Tracking the provenance of Greenland-sourced, Holocene aged, individual sand-sized ice-rafted debris using the Pb-isotope compositions of feldspars and \(^{40}\text{Ar}/^{39}\text{Ar}\) ages of hornblends

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**ABSTRACT**

The provenance of sand-sized ice-rafted debris (IRD) sourced from Greenland is currently difficult to determine. Such knowledge, if it could be ascertained with a high degree of certainty, could be applied to the Greenland-proximal marine records to improve both our understanding of modern-day spatial patterns of iceberg rafting and the past history of the Greenland Ice Sheet (GIS). Recent studies have highlighted the utility of the Pb-isotope composition of individual sand-sized feldspars and the \(^{40}\text{Ar}/^{39}\text{Ar}\) ages of individual sand-sized hornblends in this regard. However, before any such provenance toolkit can be applied to the palaeo-record, it is necessary first to determine whether this approach can be used to track the sources of known recent Greenland-proximal IRD deposition. To this end we present new records of the Pb-isotope composition and the \(^{40}\text{Ar}/^{39}\text{Ar}\) ages of individual sand-sized grains of feldspars and hornblends, respectively, from modern Greenland glacifluvial and fjord sands and Holocene to modern Greenland-proximal marine sediments. These new data demonstrate that sand-sized feldspars and hornblends glacially eroded by the GIS exhibit distinct intra- and inter-tectonic terrane differences in their Pb-isotope compositions and ages and that these differences are clearly expressed in the geochemistry and geochronology of sand-sized IRD deposited in marine sediments around Greenland. Although overlap exists between some Greenland-proximal IRD ‘source fields’ defined by these data, our approach has the potential to both better understand spatial patterns of Greenland-derived IRD in the modern day as well as during past episodes of iceberg calving.

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1. Introduction

The Arctic is one of the fastest warming regions on Earth (Bindoff et al., 2013). It also includes the Greenland Ice Sheet (GIS), which, if it were to melt completely, would contribute ~7.3 metres of global sea-level rise (Meehl et al., 2007). Understanding how the GIS will respond to anthropogenically-induced global warming over the coming century is therefore important. This need is further highlighted by the fact that glacial isostatic adjustment complicates accurate prediction and associated mitigation since the impact of deglaciation of the GIS and the West Antarctic Ice Sheet (also under threat from future melt-reduction and/or collapse; Carlson and Winsor, 2012) on sea-level would be felt differently in both near and far field regions (e.g. Mitrovica et al., 2009).

Current scenarios depicting the extent to which the GIS will deglaciate in response to future warming are heavily dependent on the capabilities of numerical climate models, which are embedded with large uncertainties (i.e. Otto-Bliesner et al., 2006). The current generation of climate models lack the capacity to accurately model melt- and growth-histories and basal dynamics of...
continental ice-sheets (Morlighem et al., 2014). To improve prediction of the likely response of the GIS to future warming it is therefore necessary to ground truth model output by simulating continental ice-sheets for a variety of past climates and comparing the results with geological observations (e.g. Thiede et al., 2011; Reyes et al., 2014). Another approach, however, would be to improve our understanding of the dynamic range of the GIS and possibility for threshold melt-reduction behaviour as a function of radiative forcing (due to changes in, e.g., boreal summer insolation and atmospheric CO₂) from an empirical-data perspective by reconstructing the GIS from the geological record for a variety of past warmer and colder-than-present climate states (e.g. Thiede et al., 2011; Reyes et al., 2014).

Reconstructions of the Holocene history of the GIS have considerably advanced our understanding of the relationship between GIS retreat and radiative forcing associated with the last deglaciation. Such studies have benefited from an abundance of spatially diverse geological evidence recorded within both terrestrial (e.g. Bennike and Björck, 2002; NGRIP, 2004; Dyke et al., 2014; Winsor et al., 2015) and GIS-proximal marine (e.g. Knutz et al., 2011; Hogan et al., 2012; Knutz et al., 2013) realms, and reveal that the spatial extent of the GIS reached a minimum by the early Holocene (∼11–7 ka) during peak boreal summer insolation. Our ability, however, to estimate the likely response of the GIS to radiative forcing under future projections for atmospheric CO₂ is limited by insufficient geological-based evidence for GIS extent during older deglacials and interglacials. Recently, evidence in the form of seismic studies (Nielsen and Kuijpers, 2013; Knutz et al., 2015), the provenance of silt-sized terrigenous sediments discharged from southern Greenland to Eirik drift (Reyes et al., 2014) and ⁴⁰K measurements on silt from the bottom of the central Greenland GISP2 ice core (Bierman et al., 2014) has provided new insights into the past history of the GIS prior to the Holocene. One currently underutilised, but complimentary, approach that could be used to enhance our understanding in this respect is to examine the provenance of individual sand-sized ice-rafted debris (IRD) deposited in Greenland-proximal marine settings to estimate past locations of GIS iceberg calving and drift. The Pb-isotope compositions and ⁴⁰Ar/³⁹Ar ages of individual sand-sized feldspars (Gwiadza et al., 1996; Bailey et al., 2012) and hornblends (Hemming et al., 1998; Knutz et al., 2013), respectively, have proven useful for tracking sources of ice-rafted sediments. Our current understanding of the potential sources for sand-sized ice-rafted hornblends and feldspars derived from Greenland is based on knowledge of only the age ranges assigned to individual tectonic terranes (Fig. 1) and on a geographically incomplete compilation of the Pb-isotope compositions of feldspars and ore galenas from bedrock samples (Gwiadza et al., 1996; Bailey et al., 2012). Little is known, however, about the age distribution and Pb-isotope composition of individual sand-sized hornblends and feldspars preferentially incorporated into icebergs sourced from specific iceberg-calving locations on Greenland to-day (Rignot and Kanagaratnam, 2006). To address this gap in our knowledge, we report the Pb-isotope composition of individual feldspars and the ⁴⁰Ar/³⁹Ar ages of individual hornblends from Greenland glaciiluvial sands collected from a series of modern-day sandurs close to a geographically diverse range of iceberg calving locations. In doing so we address the following related questions: 1) What range and diversity exists in the Pb isotope composition and age of IRD incorporated into icebergs from specific GIS calving source regions? 2) What is the Pb isotope composition and age of IRD deposited during the Holocene in Greenland-proximal marine sediments? 3) To what extent can we use such data to reconstruct the provenance of Holocene to modern Greenland-proximal IRD deposition?

2. Geology of Greenland and its major modern iceberg calving sources

Greenland consists of a number of tectonic terranes bearing metamorphic ages ranging from 3.9 Ga to 390 Ma (Fig. 1; Dawes, 2009). The geology of central and southern Greenland is predominantly Precambrian in age and is divided into three regions: the Kettilidan Mobile Belt (KMB), the Archaaean Block (AB) and the Nagssugtoqidian Mobile Belt (NMB) (i.e. Henriksen et al., 2009) (Fig. 1). The AB is primarily composed of Neoarchaean orthogneisses (2.6–3.1 Ga), although Eoarchaean orthogneisses, up to 3.9 Ga in age, occur locally in the region of Nuuk whilst paragneisses, ∼3.8 Ga in age, occur within the Isua Supracrustal Belt (Henriksen et al., 2009). The NMB is an area that is interpreted to represent a reworked marginal portion of the AB as the result of a Palaeoproterozoic (1.7–1.9 Ga) metamorphic overprint. This area lies north of ∼65°N, and comprises the Ammassalik terrane and Nagssugtoqidian orogeny (Connelly and Thrane, 2005). In central west Greenland, in the region around Disko Bugt, the NMB is characterised by the Rinkian fold belt, a several kilometre thick Palaeoproterozoic succession inter-folded with reworked Archaaean gneiss (Grocott and Pulvertaft, 1990). The KMB defines the southern-most tip of Greenland and is composed of granitoids and low- to high-grade metasediments formed within the time period ∼19–1.7 Ga (Henriksen et al., 2009).

Coastal eastern Greenland contains a mountain range that is a relic of the Lower Palaeozoic Caledonian orogeny, composed of thrust sheets and local eclogites within reworked Palaeoproterozoic basement gneisses that formed ∼390–410 Ma ago (i.e. Gilotti et al., 2008) (Fig. 1). The southern region of the Caledonides in the area of Scoresby Sund comprises granodioritic and dioritic plutons that yield intrusive ages of 420–466 Ma (Kalsbeek et al., 2008). The northernmost granites yield intrusion ages of 425–430 Ma (Strachan et al., 2001). Caledonian rocks of Scoresby Sund are separated from NMB rocks outcropping in the region of the Kangerlussuaq Fjord System by Cenozoic basaltic volcanics of the Geikie Plateau (Fig. 1).

Today, major iceberg-calving sources are located in Disko Bugt in the western NMB (∼34.8 km³/yr) and the Scoresby Sund Fjord system in the east Caledonides (∼13.2 km³/yr) (Fig. 1), with these two sources alone contributing ∼13% to the total annual GIS iceberg flux (Weidick, 1995; Rignot and Kanagaratnam, 2006). In east Greenland, the Geikie Plateau supplies the dominant lithic fragments found in sediments deposited in the nearby Kangerlussuaq trough (Alonso-Garcia et al., 2013; Andrews et al., 2014a). Other notable NMB iceberg sources in this region include multiple tidewater glaciers in the Kangerlussuaq Fjord System (∼28 km³/yr) and regions south thereof, including the Hutchinson Plateau, Uummartit Island and Helheimglletscher at the head of Sermilik Fjord (∼23–26 km³/yr; Rignot and Mouginot, 2012) (Fig. 1). The AB is a source of relatively high volumes of icebergs along the southeast (∼46–67 km³/yr, ∼13–19%; Rignot et al., 2004; Rignot and Kanagaratnam, 2006) and southwest (∼60 km³/yr, ∼17%) coasts of Greenland including notable contributions from Eqalorutsit Killit Sermit Sermiaq near Nuuk (∼6–11.5 km³/yr) and Ukaasorsuaq glacier in Sermilik Fjord (∼6–12 km³/yr; Weidick and Bennike, 2007) (Fig. 1).

The East Greenland Current (EGC) exports icebergs from east Greenland clockwise along the landward side of the Denmark Strait (Fig. 1). Along the southern tip of Greenland the West Greenland Current (a mixture of EGC and the relatively warmer North Atlantic Irminger Current) circulates icebergs calved locally from the KMB (notably Qajaquxtap Sermia and Eqalorutsit Killit Sermit tidewater glacier systems at the head of Nordre Sermilik Fjord near Narssarsuaq (∼6 km³/yr; Weidick and Bennike, 2007; Rignot and Kanagaratnam, 2006) in a northerly direction along the
West Greenland coast, where they mix with icebergs calved from the western AB in the Labrador Sea, through eastern Davis Strait and potentially on into Baffin Bay (Fig. 1). Icebergs from Disko Bugt may travel northwards into Baffin Bay, but preferentially move westwards and southwards through western Davis Strait and further south into the Labrador Sea, and occasionally into waters off Newfoundland (Bigg et al., 1996; Tang et al., 2004) (Fig. 1). Within eastern Disko Bugt calved ice preferentially moves northwards, though a number of smaller icebergs are known to move westwards into the Davis Strait (Valeur et al., 1996). Today, the Canadian Archipelago (e.g. Baffin and Ellesmere Islands) does not represent a significant source of IRD to Baffin Bay (Tang et al., 2004).

IRD sourced from northeast Greenland and from sea–ice exported from the Siberian coast of Russia by the Transpolar Drift (Pfirman et al., 1997) are also transported southwards towards our study region by the EGC. Whilst such material represents potential distal sources of IRD deposited at our study sites, their contribution is likely to be low. Though sea–ice is sand-poor (Dethleff and Kuhlmann, 2009), recent XRD-based studies of the <2 mm fraction of terrigenous sediments from Baffin Bay (Andrews et al., 2014b), the western Nordic Sea (Andrews and Vogt, 2014) and on Denmark Strait (Andrews, 2011) indicate that IRD deposited in these regions during the modern and the Late Quaternary is sourced mainly from local glacial erosion, consistent with iceberg modelling studies (Bigg et al., 1996). These observations suggest that the spatial

Fig. 1. Geological map of Greenland with interpretation of sub-ice bedrock modified from Dawes (2009; their Fig. 1). Ice-free geology at coast is illustrated by darker coloured shading. KMB = Kettelidian Mobile Belt, AB = Archean Block, NMB = Nagssugtoqidian Mobile Belt, CFB = Caledonian Fold Belt. Also shown are terrestrial and marine study sites referred to in the main text (and detailed in Table 1), as well as major glacier/fjord systems (brown text) and ice sheet divides (black lines) (Weidick, 1995). Major drainage regions are labelled as NO = North, NE = North East, CE = Center East, SE = South East, SW = South West, CW = Center West, NW = North West, Br. = Bræ, Gl. = Glacier, and Se. = Sermia. Black arrows denote simplified modern day current systems. E/WGC = East/West Greenland Current. SSF = Scoresby Sund Fan. Hut. = Hutchinson Plateau Ice Cap. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)
pattern of IRD deposition in marine-proximal Greenland settings is dominated by local sources and that any far-travelled ice and its IRD are therefore diluted during sediment deposition by local glacial meltwater plumes and iceberg calving. This study therefore moves forward on the assumption that to identify the full range of iceberg calving sources on Greenland for any given time slice requires analysis of IRD deposited at a widely distributed network of marine sites.

### 3. Materials and methods

#### 3.1. Study sites

This study examines the provenance signature of individual feldspars and hornblendes from river and fjord sands collected from eleven widely distributed locations on the western, eastern and southern margins of south Greenland and that of sand IRD deposited in the modern or Early to Late Holocene at five Greenland-proximal marine locations (Table 1; Fig. 1). Limited Pb-isotope data exist for individual feldspars in bedrock from the Palaeoproterozoic Nagssugtoqidian orogen in Disko Bugt and the Rinkian fold belt (Connelly and Thrane, 2005), which are used here to gain a first order insight into the Pb isotope composition of feldspars ice-rafted from these regions. To better understand potential heterogeneity in the provenance character of iceberg calving sources we prefer, however, to study the Pb-isotope compositions and $^{40}$Ar/$^{39}$Ar ages of individual sand-sized grains in glacialfluvial and/or marine-proximal (fjord) sediments, as opposed to isolated bedrock specimens, since the former record an integrated signal of subglacial erosion within a particular drainage basin which will better represent that incorporated into calved icebergs. Glacialfluvial samples were targeted with modern iceberg sources in mind (Table 1). Samples Qa11-01 and Kn01-08 (Table 1) from the western NMB were targeted to characterise the provenance signature of modern-day sources of Greenland icebergs and IRD, respectively, in Disko Bugt (which housed the Last Glacial Fast flowing Jakobshavn ice stream; Hogan et al., 2012) and the region south of that (Fig. 1). Glacialfluvial sand ‘525252’ sampled proximally to the east of Kuummiit (near Tasiilaq) is used to define the provenance signature of Kaarle and Knud Rasmussen Glaciers on the eastern coast of the NMB. Core top sediment from Site JR106-GC06 is also used from this region for the provenance signature of IRD sourced from the Kangerlussuaq Fjord System (Dowdeswell et al., 2010). Sample Na04-08 from Nordre Sermilik Fjord near Narssarsuaq is used to characterise the provenance of KMB-sourced IRD from southernmost Greenland. Glacialfluvial sands ‘330272’ and ‘530551’ are used here to capture the provenance character of AB-sourced IRD calved from the Kangiata Nunaata (Nuuk) and Ser-

### Table 1

<table>
<thead>
<tr>
<th>Sample ID</th>
<th>Locality/region</th>
<th>Grid ref.</th>
<th>Water depth (m)</th>
<th>Greenland terrane</th>
<th>Glacier system(s)</th>
<th>Feldspar analyses</th>
<th>Hornblende analyses</th>
</tr>
</thead>
<tbody>
<tr>
<td>Na04-08</td>
<td>Nordre Sermilik Fjord, Narssarsuaq</td>
<td>61.1’N, 45.2’W</td>
<td>River Sand</td>
<td>KMB</td>
<td>Qajuuttap Sermia, Eqalorutisit Killit Sermiit</td>
<td>6 –</td>
<td></td>
</tr>
<tr>
<td>530551</td>
<td>Sermiligaaarsuk Fjord</td>
<td>61.5’N, 48.3’W</td>
<td>River Sand</td>
<td>Western AB</td>
<td>Sermiligaaarsuk Br., Sioralk Br.</td>
<td>20 25</td>
<td></td>
</tr>
<tr>
<td>330272</td>
<td>Nuup Kangerlua River, Nuuk</td>
<td>64.2’N, 51.5’W</td>
<td>River Sand</td>
<td>Western AB</td>
<td>Kangilimnguata Sermia, Narss Se., Qamanaarsaup Se., Akullersuap Se., Kangiata Nunaata Se.</td>
<td>24 25</td>
<td></td>
</tr>
<tr>
<td>550299</td>
<td>Gyldenløve Fjord</td>
<td>64.2’N, 41.2’W</td>
<td>River Sand</td>
<td>Eastern AB</td>
<td>unnamed Gls.</td>
<td>16 25</td>
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<tr>
<td>Kn01-08</td>
<td>Kangerlussuaq Fjord</td>
<td>67.1’N, 50.4’W</td>
<td>River Sand</td>
<td>Western NMB</td>
<td>Isuunnguata Se., Russell Gl.</td>
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<td></td>
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<td>Qa11-01</td>
<td>Disko Bug</td>
<td>68.3’N, 50.5’W</td>
<td>River Sand</td>
<td>Western NMB</td>
<td>Jakobshavn Isbrae, Alangorliup &amp; Saaqarliup Se.</td>
<td>34 10</td>
<td></td>
</tr>
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<td>525252</td>
<td>Kuummiit (Tasiilaq)</td>
<td>65.7’N, 36.7’W</td>
<td>River Sand</td>
<td>Eastern NMB</td>
<td>Knud Rasmussen Gl., Kaarle Gl.</td>
<td>12 –</td>
<td></td>
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<tr>
<td>Site JR106-GC06 1W 1–3 cm</td>
<td>Mouth of Kangerlussuaq Fjord System</td>
<td>68.1’N, 32.1’W</td>
<td>877</td>
<td>Eastern NMB</td>
<td>Kangerlussuaq, Coulard, Styrregletscher, Nordfjord and Frederiksborg Gls.</td>
<td>20 5</td>
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<tr>
<td>342549</td>
<td>Zachariae Istrøm</td>
<td>78.4’N, 20.4’W</td>
<td>River Sand</td>
<td>CFB</td>
<td>Zachariae Istrøm</td>
<td>30 24</td>
<td></td>
</tr>
<tr>
<td>520823</td>
<td>Near Hochstetterbugten, Østgrønland</td>
<td>74.7’N, 21.6’W</td>
<td>River Sand</td>
<td>CFB</td>
<td>Heinkel Gl.</td>
<td>12 –</td>
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<tr>
<td>Site JR51-GC28 1–2 cm</td>
<td>Immediately north of Scoresby Sund Fan</td>
<td>71.1’N, 18.3’W</td>
<td>1600</td>
<td>–</td>
<td>Offshore</td>
<td>21 –</td>
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<tr>
<td>Site 918A IH 1W</td>
<td>Irminger Basin</td>
<td>63.5’N, 38.4’W</td>
<td>1880</td>
<td>–</td>
<td>Offshore</td>
<td>35 8</td>
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<td>Site MC696, 0–1.5 cm</td>
<td>Labrador Sea</td>
<td>64.0’N, 57.6’W</td>
<td>n/a</td>
<td>–</td>
<td>Offshore</td>
<td>18 10</td>
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<tr>
<td>Site HU-90-013-013, 2–4 cm</td>
<td>Eirik Drift</td>
<td>58.1’N, 48.2’W</td>
<td>3771</td>
<td>–</td>
<td>Offshore</td>
<td>36 10</td>
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<tr>
<td>Site 907A 1W 31–33 cm</td>
<td>Iceland Plateau</td>
<td>69.1’N, 12.4’W</td>
<td>1811</td>
<td>–</td>
<td>Offshore</td>
<td>24 –</td>
<td></td>
</tr>
<tr>
<td>PO175GKC#7 1–2 cm</td>
<td>Denmark Strait (outer Kangerlussuaq Trough)</td>
<td>66.6’N, 30.8’W</td>
<td>300</td>
<td>–</td>
<td>Offshore</td>
<td>28 –</td>
<td></td>
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</table>


*After Rignot and Mouginot (2012), Marine sediment examined is either core top sediment (i.e. modern) or deposited on Eirik Drift during the last Holocene (Vernal and Hillaire-Marcel, 2000), in the Irminger Basin during the middle Holocene, ~6 ka (St John and Kruscek, 2002), on the Iceland Plateau during the early Holocene, ~10 ka (Jansen et al., 2000) and in the Kangerlussuaq Trough over the past 150 yr (Monso-Garcia et al., 2013).*
miligaarssuk and Sioralik tidewater glaciers on the southwest coast of Greenland. Northeast Greenland glacialfluvial sands ‘520823’ (Østgrønland) and ‘342549’ (Zachariae Isstrøm) and sediments from Site JR51-GC28 recovered by piston coring of the region just north of the Scoresby Sund Fan were also analysed to characterise IRD derived from East and Northeast Greenland outlet glaciers.

To understand spatial differences in the provenance of IRD deposited in marine settings proximal to Greenland, core-top or Holocene-aged sediments were examined from the Iceland Plateau (Ocean Drilling Program, ODP, Site 907), East Greenland Shelf (Site P0175-GKC#7), Irminger Sea (ODP Site 918), Eirik Drift (Site HU-90-013-013) and the Labrador Sea (Site MC-696) (large coloured circles, Fig. 1). The ages of samples from Sites 907 and 918 (~10 ka and ~6 ka, respectively; Table 1) were determined by reference to published age models for their stratigraphies (Jansen et al., 2000; St John and Krissel, 2002), IRD examined from the East Greenland shelf above Denmark Strait, Eirik Drift and the Labrador Sea comes from either core top (i.e. modern) sediments or was deposited in the latest Holocene (Table 1).

3.2. Individual feldspar and hornblende analyses

The 40Ar/39Ar dating of hornblendes and Pb-isotope analysis of feldspars has proven useful for tracking the sources of IRD deposited in the North Atlantic Ocean (e.g. Gwiazda et al., 1996; Hemming et al., 1998; Bailey et al., 2013; Knutz et al., 2013) and the Southern Ocean (e.g. Cook et al., 2014). Unlike accessory minerals used in tectonic provenance research (e.g. zircons for U–Pb/Hf dating or apatites and titanites for Sm–Nd isotypes), our target grains are common in both fluvial and clastic-rich marine sediments.

3.2.1. Pb isotopes in individual sand-sized feldspars

Pb-isotope analyses (n = 363; Table 1) of individual sand-sized (>150 µm) feldspar were performed at the University of Southampton on a Thermo-Scientific Neptune multicollector inductively coupled plasma mass spectrometer (MC-ICPMS) coupled with a NewWave/ESI UP193fx homogenised ArF excimer laser ablation system, operating at a wavelength of 193 nm, following methods reported in Bailey et al. (2012) (see Supplementary Materials). Feldspar grains were chosen randomly to best reflect the compositional variability within the sample, resulting in a natural preference towards K-feldspar (>80%) over plagioclase (~20%), although analyses of both phases are henceforth referred to as ‘feldspar’. Most ablations were performed with a laser spot size of 150 µm, with a small minority (~1%) analysed with a 100 µm spot. The Pb-isotope compositions of at least twenty grains were determined for each sample (Table 1). When initial analyses indicated the dominance of a single population less than 20 grains were analysed (e.g. Na04-08; Feldspar provenance is evaluated through their 206Pb/204Pb and 207Pb/204Pb ratios. No additional fidelity in provenance could be achieved by also using their 208Pb/204Pb, 207Pb/206Pb and 208Pb/206Pb ratios (all Pb-isotope data are available in our Supplementary Materials).

3.2.2. 40Ar/39Ar ages of individual sand-sized hornblendes

Individual sand-sized (>150 µm) hornblende grains (n = 142; Table 1) were washed by ultrasonic treatment using deionised water and packaged within aluminium foil in preparation for neutron irradiation at the McMaster reactor (Canada). The 40Ar/39Ar ages of irradiated hornblendes were determined at the Argon Isotope Laboratory at the Open University (UK) using noble gas mass spectrometry following Sherlock (2001). Irradiated samples were loaded into the laser-extraction system, and individual hornblende grains were fused with an infrared (λ = 1064 nm) Nd-YAG laser or ablated for five minutes with an ultraviolet (λ = 213 nm) laser-ablation microprobe. Neutron flux was measured using bismuth standard GA1550 (98.8 ± 0.5 Ma, Renne et al., 1998). Methods closely followed techniques for both ultraviolet (Wartho et al., 1999) and infrared (Adams and Kelley, 1988) analyses, including correction for measured blanks both before and after unknown sample analysis. Small sample sizes and low potassium led to high individual errors in some of the grains dated. All data are reported, but only data points with individual uncertainties of ≤~3% (2 s.d.) are interpreted.

4. Results and discussion

4.1. Characterising the provenance signature of Greenland’s ice-rafted debris

4.1.1. Pb isotopic composition of feldspars from Greenland fjord and glacialfluvial sands

The Pb isotope (206Pb/204Pb and 207Pb/204Pb) compositions of 192 individual sand-sized feldspars from our target glacialfluvial sands and fjord sediments from Greenland are shown in Fig. 2. A large range in Pb-isotope values is observed in these datasets, with a high degree of separation in Pb–Pb space between feldspars derived from each of Greenland’s tectonic terranes. Compared with previous compilations of the Pb-isotope composition of these tectonic terranes based on bedrock data (Bailey et al., 2012), our new data demonstrate the significant extra fidelity that can be achieved for Greenland sand IRD provenance by examining glacialfluvial sands (Fig. 3).

The Pb-isotope signature of the majority (95%) of feldspars derived from Scoresby Sund (JR51-GC28) and Disko Bugt (Qa11-01), the largest single modern-day iceberg calving sources on Greenland, are largely distinct in 206Pb/204Pb vs. 207Pb/204Pb cross plots (henceforth 206–207 space; compare Figs. 2A, D, E). Disko Bugt feldspars form a well-defined linear array in 206–207 space with 206Pb/204Pb ratios between ~13 and 17 associated in turn with progressively increasing 207Pb/204Pb ratios (Figs. 2D). In contrast, most feldspars (n = 16, 76%) from Scoresby Sund form a linear array with positive slope in 206–207 space with 206Pb/204Pb ratios between ~17 and 19, although a small subset (n = 5, 24%) also form a separate, but tighter, array with 206Pb/204Pb spanning ~14.5 to ~16.5 (Fig. 2A). The signature of feldspars from the Scoresby Sund region (JR51-GC28) is highly comparable in 206–207 space to that which have been determined for Østgrønland (520823) and for Zachariae Isstrøm (342549) from further north (Fig. 1), although the lastmentioned source exhibits a significantly larger spread (of ~17 to ~21) for its most radiogenic 206Pb/204Pb isotope values when compared to our Scoresby Sund dataset (Figs. 2A, E).

Feldspars from Disko Bugt (Qa11-01) overlap in 206–207 space with other NMB datasets. For instance, the most radiogenic Pb-isotope compositions (206Pb/204Pb < 15) of Disko Bugt feldspars overlap partially with the more narrowly defined linear array of Pb-isotope compositions associated with feldspars from Kangerluusuaq Fjord (Kn01-08, Figs. 2D, E) ~250 km south of Disko Bugt on the western NMB and from the Kangerlussuaq Fjord System (JR106-GC06, Figs. 2C, E) from the eastern NMB (with 206Pb/204Pb values of ~13 to 15). Feldspars sourced from Tasilaaq (525252) appear distinct in 206–207 space from our other NMB-located study regions, forming a narrowly defined linear array with relatively steep gradient between 206Pb/204Pb ratios of ~15.5 and 16 (dark orange diamonds in Fig. 2C). These data show partial overlap with the Pb isotope composition of feldspars from Nordre Sermilik Fjord (Na04-08) in the southern KMB terrane, which are characterised by 206Pb/204Pb ratios of ~15.5 that form tight groupings in 206–207 space with 207Pb/204Pb values of ~15.1 (Fig. 2B).
The Pb-isotope composition of feldspars from glacialfluvial sands from the western (Fig. 2B; samples 330272 and 530551) and eastern AB (Fig. 2B; sample 550299) are characterised by the least radiogenic Pb-isotope values of all individual feldspars analysed in this study and are largely distinct in 206–207 space from each other and from the provenance signature of feldspars found in NMB (Figs. 2C–D), KMB (Fig. 2B; Na04-08) and Caledonian terrane samples (Fig. 2A). On the western coast the Pb-isotope composition of feldspars from the Nuup Kangerlua River (330272) yield the lowest $^{206}\text{Pb}/^{204}\text{Pb}$ ratios analysed in this study ($\sim$11.5 and $\sim$12.5), displaying no overlap in 206–207 space with any other analysed sample (Fig. 2E). These values agree well with the Pb-isotope composition of feldspars from Itsaq gneisses south of Isua (Kamber et al., 2003), believed to be the least radiogenic values measured anywhere on Earth and as such are unique to this area. The Pb-isotope composition of feldspars from Sermiligaaq Fjord (530551) are distinct from those which characterise Nuup and are well-defined by a linear array with $^{206}\text{Pb}/^{204}\text{Pb}$ ratios between $\sim$12.5 and $\sim$14 in 206–207 space (Fig. 2B). Similarly, the Pb-isotope composition of feldspars from Gyldenløve Fjord (550299) on the eastern coast...
of the AB form a distinct narrow linear array with $^{206}\text{Pb}/^{204}\text{Pb}$ values between ~12 and 13 for progressively increased values of $^{207}\text{Pb}/^{204}\text{Pb}$ (Fig. 2B).

### 4.1.2. $^{40}\text{Ar}–^{39}\text{Ar}$ ages of hornblendes in Greenland glacifluvial sands and fjords

The $^{40}\text{Ar}–^{39}\text{Ar}$ age distribution of 114 individual sand-sized hornblendes from six of our target Greenland glacifluvial sands and fjord sediments (Table 1) are shown in Fig. 4. Initial assessment of the age of individual hornblendes from these localities reveals populations characterised by a wide range of ages spanning the Palaeoarchaean to the middle Palaeozoic (~3800 to 380 Ma) consistent with the known age distribution of Greenland’s tectonic terranes (Henriksen et al., 2009). In concert with the Pb-isotope composition of feldspars from our potential IRD source regions, distinct differences exist, however, in the age distribution of hornblende grains from the individual terranes examined.

Minor numbers ($n = 5$, 21%) of hornblendes sourced from Zachariae Isstrom (342549), the only locality in this study from the CFB to be analysed by $^{40}\text{Ar}–^{39}\text{Ar}$ dating, appear to be characterised by both Neoarchaean to Palaeoproterozoic (2600–1900 Ma) and earliest Neoproterozoic (1000 Ma) ages (Fig. 4A). The majority ($n = 19$, 79%), however, are part of a significantly younger mode bearing Lower Palaeozoic-ages centered on ~450 Ma, consistent with resetting by Caledonian metamorphism (Fig. 4A).

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### Table 1: Greenland IRD Sources

<table>
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<tr>
<th>Neoproterozoic</th>
<th>Palaeozoic</th>
<th>Archaean</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Zachariae Isstrom</strong> (342549)</td>
<td>(KF5) n = 24</td>
<td></td>
</tr>
<tr>
<td><strong>Kanger Fjord System</strong> (JF106-S006) (East NMB)</td>
<td>n = 7</td>
<td></td>
</tr>
<tr>
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<td></td>
</tr>
<tr>
<td><strong>Sermilik Fjord</strong> (330557) (West AB)</td>
<td>n = 25</td>
<td></td>
</tr>
<tr>
<td><strong>Nuup</strong> (330272) (West AB)</td>
<td>n = 25</td>
<td></td>
</tr>
<tr>
<td><strong>Disko Bugt</strong> (Qa11-01) (West NMB)</td>
<td>n = 10</td>
<td></td>
</tr>
</tbody>
</table>

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### Marine Sites

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<th>Palaeozoic</th>
<th>Archaean</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Immingham Sea</strong> (Site 918)</td>
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<td></td>
</tr>
<tr>
<td><strong>Link Drift</strong> (Site HU90-013)</td>
<td>n = 10</td>
<td></td>
</tr>
<tr>
<td><strong>Labrador Sea</strong> (Site MC096)</td>
<td>n = 9</td>
<td></td>
</tr>
</tbody>
</table>

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**Fig. 3.** Comparison of Pb-isotope values for potential Greenland ice-rafted debris sources compiled by Bailey et al. (2012) (red transparent ‘ellipses’ and red labels) and this study (hollow multi-coloured ‘bubbles’ and black text labels) in $^{206}\text{Pb}/^{204}\text{Pb}$-$^{207}\text{Pb}/^{204}\text{Pb}$ space. The source fields reported in this study are based on Pb-isotope compositions of individual feldspars from Greenland glacio-fluvial and fjord sands (Fig. 2). The source fields shown by Bailey et al. (2012) are based on feldspars (‘fs’ and conformable ore galenas (‘g’) from circum-North Atlantic Ocean bedrock. KMB = Ketilidian Mobile Belt, E/W AB = East/West Archaean Block, W/E NMB = East/West Nagssugtoqidian Mobile Belt, CFB = Caledonian Fold Belt. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

**Fig. 4.** Comparison of $^{40}\text{Ar}–^{39}\text{Ar}$ ages of individual sand-sized (>150 μm) hornblendes from Greenland glacifluvial and fjord sands (A–F) and Holocene Greenland-proximal marine sediments (G–I). Samples are ordered in terms of geographic position, starting from the north-easternmost sample (Zachariae Isstrom, A) and moving clockwise around the GIS to the western-most samples (Disko Bugt, F). Geological timescale provided for reference. See map in Fig. 1 (and Table 1) for sample locations. CFB = Caledonian Fold Belt, NMB = Nagssugtoqidian Mobile Belt, AB = Archaean Block.
from Disko Bugt (Qa11-01) in the western KMB (n = 10) appear to be dominated by ages centered on ~1800–1900 Ma (Fig. 4F). Although the hornblende population analysed is too small to be statistically significant, the five grains dated from sediments deposited in the Kangerlussuaq Fjord System (Fig. 4B, JR106-CC06) highlight that hornblendes from this important modern-day iceberg calving source may have a similar age distribution to those transported by icebergs from Disko Bugt.

Targeted AB localities are characterised by sands populated largely by Archaean hornblendes (Figs. 4C–E). Hornblendes from Gyldenløve Fjord (Fig. 4C, 550299) and Nuup Kangerlua River (Fig. 4E, 330272) on the eastern and western AB coastlines are dominated by a single skewed mode centered on ~2600–2700 Ma. This mode is also present in the hornblende population examined from Sermiligaaarsuk Fjord (530551) on the AB, although it is less prominent at this western locality because the ages of these grains exhibit a bimodal distribution that is also characterised by the presence of a second younger mode centered on ~2100 Ma (Fig. 4D).

4.2. Provenance of IRD deposited in Greenland-proximal sediments during the Holocene

To examine how we can use our source data to reconstruct the provenance of Greenland-proximal IRD deposition, the provenance of IRD from five Greenland-proximal marine sites deposited during the early Holocene to the modern has been studied. The Pb-isotope compositions of 144 individual sand-sized feldspars deposited from these sites are shown in Fig. 5. To aid provenance determination, histograms of $^{206}\text{Pb}/^{204}\text{Pb}$ values for both source and marine core feldspars are shown in Fig. 6. The $^{40}\text{Ar}/^{39}\text{Ar}$ ages of 28 individual sand-sized hornblendes deposited on the Iceland Plateau (at ODP Site 918), on Eirik Drift (Site HU-90-013-013) and the Labrador Sea (Site MC-696) are also shown in Figs. 4G–I. Our analysis of the provenance of IRD deposited at these sites is primarily based on the observation that inter-terrace differences in the Pb isotope composition of ice-rafted feldspars (i.e. between those sourced from the NMB, CFB and AB) is larger than intra-terrace differences (between those sourced from individual localities studied here from, e.g. Kangerlussuaq Fjord (System) and Disko Bugt in the NMB). Based on the present-day westward ocean current system around southern Greenland we also assume that eastern IRD sources can imprint provenance signatures on sediment deposited off western Greenland whilst the opposite cannot be true. While such currents may also introduce sea-ice-derived sand sourced from the Arctic to our samples, if such detritus dominated IRD deposition at our study sites we would expect to find a lack of spatial heterogeneity in our provenance data (the opposite of what we find; see Fig. 2E and Fig. 4).

4.2.1. Site 907, Iceland Plateau

The majority of feldspars (n = 15; 75%) deposited at Site 907 on the Iceland Plateau during the Early Holocene form a linear array in $206\text{Pb}/204\text{Pb}$ space with $^{206}\text{Pb}/^{204}\text{Pb}$ values ~17–19, although a subset (n = 5; 25%) also cluster between values of ~15 and 16 (Fig. 5A). In terms of provenance, this bi-modal distribution fits most closely in 206–207 space with the Pb-isotope signature of CFB-derived IRD (Fig. 5A). Our Site 907 sample contains small numbers of sand-sized basalt clasts that could have been recirculated northwards from Iceland, from the Geikie Plateau calved from the southern coastline Scoresby Sund and/or from further north at Hold With Hope and Shannon Ø. Given both the strength of the EGC today, the modelled southerly drift directions east of Greenland for icebergs sourced north of Scoresby Sund (Bigg et al., 1996) and potential for small amounts of this ice to drift further south-east promoting recirculation to the site by the northerly Irminger
Fig. 6. Comparison of histograms of the $^{206}$Pb/$^{204}$Pb ratios of individual sand-sized (>150 μm) feldspars from Greenland glaciﬂuvial and fjord sands (A–L) and Holocene Greenland-proximal marine sediments (M–Q). Histograms displayed based on geographic position of samples studied: starting from the north-eastern most locality (Zachariae Isstrøm, A) moving clockwise around Greenland to the western-most localities (Disko Bugt, L). See map in Fig. 1 (and Table 1) for sample locations. CFB = Caledonian Fold Belt, E/W NMB = East/West Nagssugtoqidian Mobile Belt, E/W AB = East/West Archaean Block. (For interpretation of the references to colour in this ﬁgure, the reader is referred to the web version of this article.)

Current (i.e. Andrews et al., 2014a), we speculate that IRD deposited at Site 907 during the Early Holocene is more likely to be dominated by Caledonian sources located along the coastline of northeast Greenland.

4.2.2. Site GKC#7, Denmark Strait
Site GKC#7 lies on the Kangerlussuaq Trough margin, ~4.5° south of Scoresby Sund (Fig. 1). Radiogenic $F_{Nd}$ and Sr isotope studies (Simon, 2007) of terrigenous sediments from this area show that bulk IRD deposited here bears an early Tertiary basalt signature. A subordinate number of the individual sand-sized feldspars deposited at this site in the modern (over past 150 years) are characterised by $^{206}$Pb/$^{204}$Pb ratios > ~16 and therefore may be sourced from the CFB (Fig. 5B). The Pb-isotope compositions of the majority of feldspar from this site ($n = 24$; 86%), however, form tight clusters in 206–207 space with $^{206}$Pb/$^{204}$Pb ratios of ~12.5 to 13.5 (Figs. 5B and 6N). In terms of provenance, these grains mainly overlap in 206–207 space with a variety of western...
and eastern NMB river sands and fjord sediments (Fig. 5B). Given the location of Site GK#7 (Fig. 1) and the strength of the EGC, the nearby Kangerlussuaq Ford System (JR106-GC06) is, however, the most likely source for these grains (Figs. 6D, N). This finding is in keeping with both the significant number of sand-sized basalt clasts (~5–10% of assemblage) observed in our GK#7 sample, and an XRD-based study of the bulk provenance of the <2 mm terrigenous fraction at nearby Site MD99 2322 (Fig. 1), which shows that 90% of modern-day sediments deposited at this site are sourced locally (Andrews et al., 2014a).

4.2.3. Site 918, western Irminger Sea

Although Site 918 is situated in the western Irminger Basin immediately proximal to Greenland’s AB (Fig. 1), the Pb-isotope composition of only 2 of the middle Holocene feldspars (out of 35) deposited at this site (red data in Fig. 5C) overlap with analyses of glaciﬂuvial feldspars from this tectonic terrane (e.g. from Sermiligaarsuk Fjord, 530551). Instead, their Pb-isotope compositions overlap mainly with those of feldspars from the Palaeoproterozoic NMB (Fig. 5C). Ar/39Ar data from Site 918 are too few in number (n = 4) to allow us to identify likely speciﬁc sources of IRD deposition at this site (Fig. 4G). Yet their Palaeoproterozoic ages (~1870–2250 Ma) tentatively support the notion that Site 918 IRD is dominantly sourced from the NMB (Fig. 4B) and not from local eastern AB sources (Fig. 4C). The provenance inferred for Site 918 IRD based on these data is consistent with eastern NMB and CFB sources inferred for drop stones from nearby grab station D97-18 (Linthout et al., 2000) and with the relatively high abundance (~5–10%) of sand-sized basalt found in our sample. Given the presence of small numbers of grains in our Site 918 feldspar population that are unambiguously derived from Caledonian sources (n = 4), it is possible that some of the ‘NMB’ feldspars might instead represent ‘far-travelled’ IRD sourced from the CFB.

4.2.4. Site HU90-013-013, Eirik Drift

Many of the feldspars deposited on Eirik Drift at Site HU90-013-013 during the latest Holocene exhibit a similar distribution in 206–207 space to those deposited up-current at Site 918 (compare black and red data in Figs. 5C and 6G, and 206Pb/204Pb histograms in Figs. 6O, P). This ﬁnding implies that many of the sources responsible for IRD deposition in the western Irminger Basin during the middle Holocene are also responsible for IRD deposition on Eirik Drift during the latest Holocene. The Pb-isotope composition of two feldspars deposited at Site HU90-013-013 highlight a potential, albeit minor, contribution from sediments shed from the eastern AB to IRD deposition at this site during the modern (two black data in Fig. 4C that overlap with Gyldenløve Fjord Pb-isotope values). In support of this concept, Ar/39Ar data from this site highlight that at least some hornblends deposited on Drift are Archaean in age (Fig. 4H).

The absence of any sand-sized basalt clasts in our Site HU90-013-013 sample argues against the eastern NMB being the dominant source for feldspars deposited at this site during the latest Holocene. A number of feldspars deposited at Site HU90-013-013 with 206Pb/204Pb ratios between ~15 and 17 (n = 13) do not overlap with Pb-isotope data from Site 918 in 206–207 space (Fig. 5C). The absence of feldspars with such compositions in our Site 918 dataset indicates that these grains may not be sourced from east Greenland. Both iceberg trajectory modelling (Bigg et al., 1996) and analysis of Landsat imagery (Howat and Eddy, 2011) suggest that these feldspars may represent grains derived from important modern-day iceberg calving sources located on the KMB yet to be documented in our source datasets.

4.2.5. Site MC-696, Labrador Sea

The location of Site MC-696 in the Labrador Sea permits iceberg calving locations from all of Greenland’s tectonic terranes to be potential sources of IRD deposition at this site (Fig. 1). It is notable, therefore, that feldspars deposited at this site during the modern are largely characterised by Pb-isotope compositions that are only likely to be derived from either the NMB or KMB (blue data in Figs. 5D and 6Q). A number of these grains (n = 12) also overlap in 206–207 space with the Pb-isotope composition of feldspars from the CFB (Fig. 5D). Indeed, one data point with a high 206Pb/204Pb ratio (of ~18) implies that some icebergs calved from this region apparently survive the journey to the Labrador Sea (Figs. 5D and 6Q). Potential north-western sources (i.e. the Petermann glacier) are known to generate icebergs capable of reaching the Labrador sea (Halliday et al., 2012). Whilst ice drift models suggest both sources (north of Denmark Strait and NW Greenland) are marginally capable of supplying Site MC-696 (Bigg et al., 1996), the absence of large numbers of grains with a CFB signature makes it highly unlikely that the majority of feldspars deposited at Site MC-696 with unambiguous 206Pb/204Pb ratios (~15–16) are sourced from these far-field regions.

Owing to overlap between Proterozoic sources of Greenland IRD in 206–207 space it is not currently possible to determine the origin of most feldspars deposited at Site MC-696 at the intra-tesseral scale. Their Pb-isotope compositions arguably show good agreement in 206–207 space with NMB iceberg calving sources in Kangerlussuaq Fjord (Kn01-08, Disko Bugt (Qa11-01) and the region to its north (Connelly and Thrane, 2005) (Fig. 5D). These data are also comparable in 206–207 space with the composition of feldspars deposited on Eirik Drift (HU90-013-013) with 206Pb/204Pb ratios ~15–17 that we attribute to KMB iceberg calving (compare Figs. 5C to 6D, H).

4.3. Inferences on Holocene GIS retreat based on Greenland sand IRD provenance data

A reconstruction of the Holocene evolution of GIS iceberg calving sources based on our new datasets is beyond the scope of this study. Yet, the provenance determinations that can be made for both our new and previously published (e.g. Knutz et al., 2013) Greenland–proximal marine IRD datasets based on our new characterisations for Greenland-sourced IRD demonstrate the potential that this approach has to provide valuable context on previous inferences of the retreat history of the GIS during the Early Holocene (e.g. Bennike and Björck, 2002; Dyke et al., 2014; Winsor et al., 2015).

For instance, the Pb isotope composition of sand-sized feldspars that we report for Site 907 indicate that northeast Greenland was still supplying IRD to the Iceland Plateau at ~10 ka, which is consistent with the 14C chronology for the timing (~10–9 ka before present) for deglaciation of the coastal margin of northeast Greenland during the Early Holocene (Bennike and Björck, 2002). Based on 10Be surface exposure ages and 14C chronologies from the coastline of southwest Greenland it has been inferred that on the western AB, the GIS retreated rapidly from the coastline towards its near-modern ice margin between ~11 and 10 ka (Bennike and Björck, 2002; Winsor et al., 2015). Indeed, our Pb isotope and Ar-Ar data from Site MC-696 (64°N, 57.6°W) show that the western AB does not represent a modern source of IRD to the adjacent Labrador Sea (compare Figs. 7A and C). Recent evidence for significant deposition of Archaean ice-raﬁed hornblends at Site DA04-31 (62.3°N, 54.2°W) during the Early Holocene (Knutz et al., 2013) suggest, however, that the export glaciers on the western AB remained a signiﬁcant source of IRD to the Labrador Sea until at least ~9 ka, which our Greenland source characterisation indicates was most likely derived from the Nuuk region (the Godthaabfjord and
of southern Greenland. The generation of such datasets from sand-sized hornblends and feldspars therefore has great potential to help better understand spatial differences in the provenance of IRD deposited in the Greenland-proximal marine setting and the timing of southern GIS margin deglaciation during the Holocene and past interglacials.

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Appendix A. Supplementary material

Supplementary material related to this article can be found online at http://dx.doi.org/10.1016/j.epsl.2015.10.054.

References
