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Key Points:

- Thorium measured in seawater and sediments to quantify glacial traceelement nutrient inputs at the West Greenland margin
- Thorium-232 fluxes are elevated and are similar to other ocean margin sites with high sediment inputs
- Elevated iron fluxes support high primary productivity and reach the beyond the coastal ocean

Supporting Information:

Supporting Information may be found in the online version of this article.

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High Lithogenic and Micro-Nutrient Fluxes From the West Greenland Margin Traced by Thorium in Seawater and Sediments

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Abstract The flux of nutrients from continents to the oceans sustains oceanic primary productivity and is a fundamental component of the carbon cycle. In most regions of the world's oceans primary productivity is limited by the supply of nutrients. In particular, iron can become limiting in the open-ocean due to its low solubility. Glaciated continents have been suggested as an underappreciated source of iron to the high-latitude oceans. Yet, uncertainty remains regarding the magnitude and spatial variability of glacially derived nutrient fluxes, and the extent to which these nutrients impact open-ocean ecosystems. To quantify lithogenic fluxes at the West Greenland margin, we measured ²³²Th and ²³⁰Th in seawater and core-top sediments across the shelf and slope. Our results highlight a negative correlation between low-salinity waters and dissolved and particulate ²³²Th, suggesting a glacial source for this lithogenic isotope. We calculated dissolved ²³²Th fluxes $5-24~\mu g~m^{-2}~yr^{-1}$ (100–500 m depth), and sedimentary ^{232}Th fluxes 105–711 $\mu g~m^{-2}~yr^{-1}$, higher than typical open-ocean settings and similar to margin sites influenced by large inputs from aeolian dust and rivers. A sampling transect shows that dissolved ²³²Th fluxes increase toward Greenland, confirming that lithogenic inputs are sourced laterally from the margin. Using our ²³²Th fluxes, we estimate an elevated supply of dissolved Fe which extends over the continental slope toward the open ocean. This Fe flux is large enough to support much of the local primary productivity, highlighting the importance of lithogenic fluxes in supporting the marine ecosystem in high-latitude oceans.

Plain Language Summary Small plant-like creatures (algae) living in the surface ocean are an important food source for other marine creatures and affect the Earth's climate by drawing carbon dioxide into the ocean when they grow. In most parts of the oceans the algae's growth is limited because they do not have enough of certain nutrients, such as iron. It is possible that around Greenland, algae are supplied with iron that comes from mud ground-up by ice on the continent. There are no measurements of how much, and how quickly, mud (and iron) reaches the algae in the ocean far away (around 100 km) from the coast of Greenland. We measured two types of the radioactive element thorium in seawater, and seafloor mud, to calculate the rate that mud is supplied to the ocean off West Greenland. We found that high amounts of thorium and iron are supplied to the region, enough to account for the large quantity of algae growing there. This suggests that the ice on Greenland, and maybe also on Antarctica, helps to keep the algae in the ocean growing. Changes to Earth's ice-sheets might then impact how algae grow and absorb carbon dioxide in the future.

1. Introduction

The rate at which carbon is fixed by marine phytoplankton and exported from the surface ocean is a key variable in the global carbon cycle, and is linked to changes in the Earth's climate (Hain et al., 2014; Wilson et al., 2022). One important factor regulating carbon exported to depth (export productivity) is the availability of macro and micronutrients for phytoplankton growth (Moore et al., 2013). In some regions of the ocean, one or more key nutrients may be depleted and thus are considered limiting to primary productivity (Moore et al., 2013).

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Export production is mainly supported by the cycling of dissolved nutrients to the surface ocean from deeper waters (Moore et al., 2013); however, changing inputs of nutrients from external sources can alter regional and global net primary production and export production (Cotrim da Cunha et al., 2007). In recent years, it has been suggested that nutrient limitation in the high-latitude oceans may be alleviated by nutrients supplied by glaciers and ice sheets (Bhatia et al., 2013; Dinniman et al., 2023; Wadham et al., 2019). High inputs of macro- and micronutrients from the Greenland Ice Sheet have been measured at glacial outflows (Hawkings et al., 2014, 2016), which contain disproportionate concentrations of bioavailable elements compared to other settings (Hawkings et al., 2018; Shoenfelt et al., 2019).

Whether glacially derived nutrients relieve nutrient limitation depends on two key factors. The first is the status of the phytoplankton community with respect to nutrient limitation: additional nutrients will only stimulate new productivity if they include the currently limiting nutrient, or co-limiting nutrients. For example, in regions limited by nitrate (Moore et al., 2013), additional fluxes of Fe or phosphate will not increase primary productivity (unless by the stimulation of nitrogen fixing organisms; von Friesen & Riemann, 2020). Second, waters carrying nutrients from the glacial terminus must be transported to the nutrient-limited regions in the open ocean (Arrigo et al., 2017; Hopwood et al., 2018) before the nutrient load is removed from the water column (e.g., by biological uptake in fjords or settling and flocculation of particles; J. Krause et al., 2021; Ng et al., 2024).

There is evidence of Fe limitation in the Irminger and Iceland basins (Hopwood et al., 2018), and of a deficiency in Fe relative to nitrate in the central Labrador Sea (Tonnard et al., 2020). Conversely, data from the East and West Greenland margins show excess of Fe relative to nitrate (Hopwood et al., 2018; Tonnard et al., 2020), suggesting nitrate is the more limiting nutrient. Silicic acid, rather than Fe, appears to limit diatom growth during the spring bloom across parts of the sub-polar North Atlantic, including at the outlet of Godthåbsfjord, close to Nuuk, Greenland (J. W. Krause et al., 2019). Additionally, it has been suggested that light limitation is important during the late summer and autumn (Oliver et al., 2018). Over the continental shelf and slope near Nuuk, Greenland, recent studies indicate high chlorophyll concentrations and moderately productive diatom communities, despite low dissolved macronutrient concentrations (Hendry et al., 2019; Ng et al., 2024). Similarly, Tonnard et al. (2020) link the depletion of nitrate relative to Fe at the Greenland margins to the strong bloom and high chlorophyll concentrations. These results point towards phytoplankton at the Greenland margins that rely on a substantial supply of nutrients to maintain productivity, and may be limited by macro-, rather than micronutrients. Part of the nutrient supply to high-latitude surface waters may arise from upwelling of deep, nutrient rich, marine waters (Cape et al., 2019; Hopwood et al., 2018), diapycnal mixing (Painter et al., 2014), benthic supply (Ng, Cassarino, et al., 2020) or the export of glacially derived particles (Ng et al., 2024). Seasonal convective mixing is thought to account for the dominant flux of Fe to the surface in the Iceland and Irminger basins (Achterberg et al., 2018; Painter et al., 2014). Similarly, winter mixing can supply nutrients to the surface at the West Greenland margin prior to the spring bloom (Harrison et al., 2013); however, a shallow mixed layer persists from spring through to autumn (Arrigo et al., 2017), in part due to fresh surface waters (Rysgaard et al., 2020). Thus, upwelling is less likely during the summer bloom, when glacial melt waters may be a more important nutrient source (Arrigo

There is a significant knowledge gap in processes governing the export of glacially derived nutrients to the open ocean (Wadham et al., 2019); some nutrients are expected to behave conservatively during estuarine mixing, while others are highly non-conservative (Hopwood et al., 2020; J. Krause et al., 2021). Removal of Fe across salinity gradients in estuarine environments may be >90% (Poulton & Raiswell, 2002) and similar removal has been observed or assumed in Arctic fjords (Hopwood et al., 2020). Despite the large proportion of dissolved Fe that is removed in fjords, Fe concentrations close to fjord outlets (~5–10 nM) can still be an order of magnitude larger than those on the surface of nearby coastal (~<1 nM) or open (~< 0.5 nM) oceans (Hopwood et al., 2016; J. Krause et al., 2021; Tonnard et al., 2020). Thus, the transfer of waters from fjords to the open ocean may also be associated with a flux of nutrients. In addition to melt waters, Fe sourced from sediment resuspension has been demonstrated in other glaciated coastal settings (Sherrell et al., 2018). Nevertheless, there have been few quantitative assessments of lithogenic fluxes to the open oceans close to ice sheets, and significant uncertainty remains regarding their importance in supplying these nutrients (Hawkings et al., 2020; Hopwood et al., 2015, 2020; Wadham et al., 2019).

Terrigenous fluxes from the Greenland Ice Sheet have been estimated by combining suspended sediment concentrations (from remote sensing) and modeled ice sheet run-off (Overeem et al., 2017). These estimates indicate

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Here we make a quantitative assessment of the lithogenic supply over the West Greenland continental shelf and slope by applying a scavenging model to measurements of lithogenic ²³²Th and radiogenic ²³⁰Th in seawater and core-top sediments. We extend our quantitative assessment to estimate fluxes of dissolved Fe and argue that our flux estimates represent a plausible lower bound for the region.

2. Materials and Methods

In the following section, we outline the theoretical models, sampling methods and laboratory procedures used in our study. A more detailed description of sampling, laboratory methods and quality control is given in Supporting Information S1.

2.1. Using ²³²Th/²³⁰Th to Calculate Lithogenic Fluxes

2.1.1. Dissolved Thorium Isotopes

The method we use in this study calculates the vertical removal (scavenging) flux of ²³²Th onto particles, and by assuming steady-state, estimates the supply of ²³²Th required to balance this removal—assumed to be from dissolving lithogenic particles, analogous to aeolian dust deposition (Hayes et al., 2013; Hsieh et al., 2011). At our study sites, lithogenic material is likely delivered laterally from the continent through surface waters, such that vertical removal fluxes are directly linked to lateral lithogenic supply (Hendry et al., 2019; Supporting Information S1).

To estimate fluxes of dissolved ²³²Th (d²³²Th), we calculate the residence time of dissolved thorium by measuring radiogenic dissolved ²³⁰Th (d²³⁰Th), which is depleted in seawater compared to its parent isotope ²³⁴U, due to adsorption onto particles (which subsequently sink, 2.1.2; Bacon & Anderson, 1982). The integrated deficit of d²³⁰Th, relative to production by ²³⁴U, provides a measure of the ²³⁰Th removal rate (Broecker et al., 1973; Hsieh et al., 2011). Assuming a steady state and negligible contributions from advection and eddy diffusion, the scavenging timescale for ²³⁰Th is equivalent to a residence time (Equation 1). A vertically integrated removal flux of ²³²Th can be calculated for different depths by applying the integrated ²³⁰Th residence time to ²³²Th inventories (Hayes et al., 2013, Equation 2). Application of ²³⁰Th residence times to ²³²Th requires an assumption that the isotopes share the same chemical behavior (e.g., speciation), despite their different sources. Though subject to some uncertainty, initial studies testing this assumption suggest that the isotopes share similar speciation (Anderson et al., 2025; Hayes et al., 2017; Hayes, Fitzsimmons, et al., 2015; Pavia, Anderson, Pinedo-Gonzalez, et al., 2020).

This approach has been used to estimate fluxes of ²³²Th from dust in the open-ocean Atlantic and Pacific (Anderson et al., 2016; Hayes et al., 2013, 2017, 2018; Hayes, Fitzsimmons, et al., 2015; Hsieh et al., 2011; Pavia, Anderson, Winckler, & Fleisher, 2020), as well as in the Southern Ocean to determine glacial lithogenic fluxes (Pérez-Tribouillier et al., 2020). A modeling study indicates that 2/3 of global ²³²Th input may be linked to a sedimentary source, although ²³²Th inventories disproportionately reflect surface inputs (Xu & Weber, 2025). For the margin setting investigated here, with dynamic physical oceanography, we also consider the impacts of advection and diffusion on our flux calculations (Supporting Information S1).

In Equation 1 the integration is carried out using the trapezium rule and the activities from surface to the first sample in a depth transect are assumed to be constant (as might be expected in the surface mixed layer). For profiles with the shallowest sample at >50 m (CTD8, 9, and 24), a surface 230 Th concentration is estimated from the average of shallow samples from the other profiles (CTD4, 6 and 14; Supporting Information S1) to allow for integration to depth. The low surface concentrations mean that this approximation contributes a small amount to the total 230 Th inventory: doubling the estimated surface 230 Th concentration increases the inventory at the deepest profile depths by only 2%–5%. In Equation 1, λ_{230} is the decay constant of thorium-230, and 230 Th and 234 U are the activities of each isotope per volume of seawater. The subscript "xs" refers to 230 Th that is not derived

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from lithogenic material—estimated using the d^{232} Th concentration and 4×10^{-6} mol/mol as an estimate of 230 Th/ 232 Th released from lithogenic particles (within uncertainty of average upper continental crust, UCC, assuming secular equilibrium in the 238 U series; Roy-Barman et al., 2009; Rudnick & Gao, 2003). The average fraction of lithogenic 230 Th is 16% of the total, but is up to 54% for shallow samples with high 232 Th, such that shallow 230 Th concentrations are sensitive to the chosen lithogenic 230 Th/ 232 Th ratio. Because 230 Th profiles are integrated in Equation 1, the sensitivity of our flux calculations to this ratio is reduced. For example, at CTD006, halving the lithogenic 230 Th/ 232 Th to 2×10^{-6} mol/mol (the lower uncertainty bound for UCC; Rudnick & Gao, 2003) produces a \sim 40% decrease in 232 Th flux at 5 m depth, but only an 11% decrease at 642 m.

$$\tau_{\text{Th}_z}[\text{yr}] = \frac{\int_0^z 230 \text{Th}_{xs} dz}{\lambda_{230} \int_0^z \left(234 U - 230 \text{Th}_{xs}\right) dz}$$
(1)

²³²Th flux
$$_{Z}[\mu g m^{-2} yr^{-1}] = \frac{\int_{0}^{z_{232}} Th dz}{\tau_{Thz}}$$
 (2)

Fe flux [mg m⁻² yr⁻¹] =
232
Th flux × UCC_{Fe/Th} × S_{Fe/Th} (3)

The supply of other dissolved trace metals (e.g., Fe) can be assessed using the ratios of abundance (in upper continental crust, $UCC_{Fe/Th}$) and solubility relative to ^{232}Th ($S_{Fe/Th}$; Equation 3; Hayes, Anderson, et al., 2018). For Fe, previous studies either assumed $S_{Fe/Th} = 1$ or used estimates from leaching mineral dust (Hayes et al., 2017, 2018; Hayes, Fitzsimmons, et al., 2015) and used existing estimates of Fe/Th from the bulk or upper continental crust (Hayes et al., 2013, 2018; Rudnick & Gao, 2003; Taylor & McLennan, 1995).

2.1.2. Thorium-230 Normalization

A related method to estimate lithogenic fluxes in the modern ocean is ²³⁰Th normalization applied to core-top sediments (Anderson et al., 2016; Rowland et al., 2017). This technique relies on the assumption that the scavenging removal flux of 230 Th on sinking particles is equal to its well-established production rate from 234 U (β) in the water column above (François et al., 2004). This assumption has been demonstrated to be accurate to within ~30-40% in most of the ocean and the method has been widely applied to modern and Pleistocene sediments (Costa et al., 2020). By combining the ²³⁰Th-normalized vertical settling flux of sediment (Equation 4) with the concentration of ²³²Th in the sediment, a flux of ²³²Th can be derived for core-top sediments (Equation 5). Our dissolved and sedimentary approaches both compare the well-constrained production of d230Th against its concentration to estimate fluxes (cf. denominator, Equation 1 and numerator, Equation 4). To calculate a mass flux, only 230 Th derived from 234 U in the overlying water column is considered, 230 Th_{xs}; it is necessary to estimate and subtract lithogenic and authigenic ²³⁴U from the total. This estimation relies on assumed ratios of lithogenic and authigenic ²³⁸U/²³²Th and ²³⁰Th/²³⁸U; fluxes are particularly sensitive to the assumed ratios when the proportion of ²³⁰Th in these fractions is high (Missiaen et al., 2018). We assess this sensitivity for our sites in the supplementary information (Figure S1 in Supporting Information S1). We correct for ²³⁰Th decay since deposition by assuming an age of 1,000 and 2000 years for sediments 0-1 and 1-2 cm depths, respectively. Because core-top ages are likely to be much less than the half-life of ²³⁰Th, decay corrections have little effect on calculated mass fluxes; varying the assumed ages from 0 to 5,000 years changes the calculated mass fluxes by <5%.

Mass flux
$$[g m^{-2} yr^{-1}] = \frac{\beta [dpm m^{-3}yr^{-1}] \times z [m]}{^{230}Th_{xs,0}[dpm g^{-1}]}$$
 (4)

²³²Th flux _{Sed} [
$$\mu$$
g m⁻² yr⁻¹] = Mass flux × ²³²Th [μ g g⁻¹] (5)

2.2. Oceanographic Setting of the West Greenland Margin

Samples were collected at the West Greenland margin in July-August 2017 during the DY081 cruise, onboard the RRS *Discovery* (Hendry et al., 2019). Shallow flow (~<200 m) at the West Greenland margin is dominated by the West Greenland Current (WGC) that flows north-westwards, parallel to the coast, forming part of the sub-polar gyre (Figure 1; Holliday et al., 2007; Myers et al., 2009). During the 2017 DY081 cruise, geostrophic velocities in

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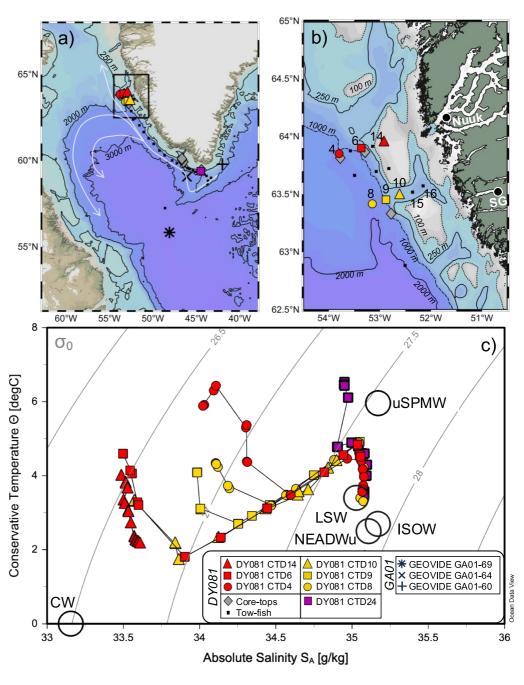


Figure 1. (a) Sample sites at the West Greenland margin during the DY081 cruise (this study, colored symbols) and three sample sites from the GEOVIDE cruise (Deng et al., 2018; large black symbols). Small black squares indicate the location of DY081 underway (tow-fish) surface particulate samples (also shown in Figure 2). White arrows schematically depict surface currents. (b) The sampling grid offshore Nuuk: the location of Nuuk and the Sermeq Glacier (SG) are indicated and bathymetry is shown on a color scale and by black isolines. (c) Hydrography of sampling sites and approximate end-member water-mass compositions (open circles) from the literature: CW = Greenland coastal water, uSPMW = upper sub-polar mode water, LSW = Labrador Sea water, ISOW = Iceland-Scotland overflow water, NEADWu = upper Northeast Atlantic deep water (García-Ibáñez et al., 2018; Rysgaard et al., 2020). Figure created using Ocean Data View (Schlitzer, 2025).

the cores of the WGC were measured to be on the order of $0.1-0.3~{\rm m~s}^{-1}$ (Hendry et al., 2019). The study area has high eddy kinetic energy (Cuny et al., 2002), and eddies move heat and buoyancy delivered by the boundary currents further into the Labrador Sea interior (Saenko et al., 2014). The hydrography at our sites (Hendry et al., 2019) is controlled by the balance of Greenland coastal water (CW, mostly <50 m), upper sub-polar mode

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water (uSPMW, increasing to \sim 500 m) (Rysgaard et al., 2020), and a contribution from a cooler, slightly saltier, water mass (>500 m, Figure 1). All our sites also show a strong thermocline, with temperatures decreasing from the surface to a depth of \sim 30–50 m.

Measurements of chlorophyll concentration and biogenic silica (i.e., diatom) productivity during the DY081 cruise (Hendry et al., 2019) indicate there is an active phytoplankton community off West-Greenland. Median biogenic silica productivity in the sampling grid offshore Nuuk (Figure 1b) was >2x the global average (Hendry et al., 2019; Nelson et al., 1995). These initial findings suggest that a relatively high supply of micronutrients (e.g., Fe) may be required to sustain the high levels of productivity, and that as macronutrient concentrations are low in the surface waters, the plankton may be limited by these nutrients.

2.3. DY081 Cruise and Sampling

Samples were collected during the DY081 cruise in July 2017 as part of the wider ICY-LAB project (Isotope CYcling in the LABrador sea; Hendry et al., 2019). Water samples (\sim 5 L, n=24) were collected from 7 CTD casts, southwest of Nuuk (6 casts), and near Cape Farewell (1 cast; Figure 1). Samples were filtered through 0.2 μ m pore-size membrane filters and collected in acid-cleaned polypropylene 6 L containers. The samples collected in cast CTD024 were filtered using a 0.45 μ m pore-size filter. After filtration, samples were immediately acidified to pH \sim 2 using \sim 5 mL of concentrated Romil UpA HCl (Anderson et al., 2012).

Surface particulate trace metal samples were collected at a depth of \sim 2 m while the ship was moving using a trace-metal clean underway towed-fish apparatus (e.g., Annett et al., 2017). Samples of unfiltered seawater (3 L) were collected in acid-cleaned HDPE bottles and then filtered through acid-cleaned Supor 0.45 μ m filters at <34 kPa using filtered air.

Sediment samples were taken from the top (0-1 cm and 1-2 cm intervals) of short (<40 cm) cores collected using a mega (multi)-corer (5 cores, 7 samples) and remotely operated vehicle (9 push-cores, 10 samples), recovered from water depths 953–1,327 m. Cores were sliced into 1 cm intervals during the cruise, and the samples were stored refrigerated prior to analysis.

2.4. Laboratory Procedures

Filtered seawater samples were processed at the University of Bristol in the Bristol Isotope Group clean laboratory broadly following procedures set out by previous studies (Auro et al., 2012; Ng, 2016; Ng, Robinson, et al., 2020; Rowland et al., 2017). The main acids used in the laboratory, HNO_3 and HCl, were distilled in-house using Teflon stills. The other reagents used were Romil SpA grade, and Milli-Q water with a resistivity of $18.2 \text{ M}\Omega$ cm.

To summarize briefly the laboratory protocol, \sim 5 L samples were weighed and spiked with known amounts of 229 Th and 236 U from two separate spike solutions. Thorium and uranium were co-precipitated with Fe(OH)₃ by the addition of concentrated ammonium hydroxide. Supernatants were separated from precipitates by a peristaltic pump and the precipitates were collected and dissolved. Uranium and thorium were separated and purified using anion exchange column chromatography. Before analysis, samples were subjected to an oxidative step involving HClO₄, H₂O₂ and concentrated HNO₃.

Surface water particulate samples were processed at Rutgers University, according to previously established methods (Annett et al., 2017; Planquette & Sherrell, 2012): filtered particles were fully digested in Teflon containers on a hotplate using a mixture of HNO₃ and HF. Sediment samples were analyzed at Lamont-Doherty Earth Observatory (LDEO) according to previously published methods (Fleisher & Anderson, 1991, 2003). Sediment was spiked with a mixed ²³⁶U-²²⁹Th spike and digested by heating samples in a mixture of concentrated HNO₃, HClO₄ and HF. Uranium and thorium were separated from the sample matrix by co-precipitation with Fe (OH)₃ and anion exchange column chromatography. Acids were ACS "Plus grade" and "Optima grade" (Fisher Scientific).

2.5. Mass Spectrometry

Dissolved Th isotopes were analyzed at the University of Bristol by multi-collector inductively coupled plasma mass spectrometry (MC-ICP-MS) following the peak-jumping routine of Auro et al. (2012) and following the

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protocols of Ng (2016). Each sample was bracketed by clean acid to monitor and correct the instrumental blank, and also by a thorium bracketing standard to correct for instrument mass bias and the ion counter yield. Corrections were also made for the tailing of the larger ²³²Th beam on ²³⁰Th and ²²⁹Th masses. Uncertainties based on the signal variability were propagated through all corrections. The measured ²³⁰Th concentration is corrected for the radioactive ingrowth of ²³⁰Th from ²³⁴U since the time of sample filtration (Anderson et al., 2012; Robinson et al., 2004).

Particulate ²³²Th surface samples were analyzed at Rutgers University as part of a multi-element method using single-collector high-resolution ICP-MS according to previously established methods (Annett et al., 2017; Planquette & Sherrell, 2012). Concentrations were quantified relative to external standard solutions, and corrected for sensitivity drift and matrix effects using added ¹¹⁵In, and for full procedural blanks. Sedimentary thorium and uranium isotopes were analyzed at LDEO by ICP-MS (Thermo Scientific Element Plus). A uranium standard (SRM129) was used to correct for instrumental mass bias and to determine detector gain. Corrections were also made for the tailing of the larger ²³⁸U and ²³²Th beams on the minor isotopes.

Full procedural blanks (one or two per batch) for dissolved 232 Th were 13 ± 16 pg $(2\sigma, n = 7)$, with only one >15 pg and for 230 Th were 0.3 ± 0.4 fg $(2\sigma, n = 7)$, with one >0.4 fg. Blank corrections make up a maximum of 4% and 5% of the total analyte mass for 232 Th and 230 Th respectively. Procedural blanks for particulate 232 Th samples were 2.79 ± 1.86 pg, which represents 1%-18% of the signal for samples reported here (mean = 6%). Based on a typical volume filtered of 2-3 L, our detection limit for particulate 232 Th in seawater using this method is 3.9-5.9 pg kg $^{-1}$. One full procedural blank for sediment samples measured at LDEO contained 1.1 ± 0.02 ng 232 Th $(\pm 2\sigma)$ and 40 ± 1.1 fg $(\pm 2\sigma)$ 230 Th, representing <0.6% and <0.4% of the respective analytes.

3. Results

3.1. Surface Particulate ²³²Th Concentrations

Surface particulate 232 Th (p 232 Th) concentrations show a large range from 10 to 552 pg/kg. High concentrations >300 pg/kg are seen at the Nuuk sampling grid and off Narsaq (\sim 60°N, 47°W) (Figure 2a), where low salinity waters are also present. Two distinct negative correlations exist between p 232 Th and salinity, specific to longitude ranges, with the relationship for samples collected <50°W showing a shallower gradient than those collected >50°W (closer to Nuuk) (Figure 2c). The two regression lines converge around a salinity of 34 and p 232 Th concentration of \sim 75 pg/kg. Concentrations of p 232 Th are much lower (generally \leq 20 pg/kg) at sites with higher salinities and/or further from the coast.

3.2. Dissolved ²³²Th and ²³⁰Th Concentrations

Dissolved 232 Th concentrations ranged from 54 to 360 pg/kg, with a mean of 138 ± 86 (1σ). For each CTD cast, the values are highest in the shallowest samples, and decrease from the surface to a mid-depth minimum around 500 m. Results from CTD004 also show small but consistent increases in the 4 samples from \sim 500 to 1,050 m. Deeper than 500 m, concentrations of 232 Th are highest closer to the coast (at a given depth) for both transects at Nuuk. The two transects show a similar range of values. Three samples from CTD024 off the southern tip of Greenland, which were filtered using 0.45 μ m filters, have similar values to those collected off Nuuk.

Dissolved $^{230}\mathrm{Th_{xs}}$ concentrations range from 0.9 to 4.5 fg/kg and generally increase with depth from the surface (Figure 3b). Below $\sim \! 100$ m, concentrations increase in a linear fashion, while above 100 m the gradient with depth is steeper as concentrations decrease toward the surface. Two samples measured at the same depth at CTD004 are within 2σ uncertainty of each other. Below $\sim \! 100$ m at the northern Nuuk transect, concentrations are slightly higher offshore (cf. CTD006 and CTD004), while concentrations are invariant with distance from the coast at the southern transect. Concentrations at Cape Farewell (CTD024) are similar to those at Nuuk at the same depths and also increase with depth. Shallower than 100 m the DY081 data are similar to those reported from the GEOVIDE cruise (Deng et al., 2018), around 1–2 fg/kg near the surface, but below 100 m DY081 data are generally lower at equivalent depths, by $\sim \! 0.5$ fg/kg for data near Cape Farewell and by $> \! 1$ fg/kg for stations in the central Labrador sea (Deng et al., 2018; Moran et al., 1997, 2002).

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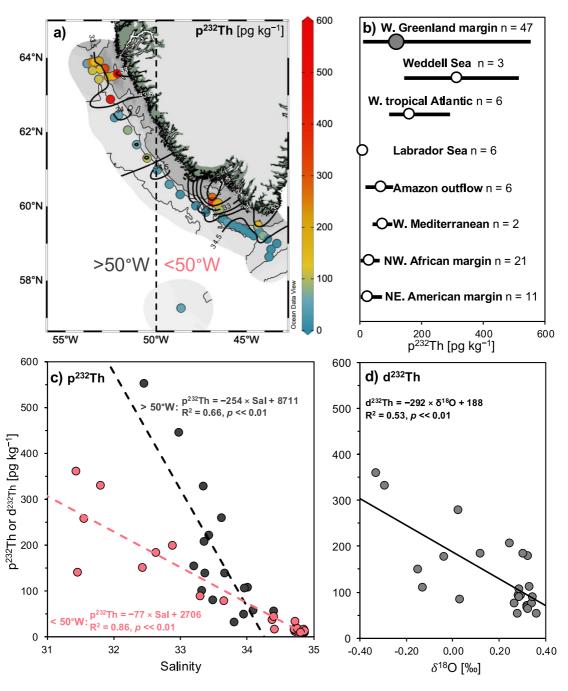


Figure 2. (a) Surface concentration of p^{232} Th (pg kg⁻¹) is shown on the color scale. Black contours show the average salinity of underway samples. (b) Compilation of p^{232} Th concentrations (<500 m depth): symbols = average, bars = range and n = number of samples. Data are from: the Weddell Sea (Venchiarutti et al., 2011); the western tropical Atlantic (station 2; Hayes et al., 2017); the Labrador Sea and near the Amazon outflow (Moran et al., 2002); the western Mediterranean (Gdaniec et al., 2018); the northwest African and northeast American margins (Hayes et al., 2018). (c) Correlations between salinity and p^{232} Th for surface underway samples: black symbols > 50°W; red symbols < 50°W. (d) Correlation between dissolved p^{232} Th concentrations (all depths, pg kg⁻¹) and p^{232} Th concentrations (all de

3.3. Thorium-230-Derived Residence Times and Dissolved ²³²Th Fluxes

Residence times of thorium (τ_{Th}) range from 1.6 to 6 years, and generally increase with depth from the surface, although the shallowest three data points from CTD004 show a reversal of this trend in the top 50 m (Figure 3). The three sites in the northern Nuuk transect show a spread in τ_{Th} at <100 m, with values converging below this depth.

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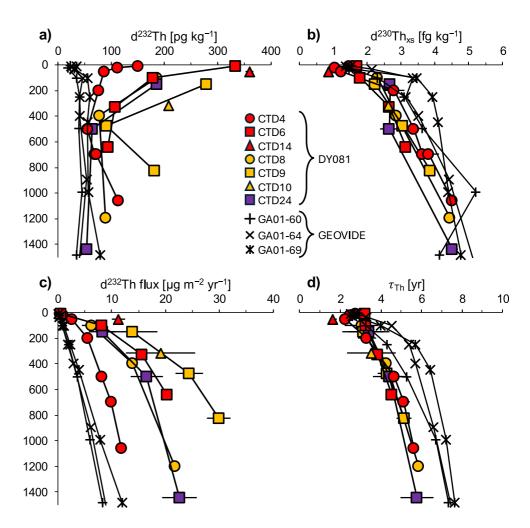


Figure 3. Concentrations (a), (b) of dissolved 232 Th and 230 Th $_{xs}$, integrated 232 Th flux (c) and 230 Th residence time (d) near Nuuk and Cape Farewell during the DY081 cruise. The deepest samples from each profile are within 30 m of the seafloor except for CTDs 8 and -9 (deepest samples 486 and 489 m above the seafloor). Data from the GEOVIDE cruise (black symbols, Deng et al., 2018) are also shown; only GEOVIDE data within the depth range of DY081 samples are shown. Residence times and 232 Th fluxes from GEOVIDE data (Deng et al., 2018) are calculated assuming 238 U = 3.15 ng/g and 234 U/ 238 U activity ratio of 1.147.

Integrated fluxes of d^{232} Th increase sharply with depth and range from 0.3 to $10 \,\mu g \, m^{-2} \, yr^{-1}$ in the surface 50 m and 8–30 $\mu g \, m^{-2} \, yr^{-1}$ at depths >300 m (Figure 3). Fluxes at different CTD sites converge toward the surface and diverge at greater depths, with steeper gradients with depth in the upper 500 m. At both Nuuk transects, fluxes are generally higher closer to the coast. Fluxes from CTD024 (Cape Farewell) lie within the range of fluxes at Nuuk.

3.4. Sedimentary 230 Th and 232 Th Concentrations and 230 Th-Normalized Fluxes

Thorium-232 concentrations in core-top sediments range from 3.48 to 10.76 $\mu g~g^{-1}$, (average 4.91 \pm 1.74, 1σ), indicating that a substantial portion of the sediment is lithogenic (upper continental crust $\sim\!10.5~\mu g~g^{-1}$; Rudnick & Gao, 2003). Total 230 Th (activity) concentrations ranged from 0.56 to 1.44 dpm g^{-1} . Due to the large lithogenic fraction and shallow water depth of the sample sites, the proportions of unsupported 230 Th (230 Th $_{xs}$) are low and sensitive to the assumed values of 238 U/ 232 Th and 230 Th/ 238 U ratios (activity ratios of 0.45 and 0.816 respectively) gives 230 Th $_{xs}$ concentrations from 0 to 1.04 dpm g^{-1} , which corresponds to 230 Th normalized mass fluxes of 23–120 g m $^{-2}$ yr $^{-1}$, and 232 Th fluxes 105–711 $\mu g~m^{-2}$ yr $^{-1}$ (ignoring 230 Th $_{xs}$ values within error of zero). The range of 232 Th fluxes changes to 91–412 $\mu g~m^{-2}$ yr $^{-1}$ (238 U/ 232 Th = 0.3; 230 Th/ 238 U = 0.75) and 133–944 $\mu g~m^{-2}$ yr $^{-1}$ (238 U/ 232 Th = 0.6; 230 Th/ 238 U = 0.95) when changing the lithogenic correction factors. Our choice of lithogenic

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correction factors and the sensitivity of ²³²Th fluxes is detailed in the (Figure S1 and Table S1 in Supporting Information S1). We note that the uncertainty introduced to ²³²Th fluxes by chosen lithogenic ratios will likely be systematic for a given location because the sediments are of similar age and share a similar lithogenic source. Because of this uncertainty, we interpret the range of ²³²Th fluxes calculated across our multiple sampling sites.

4. Discussion

4.1. High Lithogenic Signals Linked to Glacial Waters

The spatial pattern of surface water p^{232} Th concentrations (Figure 2a) highlights key areas of very high lithogenic particulate load at the Greenland margin. As might be expected, the (>0.45 µm) particulate concentrations are greater than those observed in the open ocean (e.g., Moran et al., 1997) and show that large amounts of lithogenic particles reach the waters overlying the continental shelf and slope. The association of high p^{232} Th with low-salinity surface waters suggests a strong link between glacially derived waters and particulate lithogenic material, and suggests a sufficiently long residence time for lithogenic particles to maintain quasi-conservative behavior in the shelf-slope region surface waters (Figure 2c). This behavior is consistent with the rapid transport of fjord-derived particles across the shelf (Figure S2 in Supporting Information S1). The glacial origin of the low salinity waters is further supported by a strong correlation between δ^{18} O and salinity in surface waters (0–10 m; $R^2 = 0.95$; Hendry et al., 2019). This association between glacially derived waters and high lithogenic contents is also supported by the d^{232} Th data, with moderate correlations between δ^{18} O (and salinity) and d^{232} Th concentrations across all sample depths (Figure 2d).

It is possible that the relationship between freshwater tracers and ²³²Th is a distal signal advected from the Arctic or East Greenland in the boundary current system. However, we observed two distinct relationships between salinity and p²³²Th at our sampling locations, which suggests that more localized processes may alter the lithogenic loading per unit freshwater in different source regions/catchments (e.g., differences in glaciation style, geomorphology, lithology, particle removal rates and hydrography of fjord systems). The steeper gradient for samples >50°W (Figure 2c) implies a higher relative lithogenic content for meltwaters derived from around Nuuk than from the sites further south and east. This result is consistent with a recent study that calculated that the outlet from SG into Sermilik fjord (Figure 1b), (~50 km from our highest p²³²Th concentration site), is thought to contribute ~25% of the suspended sediment flux from the entire Greenland ice sheet (Overeem et al., 2017). Indeed, satellite images show plumes of material emanating from the fjords toward our sampling sites (Figure S2 in Supporting Information S1), sometimes crossing the shelf within ~2 days (Supporting Information S1). Such rapid transport of particles across the shelf could explain how the different p²³²Th versus salinity correlations are maintained, despite the strong boundary current (i.e., the transport time in the WGC for ~680 km between Cape Farewell and the sampling grid at Nuuk at 0.35 m/s is ~26 days).

Local variations in geology may also influence the amount of ²³²Th exported from a catchment. Stream sediments enriched in lanthanum (with which Th correlates; Rudnick & Gao, 2003; Steenfelt, 2012) and U/Th mineralization (Armour-Brown et al., 1983) both argue for elevated ²³²Th in source lithologies around Southern Greenland (i.e., closer to our sites <50°W). Likewise, younger rocks around the southern tip of Greenland may have higher ²³²Th concentrations than Archaean rocks dominant in the area around Nuuk (Henriksen et al., 2009; Taylor & McLennan, 1995). Therefore, lithological influences probably act to reduce p²³²Th concentrations at sites >50°W; higher sediment inputs (Overeem et al., 2017) must outweigh these factors to produce the steeper relationship between p²³²Th and salinity at these sites.

Resuspension of sediment at the ocean floor can elevate dissolved and particulate 232 Th concentrations in the water column (Hayes et al., 2018; Schlitzer et al., 2018). Elevated turbidity is measured at ~170 m and 350–460 m, toward the head of the Nuuk trough (CTD16 and -15; Figures 1b, Figure S6 in Supporting Information S1), but this signal is not seen at our sample locations further offshore (CTD8, -9, -10), or at the northern Nuuk transect (CTD4, -6, -14; Figure 1). We are therefore confident that the majority of particulate 232 Th is supplied at or near the surface. The shallow maxima in 232 Th concentrations are also consistent with a major source at the surface. However, there are hints of a moderate subsurface shelf/slope source of 232 Th at CTD4 and -9, where 232 Th concentrations increase below ~500 m (Figure 3). It is possible that 232 Th sourced from sediment resuspension (or released from pore-waters during resuspension; Cochran et al., 1986) could be transported along isopycnals toward CTD08 and -09, explaining some of the elevation of 232 Th toward the surface at those sites.

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4.2. Lithogenic Signals at the Greenland Margin in a Global Context

4.2.1. Dissolved and Particulate ²³²Th Concentrations

The highest DY081 p^{232} Th concentrations from close to Nuuk and Narsaq are an order of magnitude, or more, higher than surface open ocean measurements from the Labrador Sea (Figure 2b; Moran et al., 1997, 2002). The average surface p^{232} Th concentration oceanward of the 1,000 m isobath at Nuuk, ~88 pg/kg, is high compared to previous surface measurements in the central Labrador Sea, ~10 pg/kg (Moran et al., 2002). Compared to other regions with high lithogenic input, DY081 surface p^{232} Th concentrations are still considerable (Figure 2b). For example, in the western tropical Atlantic, close to the Amazon outflow, concentrations are 62–108 pg/kg (Moran et al., 2002) and in the western Mediterranean are up to 106 pg/kg at 100 m, although <50 pg/kg in general (filtered onto 1 μ m filters; Gdaniec et al., 2018). At the east Atlantic margin p^{232} Th concentrations are <20 pg/kg (at depths <200 m in the 0.8–51 μ m size fraction), while those at the northwest Atlantic margin offshore North America are up to ~70 pg/kg (Hayes et al., 2018). Close to the West Antarctic Peninsula, similarly high p^{232} Th concentrations, up to ~300 pg/kg, have been measured in waters ~200 m (Venchiarutti et al., 2011).

The DY081 d²³²Th concentrations are also high compared to the east Atlantic margin, up to 175 pg/kg, under the influence of high fluxes of Saharan dust (<50 m depth; Hayes, Anderson, et al., 2018) and as high as surface waters influenced by both dust and river inputs, up to ~230 pg/kg, from the western tropical Atlantic (Hayes et al., 2017). Close to the glaciated West Antarctic peninsula, d²³²Th concentrations are 100–110 pg/kg at their subsurface maximum ~150–180 m water depth (Venchiarutti et al., 2011). The evidence, therefore, from both dissolved and particulate ²³²Th concentrations suggests a strong contribution of lithogenic material to seawater chemistry at the Greenland margin, comparable to other areas of high lithogenic inputs. This observation is consistent with satellite data, indicating a high input of suspended sediment from glacial outflows (Overeem et al., 2017) and importantly suggests that elevated lithogenic loads are not completely trapped in fjord systems.

The GEOVIDE sites GA01-60 and -64 (Deng et al., 2018), are ~70-100 km offshore (Figure 1), oceanward of the 1,500 m isobath off the southern tip of Greenland, and show much lower surface d²³²Th surface concentrations, suggesting that waters with high lithogenic content may be confined quite close to the shelf around Cape Farewell. Differences in ²³²Th depth profiles moving offshore are also seen between the West Antarctic Peninsula and Drake Passage (Venchiarutti et al., 2011) and such differences between open-ocean and margin ²³²Th profiles have also been predicted by modeling due to water mass exchange (Roy-Barman, 2009). The convergence of some GEOVIDE and DY081 ²³²Th concentrations below ~500 m (e.g., CTD24) indicates that either scavenging or water-mass exchange may reduce the spatial differences at these greater depths.

4.2.2. Dissolved ²³²Th Fluxes

The calculated ²³²Th scavenging fluxes indicate a strong source of lithogenic material derived from the continent (Figure 3). Compared to vertical fluxes calculated from the data of Deng et al. (2018) around Cape Farewell and the central Labrador Sea (GEOVIDE cruise), DY081 data are elevated at all depths except the top ~50 m, where the fluxes converge. Below ~200 m the DY081 fluxes are up to ~3x higher (at equivalent depths) than those typically observed in the open ocean, (<5.5 µg m⁻² yr⁻¹ in the South Atlantic and North Pacific; Deng et al., 2014; Hayes et al., 2013) (Figure 4a). This is not unexpected for a continental margin setting, but simply demonstrates that scavenged ²³²Th fluxes at the West Greenland margin are indeed elevated and that the region is influenced by a horizontal supply of glacially derived lithogenic trace-metals that extends into the Labrador Sea, and is not confined to fjords only (Hopwood et al., 2015). Furthermore, ²³²Th fluxes at the West Greenland margin are comparable to, or higher than, those observed at continental margins influenced by some of the world's largest lithogenic sources: Saharan dust and the Amazon and Orinoco Rivers (Figures 4a and 4c; Hayes et al., 2017; Hayes, Anderson, et al., 2018). The depth-trend in ²³²Th fluxes is probably a result of a violation of one or more of the scavenging model assumptions (Supporting Information S1); explaining these trends is a topic of active research and although there is no consensus on an optimum integration depth, most studies consider depths ~50-500 m for estimating dust inputs (Anderson et al., 2016; Deng et al., 2014; Hayes et al., 2013, 2017; Pavia, Anderson, Winckler, & Fleisher, 2020). We present full ²³²Th flux depth profiles for comparison to the literature, but expect fluxes at depths <100 m to be more severely impacted by violations of model assumptions (Supporting Information S1) and so consider a set range of integration depths in our discussion of Fe supply (Section 4.3.1).

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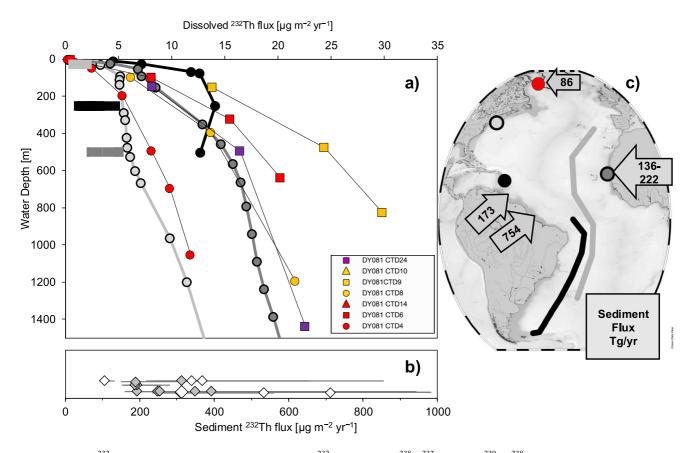


Figure 4. (a) Dissolved 232 Th fluxes (thin lines) and (b) core-top sediment 232 Th fluxes using 238 U/ 232 Th = 0.45, 230 Th/ 238 U = 0.816 (diamonds: gray = Southwest Greenland, white = Nuuk; distributed vertically for clarity) from the West Greenland margin. Dissolved 232 Th fluxes (a) from near the North America (light gray), Africa (dark gray), South America (black) are also shown (greyscale circles, thick lines; Hayes et al., 2017; Hayes, Anderson, et al., 2015, Hayes et al., 2018). Openocean 232 Th fluxes at fixed integration depths are shown (solid bars) from the Atlantic (25 m light gray), Hsieh et al., 2011; 250 m black, Deng et al., 2014) and North Pacific (500 m, dark gray, Hayes et al., 2013, not shown on map). The map shows the compiled sites from the Atlantic with estimates of terrigenous sediment input from the Amazon and Orinoco rivers (Peucker-Ehrenbrink, 2018), Saharan dust (Yu et al., 2019) and glacial sediment input around Nuuk (Overeem et al., 2017) in Tg/yr. Error bars on sediment 232 Th fluxes indicate the range of fluxes by varying lithogenic activity ratios between 238 U/ 232 Th = 0.3–0.6 and 230 Th/ 238 U = 0.75–0.95 while keeping 230 Th_{xs} > 0 (the full range of possible sediment 232 Th fluxes is shown in Figure S1 in Supporting Information S1). Dissolved Fe fluxes estimated in this study are directly proportional to 232 Th fluxes (for S_{Fe/Th} = 1 and Fe/Th = 3,271 g/g: Fe/ 232 Th ~ 3.3 mg/μg ~ 60 μmol/μg. For S_{Fe/Th} = 0.4: ~1.3 mg/μg ~ 23 μmol/μg).

The increased proximity of DY081 sites to the continent compared with the literature sites (\sim 40–50 vs. \sim 200 km at West African margin) could partly explain higher 232 Th values. A second line of evidence for elevated lithogenic fluxes at our sites comes from core-top sediments, with average total 232 Th fluxes, 312 \pm 148 µg m $^{-2}$ yr $^{-1}$, (\geq 2x late-Holocene 230 Th-normalized 232 Th fluxes at the South American and African margins; Rowland et al., 2021). Multiplying the average total 232 Th fluxes by an estimate of S_{Th} \sim 3–5% (Baker et al., 2020; Hayes et al., 2013) gives dissolved fluxes similar to our scavenging estimates at \sim 500 m (i.e., \sim 10–15 µg m $^{-2}$ yr $^{-1}$). A further check on our flux estimates can be made using the suspended sediment flux estimated for glaciers around Nuuk from satellite data (Overeem et al., 2017). Assuming an exponential decrease in flux with distance, we estimate \sim 1% of the suspended sediment flux from glaciers could reach our sites (Text S4 in Supporting Information S1), giving a total possible 232 Th flux of \sim 9.2 \times 10⁶ g 232 Th/yr. Distributing this flux over a square 100²–150² km² around our sites suggests a total potential vertical 232 Th flux of \sim 410–920 µg m $^{-2}$ yr $^{-1}$, generally higher than our core-top 232 Th fluxes (Supporting Information S1).

4.2.3. Do Physical Transports Lead to Overestimates of d²³²Th Supply?

The vertical gradients of 230 Th and 232 Th in the upper \sim 250 m will act to transport 230 Th into the surface layer, and 232 Th out of the layer, leading to underestimates of 232 Th removal fluxes in each case (Supporting Information S1). Boundary scavenging near ocean margins (Anderson et al., 1983) may also mean that our dissolved and

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sedimentary 232 Th fluxes are too low (e.g., only \sim 70% of their true values; Costa et al., 2020; Hayes, Fleisher, et al., 2015).

The strong near-surface 232 Th gradient across the shelf and strong currents at our sites could introduce additional terms to 232 Th mass balance that are not accounted for by our model. Near Nuuk we calculate a potential advective supply of 232 Th in the upper 50 m across the shelf that is >10x larger ($\sim 380~\mu g~m^{-2}~yr^{-1}$) than our highest scavenging removal fluxes (Supporting Information S1). Although we expect much of this supply to be removed along the shelf by the WGC (Supporting Information S1), the size of the potential advective supply implies that total 232 Th supply at our sites could be larger than we estimate from scavenging removal alone, and is possibly dominated by advection instead of particle dissolution (Supporting Information S1).

Such large advective fluxes imply short ²³²Th residence times, raising the possibility that measured ²³²Th concentrations are not at steady state, but could represent a more transient elevation (e.g., due to input from fjords during the melt season). In such a scenario, scavenging removal fluxes would be overestimates of the long-term mean. However, given the additional ²³²Th potentially supplied by advection, and the agreement with our estimates calculated using alternative methods (4.2.2), we consider it unlikely our scavenging based fluxes overestimate the total ²³²Th supply.

4.3. Fe Fluxes and Biological Requirements

4.3.1. Fe Fluxes at the West Greenland Margin and Globally

The vertical removal flux of d²³²Th has been used to estimate dissolved Fe fluxes across a range of oceanographic settings including the tropical Pacific, western tropical Atlantic and across the North Atlantic (Hayes et al., 2013, 2017, 2018; Hayes, Fitzsimmons, et al., 2015). These studies have mainly focused on assessing aeolian supply to open ocean areas. The approach assumes that Fe and ²³²Th are both supplied predominantly from the dissolution of lithogenic material at a fixed ratio such that variation in Th inputs scale with Fe supply (i.e., it is assumed that no sources exist that supply Th without associated Fe, and vice versa).

In our glacial margin setting the source of 232 Th is dissolution of lithogenic material, so we assume an Fe/Th ratio of 3,271 g/g, a composition typical for the upper continental crust (Hayes, Fitzsimmons, et al., 2015; Taylor & McLennan, 1995). For the area around Nuuk, the presence of dominant Archaean rocks (Henriksen et al., 2009) means that the ratio may be substantially larger (> 10^4 g/g; Taylor & McLennan, 1995), making our Fe fluxes conservative for this region. The ratio of Fe:Th fractional solubility ($S_{Fe/Th}$) was estimated to be 1 by earlier studies (Hayes, Fitzsimmons, et al., 2015), but more recent leaching experiments on Saharan dust imply that Fe and Th are not released congruently, suggesting a wider range of $S_{Fe/Th}$ (0.4–1.3; Hayes, Anderson, et al., 2018). Independent of the $S_{Fe/Th}$ and Fe/Th values, our iron fluxes represent a lower bound as they are derived from 232 Th fluxes that are likely underestimated due to the local hydrography (Supporting Information S1).

The average dissolved Fe flux across all sites ranged from 16 to 52 mg m $^{-2}$ yr $^{-1}$ for solubility ratios of 0.4 and 1.3, respectively (279–906 µmol m $^{-2}$ yr $^{-1}$; Table S2 in Supporting Information S1). As the Fe fluxes are proportional to 232 Th fluxes they share the same spatial patterns and comparison to other global sites in Figure 4 (Figure S5 in Supporting Information S1). Due to violations in scavenging-model assumptions at shallow depths (Supporting Information S1; Pavia, Anderson, Winckler, & Fleisher, 2020), we consider Fe fluxes at greater integration depths (100–200 and 200–500 m) to give better estimates (Table S2 in Supporting Information S1; Figure 5). Considering these depths, our lower bound for iron fluxes at the West Greenland margin is comparable to those found in other high lithogenic input settings (Figure 4; Table S2 in Supporting Information S1), such as near the Amazon River (Hayes et al., 2017) or under the Saharan dust plume (Hayes et al., 2018).

If supplies of Fe and Th become decoupled before reaching our study sites, for example, if Fe is released from sediments due to reductive dissolution (Tonnard et al., 2020), then this method will underestimate the fluxes of Fe. Despite these uncertainties, our flux estimates provide the first quantitative constraints on the supply of Fe from dissolving lithogenic particles at the West Greenland margin. As lithogenic particles are supplied laterally (Figure 2), our Fe flux estimates constrain the horizontal Fe supply that must balance the vertical removal from West Greenland.

We also make an additional estimate of dissolved Fe supply based on our core-top 232 Th fluxes and an estimate of fractional Fe solubility (S_{Fe}). We estimate S_{Fe} as 2.55%, from the average of two Alaskan glacial flour samples

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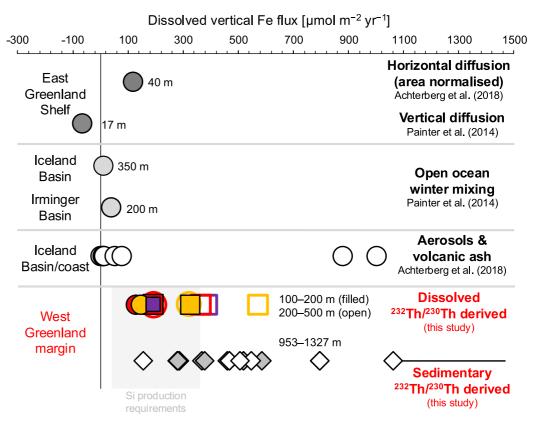


Figure 5. Estimated dissolved Fe vertical fluxes in the high-latitude North Atlantic from different sources. Fe flux estimates from dissolved 232 Th/ 230 Th are calculated for depths 100–200 m (filled symbols) and 200–500 m (open symbols) using $S_{Fe/Th}=0.4$ and Fe/Th = 3,271 g/g. Dissolved Fe flux estimates derived from core-top sediment 232 Th/ 230 Th for Nuuk (white diamonds) and Southwest Greenland (gray diamonds) were calculated using $S_{Fe}=2.55\%$ and Fe/Th = 3,271 g/g, and lithogenic activity ratios 238 U/ 232 Th = 0.45 and 230 Th/ 238 U = 0.81. Error bars show fluxes when estimated with 238 U/ 232 Th = 0.3–0.6 and 230 Th/ 238 U = 0.75–0.81 (the error bar for the lowest flux is smaller than the symbol size). The possible range of Fe demand by siliceous primary producers is indicated by a gray shaded region (main text, Section 4.3.2). Horizontal fluxes at the East Greenland shelf were normalized to an approximate shelf-area in the original study (Achterberg et al., 2018). Negative fluxes indicate net Fe removal. The highest Fe fluxes from aerosols include direct input from a volcanic eruption during Spring 2010 (Achterberg et al., 2018). Water depths over which fluxes are calculated or integrated are shown.

(Schroth et al., 2009). Although the S_{Fe} estimate is based on samples from a different location, primary Fe(II) silicates contribute most of the Fe in both the Alaskan and DY081 sediments (Fe(II)/total Fe ~35–80%; Shoenfelt et al., 2019); this likely exerts a strong influence on Fe solubility (Hawkings et al., 2018). Our dissolved Fe flux estimates based on these assumptions and core-top data range from 156 to 1,062 μ mol m⁻² yr⁻¹ (average 466 \pm 222 μ mol m⁻² yr⁻¹, 1 σ), consistent with significantly elevated Fe fluxes at our sites (Figure 5).

4.3.2. Do Physical Transports Bias Fe Supply Estimates?

Because the supply of 232 Th could be dominated by advection (4.2.3), rather than particle dissolution, it is possible that the supply of Fe and Th become decoupled along the advection pathway (Hayes et al., 2018), and our model overestimates Fe supply (Equation 3). However, because the potential advective 232 Th flux is \sim 10x our supply estimate based on scavenging removal fluxes, our estimates of Fe supply would remain conservative unless iron was removed relative to Th by the same factor (i.e., $>\sim$ 10x).

Dissolved Fe and Th are both expected to be strongly removed across salinity gradients in fjords; data from the Baltic Sea (for Th) and the fjords around Nuuk (for Fe) indicate this removal process occurs at a similar salinity for both elements (\sim 10–15) (Andersson et al., 1995; Hopwood et al., 2016), with concentrations decreasing by a factor of 18 (Fe) and 28 (Th). Additionally, in particle-rich coastal and estuarine environments, Fe and Th have been shown to have very similar short residence times of \sim 1–20 days, suggesting rapid removal of both elements

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at similar rates (McKee, 2008; Santschi et al., 1980; Shen et al., 2024). Thus, it seems unlikely that the large lateral advective flux of d²³²Th (or Fe) could be supported by the direct export of d²³²Th and Fe from fjords, and that the elements would not be expected to fractionate strongly even in such a scenario. Instead, the very high concentrations of p²³²Th and the high ²³²Th fluxes measured in core-top sediments both suggest the dissolution of lithogenic particles as an important supply term for ²³²Th (and Fe) at our sites.

4.3.3. Fe Supply and Uptake at Nuuk

The Fe fluxes presented in this study are comparable to, or higher than, other sites situated in areas of high lithogenic inputs measured using the same methods (e.g., the Amazon river and dust inputs; Hayes et al., 2017; Figure 4). This supports the notion that fluxes of lithogenic material from Greenland are globally significant (Overeem et al., 2017) and are likely to be of importance in supplying lithogenic nutrients at least on a regional scale. It is likely, given its continuously glaciated nature, that the rest of the margin supplies lithogenic nutrients to the ocean, though inputs at the western margin are expected to be the largest (Overeem et al., 2017). However, the relative importance of a given flux of Fe in terms of biogeochemistry is determined by the requirements of phytoplankton in the local area. During the DY081 cruise, biogenic silica production was estimated using incubation experiments (Hendry et al., 2019) to be \sim 2–14 mmol m⁻² day⁻¹ around Nuuk. These estimates made during summer may well represent an upper estimate for the annual productivity at these sites (due to favorable conditions for diatom growth), but they can be used to assess the importance of the Fe fluxes calculated here to a first approximation.

Using typical diatom molar uptake ratios of Si:C ~ 0.13 (Brzezinski, 2004) and C:Fe $\sim 10^5$ as estimated by Tagliabue and Arrigo (2005) for diatoms in the Ross Sea (which is also at high latitude, surrounded by glaciated continental margins and likely has high Fe input) we can then estimate the Fe supply required by diatoms around Nuuk to sustain the measured biogenic silica production. Although this approach has caveats, including the poorly constrained stoichiometry of nutrient uptake by diatoms—which is dependent on the degree of Fe stress amongst other factors (Hutchins & Bruland, 1998; Koch & Trimborn, 2019)—our calculations allow a first approximation of the Fe requirements of diatoms at our sample sites.

Scaling up the measured Si productivity to an annual productivity estimate gives a range of Fe supply required by diatoms around Nuuk of ~60-400 µmol m⁻² yr⁻¹ (3-22 mg m⁻² yr⁻¹), which, the large uncertainties notwithstanding, overlaps the range of average estimates of Fe flux calculated here. Assuming a shorter, perhaps more realistic, growing season length of ~8 months at our sites (Arrigo et al., 2017) indicates required Fe fluxes of \sim 40–260 µmol m⁻² yr⁻¹. The supply of dissolved Fe from the lithogenic material is thus likely to contribute a dominant proportion of the Fe required for diatom growth at Nuuk, even when considering the lower value of S_{Fe/} The (Figure 5). Additional supply of Fe due to upwelling or due to diapycnal diffusivity is also possible; however, upwelling can be suppressed during the spring and summer due to stronger stratification (Arrigo et al., 2017; Oliver et al., 2018), and the dominant wind directions are seldom favorable for upwelling (Hopwood et al., 2015). Vertical diffusivity has been shown to supply only small amounts of Fe to the surface mixed layer in the Irminger basin (Achterberg et al., 2018), and leads to removal of Fe at the East Greenland shelf (Painter et al., 2014). Deeper mixing supplying dissolved Fe from greater depths may become more important during the winter (Achterberg et al., 2018). Thus, the flux of Fe from the dissolution of lithogenic material seems likely to account for the most important supply term. This conclusion is supported by the fact that our chosen values for UCC Fe/Th and S_{Fe/Th} (Figure 5) and the effects of lateral and vertical diffusion (Supporting Information S1) mean that our Fe estimates probably represent a lower bound.

4.3.4. Regional Context of Fe Supply

Our aim is to assess whether lithogenic fluxes are particularly elevated at the Greenland margin, so that we choose values for $UCC_{Fe/Th}$ and $S_{Fe/Th}$ that are conservative with respect to the Fe flux magnitude, allowing us to provide a lower bound on these fluxes for our sites.

A previous study calculated and compiled Fe fluxes to surface waters in the Irminger and Iceland basins from a range of processes, including aeolian deposition, horizontal supply from shelves, vertical mixing and diapycnal diffusivity (Achterberg et al., 2018). A horizontal Fe flux estimate for the East Greenland shelf—converted to a vertical flux equivalent by Achterberg et al. by normalizing to a surface area—is 117 μ mol m⁻² yr⁻¹, lower than our vertical Fe fluxes estimated for the West Greenland margin (>128 μ mol m⁻² yr⁻¹ for S_{Fe/Th} = 0.4; Table S2 in

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Supporting Information S1; Figure 5). The dissolved Fe fluxes we reconstruct for the West Greenland margin from sediments and seawater are higher than the other fluxes calculated or compiled by Achterberg et al., except for the input of Fe-rich ash from a volcanic eruption (Figure 5). This finding highlights the potential significance of the large glacially derived lithogenic supply to the nearby coastal ocean.

Achterberg et al. concluded that the large fluxes measured at the East Greenland shelf could not propagate into the basin interior (i.e., limited to <100 km) due to strong density gradients in the East Greenland current. For our sites on the west Greenland margin, isopycnals are inclined downwards toward the continental slope and flatten at shallower depths toward the open ocean hundreds of kilometers offshore (Lee, 2007; Sarthou et al., 2018; Figure S6 in Supporting Information S1). Combined with high horizontal eddy diffusivity in the region, the density structure could provide a mechanism for the mixing of sub-surface margin waters (i.e., ~100–300 m), with a strong lithogenic imprint, toward the surface waters of the interior of the Labrador sea (Funk et al., 2009; Kawasaki & Hasumi, 2014; Figure S6a in Supporting Information S1). Thus, the Fe fluxes at the West Greenland Margin may be more significant in supplying Fe to the open-ocean Labrador Sea than holds for the equivalent setting on the East Greenland Margin for the Irminger basin. Likewise, GEOVIDE cruise results show elevated particulate Fe concentrations and fluxes off the West Greenland margin compared with the eastern margin (Gourain et al., 2019; Lemaitre et al., 2020).

5. Conclusion and Outlook

In this study, we have presented some of the first estimates of lithogenic fluxes to the ocean at the West Greenland margin. Concentrations of the isotope ²³²Th suggest that a significant lithogenic source accompanying low-salinity melt waters is transported offshore from Nuuk; concentrations of both p²³²Th and d²³²Th are as high or higher than at other sites with high lithogenic input. Using measurements of ²³²Th/²³⁰Th in filtered seawater and sediments, we show that vertical ²³²Th fluxes in our study region are high, though not unprecedented, when compared with other continental margins. Our dissolved ²³²Th fluxes, although likely to be underestimates (Supporting Information S1), are much higher than those in the open-ocean, and the elevated ²³²Th concentrations and fluxes reach oceanward of the 1,000 m isobath. These results confirm that the Greenland margin, and possibly glaciated margins more generally, do act as important sources of lithogenic material (including micronutrients) to the ocean, and that the estuarine process in fjords does not completely remove the high glacial inputs at these sites (Arrigo et al., 2017; Bhatia et al., 2013; Hopwood et al., 2015, 2020). Our core-top ²³⁰Th-normalized ²³²Th fluxes also provide a modern reference point for studies of changing glacial lithogenic fluxes through the late Pleistocene (e.g., Zhou & McManus, 2024).

Our ²³²Th/²³⁰Th data suggest that fluxes of Fe at the West Greenland margin are higher than those at the East Greenland shelf (Achterberg et al., 2018). We calculated that the Fe fluxes from lithogenic inputs are probably large enough to support all of the silica productivity measured during the DY081 cruise, confirming that glacially derived lithogenic material can be important for sustaining primary productivity in the region (Ng et al., 2024). We suggest that high lithogenic inputs combine with local hydrography to advect the signals toward the basin interior, potentially supplying lithogenic nutrients off the shelf to primary producers in the Labrador Sea. Thus, our results support the more general hypothesis that glacially derived lithogenic nutrients do supply open-ocean ecosystems (Hawkings et al., 2020; Wadham et al., 2019).

The application of our method in a setting with very dynamic physical oceanography means our estimates would benefit from comparison to future independent estimates, such as sediment traps or ²³⁰Th-normalization of filtered particles (Anderson et al., 2016). These kinds of supporting data may become available in the future as the high-latitude oceans and glaciated continental margins remain in focus as key areas of global change.

Conflict of Interest

The authors declare no conflicts of interest relevant to this study.

Data Availability Statement

All the data on which this study is based can be found in Rowland (2025) and Hendry et al. (2024).

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