractions (i.e., inherited from detrital minerals and produced from authigenic and diagenetic U-uptake). In particular, ^{234}U excesses or deficits *vs* ^{238}U ($-115‰ < \delta^{234}U < +126‰$) are observed throughout the sequence, suggesting occasional U-uptake from the water column and/or some late diagenetic mobility along discrete redox gradients, despite the overall low and little variable organic carbon content (0.3 ± 0.1%) observed. The above uncertainties in $^{231}Pa_{xs}$ and $^{230}Th_{xs}$ estimates and the large variability in geochemical and sedimentary fluxes off the northeastern LIS margin, lead us to downgrade the potential paleoceanographic information yielded by these isotopes in such a setting. Nonetheless, the H2 and H1 layers are highlighted by very low initial excesses in both $^{230}Th_{xs}$ and $^{231}Pa_{xs}$, indicating their extremely fast deposition. Throughout most of the sedimentary sequence, the calculated initial $^{230}Th_{xs}$ fluxes are nearly in balance with ^{230}Th production in the overlying water column. Exceptions are the H2 layer, an interval succeeding H1, and the post-glacial sediment. The estimated initial ($^{231}Pa_{xs}/^{230}Th_{xs}$) ratios are generally lower than their production rate in the water column (i.e., 0.092), indicating nearly continuous preferential export of $^{231}Pa_{xs}$ over the last ~37 cal ka BP, thus the persistence of some deep currents throughout the interval.

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1. Introduction

Deep-water convection in subarctic seas is a key component of the North Atlantic climate system as it regulates ventilation and heat-exchange rates with the atmosphere (e.g., Broecker, 1991). In this respect, the Labrador Sea constitutes a transitional basin between the Arctic and the North Atlantic. Freshwater fluxes, either from the Arctic Ocean or from surrounding ice sheets, notably the Laurentide Ice Sheet (LIS), have tightly controlled convection in this basin (Labrador Sea Water – LSW; Lazier, 1973; Hillaire-Marcel et al., 2001). Major melting events from the northeastern LIS margin have been associated with abrupt climate/ocean instabilities, in particular most of the millennial-scale Heinrich (H) events which resulted in the deposition of recognizable layers throughout the North Atlantic sea floor, based on their sedimentological and geochemical specific features (Hemming, 2004; Rashid et al., 2012, and references herein). Sedimentary processes and fluxes in the Labrador Sea have accordingly undergone significant changes, essentially driven by the sporadic release of large amounts of terrigenous material from the LIS (e.g., Andrews et al., 1994), and/or through changes in the oceanographic regime (e.g., Wu and Hillaire-Marcel, 1994), as well as due to variations in primary productivity (e.g., Hillaire-Marcel et al., 1994b).

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Here, based principally on measurement of isotopes of uranium (^{234}U, ^{238}U), thorium (^{230}Th, ^{232}Th) and protactinium (^{231}Pa) in a ~9 m-long core (HU08-029-004PC) raised from the lower continental slope off the Hudson Strait outlet, we intend to document changes in terrigenous fluxes and sedimentary dynamics off the northeastern LIS meltwater route during the last ~37 ka. Initial excesses in ^{230}Th and ^{231}Pa [(^{230}Th$_{xs}$)$_0$ and (^{231}Pa$_{xs}$)$_0$], i.e., the fractions of these isotopes unsupported by parent isotopes in the sediment at the time of their deposition, have been estimated using a calibrated ^{14}C-chronology of the sedimentary sequence (Gibb et al., 2014) to carry back in time ^{230}Th and ^{231}Pa activities, and calculate their post-depositional in situ production (e.g., Anderson et al., 1990; Veiga-Pires and Hillaire-Marcel, 1999). U-, Th- and Pa-concentrations and activity ratios were measured with a centennial to millennial resolution due to the highly variable sedimentation rates at the site (Gibb et al., 2014). In order to link U, Th and Pa isotopes and sediment properties, CAT-scan images have been used to document sedimentological features and complemented by measurements of the coarse fraction content (used as proxy for IRD abundance), of organic and inorganic carbon concentrations (C_{org} and C_{inorg}), and semi-quantitative mineralogical analyses of the bulk sediment. Based on micropaleontological data and transfer function reconstruction of paleo-sea surface conditions, Gibb et al. (2014) identified three distinct climatostratigraphic intervals: a glacial interval when productivity was practically nil (~36.6–12.2 cal ka BP), a deglacial interval characterized by low salinity conditions due to the transit of huge amounts of meltwater (~12.2–8.3 cal ka BP) and the post-glacial, when productivity rose rapidly to full interglacial values (≤~8.3 cal ka BP). Gibb et al. (2014) highlighted the fact that if the site did record major meltwater pulses linked to H3 (?), H2, H1 and the final drainage of Lake Agassiz, it did not record the Younger Dryas nor the H0 event (see Andrews et al., 1994; Hillaire-Marcel et al., 1994a; Pearce et al., 2013, for information on the H0 event in the Labrador Sea). Building on Gibb and others' work (2014), we intend to further document here detrital sediment sources and fluxes as well as sedimentary processes, in the western most area of Labrador Sea, based on U- and Th-series isotopes. In a context of continental slope processes and at the emplacement of the LIS margin, these tracers could provide information about the past ice-sheet dynamics as well as about the variability of deep currents (e.g., Yu et al., 1996; Veiga-Pires and Hillaire-Marcel, 1999).

2. Materials and methods

The 895 cm-long core HU08-029-004PC (henceforth 004PC) was retrieved from a water depth of 2674 m on the lower Labrador continental slope, approximately 180 km seaward from the Hudson Strait shelf edge (61°27′N, 58°2′W; Fig. 1; Campbell and de Vernal, 2009).

2.1. Sedimentological parameters

Once core sections were split lengthwise, the sediment was visually examined for its structure, texture and color. Sections were then scanned at high resolution (~0.1–1 mm) by computerized coaxial tomography (CAT-scan) technique to acquire 3D X-rays images (St-Onge and Long, 2009). CAT-scan gives a view on the internal structure of the sediment cores, especially specific features such as IRD. Core sections were also photographed at 500 dpi (dots per inch) using a high resolution Smartcube™ Smart Camera Image Scanner (SmartCIS).

Qualitative and semi-quantitative ($\pm 1\sigma \approx 5\%$) analyses of mineralogical assemblages were made using X-ray diffraction (XRD) following procedures described in Last (2001), Moore and Reynolds (1997) and Thorez (2003). A homogeneous powder of the <63 µm bulk fraction was scanned using a Siemens D5000™ diffractometer, with a CoKα1, 2 X-radiations ($\lambda = 1.76896$) and a Si detector, between 2° and 50° 2θ angles. Qualitative and semi-quantitative estimates were based on peak identifications and relative intensity measurements of their X-ray patterns using the Diffracplus EVA™ software.

Total carbon- (C) content, inorganic carbon (C_{inorg}) and organic carbon (C_{org}) were analyzed on the bulk-fraction at 8 cm-intervals and reported in percentage *vs* dry sediment weight (dw%; Fig. S3). Aliquots of ~9 mg were analyzed for their total C-content using an NC 2500TM elemental analyzer (Carlo-Erba™). Aliquots were then fumigated for ~24 h with vapors of 12 M HCl to dissolve carbonate minerals and measure the residual C_{org}-content (Hélie, 2009). Values of C_{inorg} were finally estimated by subtracting the C_{org} values from the total C values. However, this dissolution procedure may have not removed all the fraction of the C_{inorg} (dolomite can be partly refractory to acid treatment) or might be slightly biased by the dissolution of HCl-leachable compounds, both resulting in minor anomalies in C_{org}-estimations. An internal precision of 0.1% ($\pm 1\sigma$) was nonetheless estimated based on duplicated analyses from the analog coulometer technique (see Fig. S3). Assuming that the C_{inorg} content is representative of calcite and dolomite abundances (based on X-ray diffraction scans; Moore and Reynolds, 1997; Last, 2001; Thorez, 2003), estimates of their respective dry weight percentages within the sediment were calculated.

The core chronology was established by Gibb et al. (2014) and constrained by radiocarbon (^{14}C) dates obtained from hand-picked planktic foraminifera *Neogloboquadrina pachyderma* sinistral from the dw% > 106 µm sediment fraction (Fig. 2). Accelerator mass spectrometry (AMS) measurements were made at the *Lawrence Livermore National Laboratory* and at the *National Ocean Sciences AMS Facility*. Radiocarbon ages were calculated using the Libby half-life of 5568 years and normalized to a δ^{13}C-value of −25‰. A marine reservoir correction of 400 years was applied ($\Delta R = 0$; see a discussion in the supplementary material of Hillaire-Marcel et al., 2007), and the ages were converted to calibrated years using Oxcal 4.2 (Ramsey, 2008) and the Marine09 calibration curve (Reimer et al., 2009). The *p-sequence* model in OxCal was selected for estimating the probability of each age. This model is based on a Bayesian approach taking into account the probability distribution of calibrated ages, the sample depths and changes in deposition rates (see Gibb et al., 2014, for more details). Three ^{14}C ages depict in particular an age interval of 8441–8179 cal ka BP for the 85–36 cm layer, with an average value of 8327 cal ka BP, deposited during the final drainage of Lake Agassiz (e.g., Barber et al., 1999; Hillaire-Marcel et al., 2007; Lewis et al., 2012), although the sediment does not show here a significant increase in detrital carbonates in contrary to what is generally observed (e.g., Hillaire-Marcel et al., 2007; Nicholl et al., 2012). Linear interpolation between each modeled age were used to calculate the sedimentation rates and the age–depth relationship, which was finally used to plot proxy data against age as well as to carry back in time radioactive disequilibria in the U-series isotopes. The modeled results provide a basal age of ~36.6 cal ka BP. Sedimentation rate is hence ~24.4 cm ka^{-1} on average but depicts large variations downcore (see Fig. 2).

2.2. Radionuclide analyses

Several techniques were combined for the measurement of actinides. ^{238}U, ^{234}U, ^{232}Th and ^{230}Th separation and extraction were performed following analytical procedures described in Lally (1992). Sampling was made at 12 cm intervals from bottom up to a depth of 140 cm depth downcore, then at 8 cm–4 cm intervals,

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