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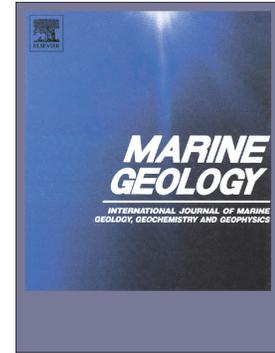
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Tracing marine cryptotephra in the North Atlantic during the Last Glacial Period:

Protocols for identification, characterisation and evaluating depositional controls

Peter M. Abbott^{1,2,3,*}, Adam J. Griggs¹, Anna J. Bourne^{1,4}, Siwan M. Davies¹

¹Department of Geography, College of Science, Swansea University, Singleton Park,
Swansea, SA2 8PP, UK

²School of Earth and Ocean Sciences, Cardiff University, Park Place, CF10 3AT, Cardiff, UK

³Institute of Geological Sciences and Oeschger Center for Climate Change Research,
University of Bern, Baltzerstrasse 1+3, Bern 3012, Switzerland

⁴Geography and Environment, University of Southampton, University Road, Southampton,
SO17 1BJ, UK

*Corresponding author (abbottp@cardiff.ac.uk)

Abstract

Tephrochronology is increasingly being utilised as a key tool for improving chronological models and correlating disparate palaeoclimatic sequences. For many sedimentary environments, however, there is an increased recognition that a range of processes may impart a delay in deposition and/or rework tephra. These processes can affect the integrity of tephra deposits as time-synchronous markers, therefore, it is crucial to assess their isochronous nature, especially when cryptotephra are investigated in a dynamic marine environment. A methodology for the identification and characterisation of marine cryptotephra alongside a protocol for assessing their integrity is outlined. This methodology

was applied to a wide network of North Atlantic marine sequences covering the last glacial period. A diverse range of cryptotephra deposits were identified and, based on similarities in physical characteristics (e.g. glass shard concentration profiles and geochemical homogeneity/heterogeneity), indicative of common modes of tephra delivery and post-depositional reworking, a deposit type classification scheme was defined. The presence and dominance of different deposit types within each core allowed an assessment of spatial and temporal controls on tephra deposition and preservation. Overall, isochronous horizons can be identified across a large portion of the North Atlantic due to preferential atmospheric dispersal patterns. However, the variable influence of ice-rafting processes and an interplay between the high eruptive frequency of Iceland and relatively lower sedimentation rates can also create complex tephrostratigraphies in this sector. Sites within a wide sector to the south and east of Iceland have the greatest potential to be repositories for isochronous horizons that can facilitate the synchronisation of palaeoclimatic records.

Keywords: Quaternary; palaeoceanography; tephrochronology; North Atlantic; transport and deposition; marine cores; glass shard concentrations

1. Introduction

Deposits of volcanic ash, tephra, can act as time-synchronous marker horizons linking palaeoclimatic sequences to help improve chronological models and assess the relative timing of climatic changes (Lowe, 2011). Two fundamental principles that underpin the application of tephrochronology are the rapid deposition of ash at all sites, i.e. instantaneous in geological terms, and that the stratigraphic position of the ash in a sequence directly relates to the timing of the volcanic eruption. Processes that either delay the transportation of ash

particles to a site or rework the material following initial deposition can have major impacts on the integrity of deposits as well-resolved isochronous markers. The operation of such processes has been investigated in many sedimentary environments (e.g. Ruddiman and Glover, 1972; Austin et al., 2004; Davies et al., 2007; Brendryen et al., 2010; Payne and Gehrels, 2010; Pouget et al., 2014; Todd et al., 2014; Hopkins et al., 2015; Watson et al., 2015; Zawalna-Geer et al., 2016) and are particularly crucial for cryptotephra, due to the absence of any visible stratigraphic features that would identify the position of the isochron, and hence the timing of deposition, and draw attention to any post-depositional reworking (Davies, 2015). For the marine environment it is critical to consider these processes due to its dynamic nature and the wide range of potential influences, especially when investigating sediments from glacial periods and high-latitude settings where ice-rafting processes could be a significant complicating factor.

Isochronous tephra deposits are formed in the marine environment if primary tephra fallout is deposited on the ocean surface, rapidly transported through the water column, deposited on the seabed and then preserved in the sediment by subsequent marine sedimentation (Figure 1). However, deposition onto other surfaces, e.g. ice sheets and sea-ice, subsequent rafting, and post-depositional reworking and redistribution processes, such as bioturbation and sedimentary loading, can have a major impact on the integrity of tephra deposits in this environment (Figure 1). For instance, these processes can affect the stratigraphic position of a tephra, a pertinent issue for marine sequences due to their lower resolution relative to other records, and potentially compromise the use of the deposit as an isochron. Therefore, it is essential that a full assessment of the sedimentation and depositional processes influencing the preservation, form and isochronous nature of marine cryptotephra deposits is undertaken.

This is especially important if tephra or cryptotephra horizons are to be used as tie lines to assess the relative timing of climatic changes between depositional environments.

Here we present an optimised protocol for marine cryptotephra studies that builds on previous studies, such as, Austin et al. (2004), Brendryen et al. (2010), Abbott et al. (2011, 2013, 2014, 2016), Davies et al. (2014) and Griggs et al. (2014), which used similar methods and indicators to assess visible or cryptotephra deposits within single core sequences. Our examples are derived from a range of depositional settings in the North Atlantic region (Figure 2), but the methodological approach could be applicable to many other marine settings. Within our approach, cryptotephra are identified and characterised using density separation, magnetic separation and electron probe micro-analysis (EPMA) techniques. We then employ a series of indicators to assess the isochronous nature of tephra deposits in the North Atlantic. These include (i) high-resolution shard concentration profiles, (ii) glass shard size variations, (iii) comprehensive single-shard geochemical analysis, and (iv), when available, co-variance with ice-rafted debris (IRD). With a focus on the time-period between 60-25 cal ka BP in the North Atlantic we define several key types of cryptotephra deposit. These are manifested as variations in glass shard concentrations, that share characteristics, such as shard concentrations profiles and geochemical compositions, which are interpreted as being indicative of common transport, depositional and post-depositional processes. The cryptotephra deposit types provide a basis for assessing the dominant controls on tephra deposition in different areas and time periods. Given the widespread core network employed in this study we pinpoint sectors of the North Atlantic Ocean that preferentially preserve isochronous deposits and these underpin a marine tephra framework presented in Abbott et al. (in revision). These horizons are the most valuable for establishing independent high-

precision correlations to the Greenland ice-core records to assess the relative timing of abrupt climate changes.

2. Methodology

2.1 Core Network

Thirteen marine sequences are included in our core network and each record was investigated using the same methodological approach (Figures 2 and 3; Table 1). Cores with well-developed proxy records were prioritised due to the overarching goal of assessing the relative timing of abrupt climate changes during the last glacial period. In addition, cores from areas with high sedimentation rates and sufficient material for contiguous tephra sampling were selected. Overall the network has a wide geographical spread. However, in some instances paired cores from nearby locations were investigated to assess the stratigraphic integrity of individual tephra deposits. It was not always possible to fulfil all of these requirements. For instance, contiguous samples were not available from MD95-2024 and two sites, M23485-1 and GIK23415-9, do not have well-resolved records of abrupt climate changes. However, these sites were included to increase the geographical extent and capture a wide range of depositional settings.

2.2 Identification of cryptotephra deposits

Cryptotephrae were identified and characterised according to the methodological protocol outlined in Figure 3. Although most aspects of this marine-focussed methodological approach have been described separately in previous studies, here we synthesise the full procedure.

Core sequences were initially analysed at a low-resolution (5 or 10 cm sampling intervals) using contiguous samples, i.e. samples taken along the whole length of depth intervals with no gaps between samples, to provide an initial quantified assessment of tephra content (i.e. glass shards) for the whole period of interest. Selected intervals were then reanalysed at a high-resolution (1 cm depth intervals) depending on a range of factors, outlined in Section 3, consistent with other studies of both marine and terrestrial sequences (e.g. Pilcher and Hall, 1992; Lane et al., 2015; Matthews et al., 2015). Both low and high-resolution samples were processed according to the workflow outlined in Figure 3.

Within the protocol, samples are sieved to isolate glass shards in three recommended size fractions ($>125\ \mu\text{m}$, $80\text{-}125\ \mu\text{m}$ and $25\text{-}80\ \mu\text{m}$). This separation and focus on fine-grain sizes is a development from prior studies that focused on coarser grain size fractions (e.g. $>150\ \mu\text{m}$ - Austin et al., 2004, Voelker and Haflidason, 2015; $63\text{-}125\ \mu\text{m}$ and $125\text{-}150\ \mu\text{m}$ – Brendryen et al., 2010), most typically utilised in the identification of foraminifera. The development was driven by the increased identification of cryptotephra as fine-grained deposits in distal sequences (Davies, 2015). The smallest grain-size fraction ($25\text{-}80\ \mu\text{m}$) was split using heavy liquid separation into density fractions most likely to contain glass shards, a method initially developed to identify tephra or cryptotephra in terrestrial sediments (Hodder and Wilson, 1976; Turney, 1998; Blockley et al., 2005). Magnetic separation is an additional step utilised to separate paramagnetic basaltic material from minerogenic material with a similar high density ($>2.5\text{g/cm}^3$; Griggs et al., 2014). Whilst magnetic separation is infrequently employed for terrestrial sequences, e.g. Mackie et al. (2002), it is routinely applied in this investigation to aid the isolation and identification of basaltic glass. The high number and proportion of basaltic horizons, relative to rhyolitic horizons, identified in this study demonstrates the value of including this step of magnetic separation within marine cryptotephra studies in the North

Atlantic. During low-resolution analysis, magnetic separation was only utilised on the 25-80 μm size fraction, because the time required for this process was longer than the time required to count shards from an unseparated sample of the coarser-grained fractions. However, during preparation of samples for geochemical analysis these coarse-grained fractions were magnetically separated alongside the 25-80 μm fraction to provide a purer basaltic glass sample.

If a low-resolution tephrostratigraphy was being constructed, all fractions were inspected for glass shard content using optical microscopy (i.e. $>125 \mu\text{m}$, $80-125 \mu\text{m}$, $2.3-2.5 \text{ g/cm}^3$ and the $>2.5 \text{ g/cm}^3$ magnetic fraction; step 12 in Figure 3). However, when glass shard concentration profiles were refined at a higher 1 cm sampling resolution, some fractions were not inspected. For example, if no rhyolitic material was present at a low resolution then the $2.3-2.5 \text{ g/cm}^3$ fraction was not inspected.

Depending on the nature of the samples and the glass contained within a sequence, alternative or additional steps were occasionally adopted (Figure 3). For instance, in some cores sediment clusters, that appear to consist of sediment bound together by biogenic silica, were observed (see also Ponomareva et al., 2018). These clusters were broken down using a weak treatment of sodium hydroxide (NaOH) (step 5). This chemical treatment could also be undertaken after step 3 of the method if clusters are known to be present following initial investigations. In such cases, the HCl should be washed out of the sediments, but no re-sieving is necessary. NaOH has previously been used in cryptotephra studies to remove biogenic silicates (e.g. Rose et al., 1996), with samples warmed to 90°C for 4 hours. However, it was found that treatment at room temperature for 1 hr was sufficient to disaggregate the sediment clusters in this study. As a precaution, NaOH treatment was

avoided when samples were being prepared for geochemical analysis, as it has been suggested that NaOH could cause geochemical modification (e.g. Blockley et al., 2005). However, other studies have shown that such treatments do not affect the glass composition (e.g. Steinhäuser and Bichler, 2008) and experimentation by Ponomareva et al. (2018) indicates that electron-probe micro analysis (EPMA) analyses are unaffected by this weak NaOH treatment.

To quantify exceptionally high shard concentrations ($\sim >10,000$ per 0.5 g dry weight sediment (dws)), samples were spiked with Lycopodium spore tablets containing a known quantity of pollen grains (step 10 in Figure 3). The threshold of 10,000 glass shards per 0.5 g dws is recommended as this roughly equivalent to the number of pollen spores present in a tablet. The ratio between glass shards and pollen grains is then used to quantify shard concentrations (e.g. Griggs et al., 2014). This is an adaption of a standard pollen counting approach previously applied to tephra and cryptotephra studies by Gehrels et al. (2006). Typically, it is not known if this quantification approach is required until low-resolution analysis has been conducted. Hence, if high shard concentrations were observed in low-resolution samples and it became apparent that shard concentrations would exceed 10,000 shards, then counting was halted and the additional step of spiking samples was incorporated into high resolution analysis of those sections.

2.3 Geochemical analysis of cryptotephra deposits

Shard concentration profiles are employed to select samples for geochemical analysis using the criteria outlined in Section 3. Samples were re-processed using steps 1-9 of the procedure in Figure 3, but, the fractions of interest were then mounted in epoxy resin on 28×48 mm

microprobe slides to permit the sectioning of the glass shards (Figure 3). When high shard concentrations were present, all material from the fraction was mounted directly on to the slides. When glass shards were present only at a low concentration ($\sim < 50$ per 0.5 g dws) they were picked onto a microprobe slide using a micromanipulator. Shards prepared by this method are easier to locate during sectioning and EPMA analysis. Flat and polished sections through the individual glass shards were produced for EPMA analysis using decreasing grades of silicon carbide paper and 9, 6 and 1 μm diamond suspensions.

EPMA was conducted at the Tephra Analytical Unit, University of Edinburgh, using a Cameca SX100 with five wavelength dispersive spectrometers over a number of analytical periods. All shards were analysed using the same operating conditions outlined in Hayward (2012). Pure metals, synthetic oxides and silicate standards were used for calibration. The secondary standards of Cannelto Lami Lava, Lipari and BCR2g were analysed at regular intervals to monitor for instrumental drift within analytical sessions, to assess the precision and accuracy of analysed samples and to provide a cross-check of the comparability of analyses between analytical periods. A comparatively large number of shards (~ 20 -40 individual shards) were analysed for each deposit to provide comprehensive characterisations that underpin the assessment of taphonomic processes, depositional controls and the isochronous nature of deposits. For all analysis and data comparison the major element data were normalised to an anhydrous basis, i.e. 100 % total oxides. However, the geochemical data utilised here are provided as raw analyses in the Supplementary Data alongside secondary standard analyses.

3. Constructing a tephrostratigraphy

The two major indicators that we employ to assess the integrity of marine tephra deposits are (i) contiguous high-resolution shard concentration profiles and (ii) rigorous geochemical characterisation of the glass shards. These are the key aspects of the tephrostratigraphies defined in this work. Constructing a tephrostratigraphy, however, involves a series of selections and we illustrate our approach, which aimed for consistency and comparability between cores, with reference to the record of brown (basaltic) shards in the MD99-2251 core from the Iceland Basin between 1650-1950 cm depth (Figure 4). There was a distinct lack of colourless shards in this core section but a slight increase was observed towards the base, which can be related to reworking and redistribution of the underlying North Atlantic Ash Zone II (NAAZ) II (see Section 4).

First a low-resolution shard concentration profile is constructed to determine the overall presence of cryptotephra and to define the background level of glass shards within a sequence (e.g. Figure 4a). All notable shard peaks were then re-analysed at a high-resolution (1 cm sampling interval) to refine their stratigraphic position. This step is crucial because the peak in concentration is typically thought to represent the timing of atmospheric fallout from a volcanic event (e.g. Ruddiman and Glover, 1972; Jennings et al., 2002; Davies et al., 2012). Theoretically it is possible for the maximum shard concentration peak to lie below the original depth of deposition, based on an interplay of the extent of mixing within and depth of the mixing layer and the sedimentation rate at the site, but, the impact of such mixing has been assessed as negligible in practice (Berger and Heath, 1968; Ruddiman and Glover, 1972). Indeed, our focus on high sedimentation rate sites would negate this effect, however, it is recommended that the potential influence of mixing on the isochron position is considered for individual horizons if they are to be used as isochronous tie-lines between sequences.

Selecting which peaks to refine at a 1 cm sampling resolution depends on the peak versus background concentrations, the shape and discreteness of peaks and replication across grain-size fractions (e.g. Figure 4a). To some extent there is subjectivity in the selection of peaks and no consistent concentration thresholds could be defined because of variability in peak and background shard concentrations both within and between the core sequences. In most instances, but not exclusively, shard concentrations in the 25-80 μm fraction displayed the greatest variability and occurrence within the records and were the prime criteria for these selections (e.g. Figure 4a). For some cores, high-resolution investigations were extended over intervals wider than the main peaks to provide a greater constraint on shard concentration variations (e.g. between 1678-1698 cm in MD99-2251; Figure 4a) and/or additional samples were analysed to determine if smaller peaks were due to increased input of material from a volcanic event or general fluctuations in background shard concentrations (e.g. between 1869-1874 cm and 1879-1884 cm in MD99-2251; Figure 4a). In addition, the time required for processing and analysing the number of selected samples was considered.

Reanalysing selected sections at a high-resolution allows an integrated shard concentration profile to be constructed (e.g. Figure 4b) that, in general, constrains the shard peaks to vertical distances of 1 or 2 cm in a core, and higher concentrations were normally observed in the high-resolution counts (e.g. peaks at 1680-1681 cm and 1904-1905 cm depth in MD99-2251; Figure 4b). This observation was anticipated because the low-resolution counts should provide an average of the glass shard concentration over the sampling interval and has been observed for other cores within the network. However, in some cases lower peak concentrations or very few shards were observed in the high-resolution samples (e.g. the 1869-1874 and 1879-1884 cm sections in MD99-2251; Figure 4b). This mis-match may be due to uneven lateral distribution of glass shards within core sequences, a lack of horizontal

continuity or glass shards being constrained in pods or lenses. Glass shard distributions of this nature have been observed in thin section (2D) and X-ray microtomography (3D) analysis of North Atlantic marine tephra sediments (Griggs et al., 2014, 2015). These additional 2D and 3D methods can provide further sedimentological information to aid isochron placement and the interpretation of post-depositional processes, however, at present they have not been widely applied to tephra deposits in our network.

Once an integrated tephrostratigraphy is defined, shard peaks are selected for geochemical analysis to allow the assessment of volcanic source and deposit integrity. Peaks were selected using criteria akin to those used to pinpoint samples for high-resolution analysis, i.e., discreteness relative to background concentrations, replication across grain-size fractions and processing and analysis time (e.g. Figure 4b). On occasions, glass shards from long upward tails in deposition or secondary peaks were analysed to provide further insights into the nature of individual deposits (see Section 4.1).

4. Results

4.1 Classification of individual tephra deposits

We utilised the approach outlined above to construct a tephrostratigraphic record for all cores within our network and tephra deposits were identified in the vast majority of records. Glass shard concentration profiles, geochemical characterisations and other indicators, such as shard size and co-variance with IRD, were integrated for these cryptotephra deposits to define a deposit type classification scheme (Table 2). Six deposit types that each share similar physical characteristics reflecting common modes of delivery and post-depositional

reworking are identified (Table 2). This classification scheme is mainly based on deposits of brown glass shards (i.e. basaltic material) due to the relative lack of colourless shard deposits. However, Type 3, is an exception and is based on deposits that are most commonly associated with colourless shards related to NAAZ II, the most widespread silicic tephra found within our core network.

Deposit Types 1,2 and 3 are all characterised by distinct concentration peaks, however, their profiles vary in form, displaying discrete (e.g. Figure 5a(i)), bell-shaped (e.g. Figure 6a(i)) and asymmetric (e.g. Figure 7a(i)) forms, respectively, and in vertical spread ranging from 1 cm to up to 100 cm (Table 2). These contrasting features are attributed to variable shard concentrations between the deposit types and differential influence of post-depositional reworking. For instance, the low shard concentrations of Type 1 deposits contribute towards their discreteness. Whilst this discreteness may result from limited post-depositional reworking, it is also possible that the low concentration of glass shards deposited at the seabed is not an adequate tracer of such activity. Reworking such as bioturbation, however, would most likely not impact the isochron position (see Section 3). In contrast, the higher glass shard concentrations associated with Type 2 deposits allows the glass shards to act as a tracer for bioturbation (e.g. Ruddiman and Glover, 1972; Griggs et al., 2015), which creates the upward and downward tails in deposition and roughly bell-shaped profile. This shard distribution pattern has often been viewed as the classic form of tephra deposits preserved in marine records (e.g. Ruddiman and Glover, 1972).

For Type 3 deposits the extremely high shard concentrations rapidly isolated underlying sediment from bioturbative activity and restricted downward migration of shards, as observed for the FMAZ II deposit in Griggs et al. (2015). The upward tail and continued deposition of

glass is primarily attributed to secondary deposition of glass shards from the same volcanic event from the surrounding sea-bed due to bottom current transportation. Bioturbative reworking may have also contributed towards increasing the overall vertical spread of these deposits. In combination these two factors create the observed asymmetric profile (e.g. Figure 7a(i); Table 2). Additional samples in the overall declining concentration profile of Type 3 deposits were sometimes analysed, particularly when subsidiary peaks were observed, in case any subsequent volcanic events were obscured within the upward tail. In all instances these additional analyses had an identical composition to shards in the main peak, corroborating the assertion that the upward tail was formed mainly through reworking of material from a single eruption (e.g. Figure 7a(i)).

Deposit Types 1, 2 and 3 are most likely derived from single depositional events, yet their isochronous nature can only be fully determined by assessing the relative homo/heterogeneity of their geochemical signature. Type 1 and 3 deposits have a homogeneous major element signature, i.e. all analysed shards form a single geochemical population most likely sourced from one volcanic eruption, which strongly suggests that they were deposited via primary fall and are useful isochronous tephra markers (e.g. Figure 5a(ii) for Type 1 deposits and Figure 7a(i) for a Type 3 deposit). Type 2 deposits are sub-divided into Type 2A, which have a homogeneous composition, and Type 2B, which have a heterogeneous composition with the analysed shards forming multiple populations and/or revealing a wide spread of analyses with high variability and limited consistency. Figures 6a and 6b provide examples of homogeneity based on major element analyses for two Type 2A deposits, whilst, Figure 5b provides examples of the heterogeneity observed for two Type 2B deposits. This sub-categorisation is important as the homogeneous Type 2A deposits are likely to be isochronous, akin to Type 1 and 3 deposits, while the heterogeneity of Type 2B deposits most likely reflects the

deposition of products from multiple eruptions and probably secondary transport processes that affect the isochronous nature of the horizons. For example, geochemical heterogeneity is a key indicator of transport via iceberg rafting and/or the amalgamation of the products of closely timed eruptions (Griggs et al., 2014). An additional line of evidence for Type 2B deposits is co-variance of shard concentrations with IRD records. The relative proportion of shards across the different grain-size fractions can also help determine transport processes because sea-ice rafting typically transports shards larger than would be expected via primary fallout to distal sites (e.g. Austin et al., 2004). Overall, for Type 2 deposits a careful assessment of a range of key indicators is required to determine their value as isochronous deposits.

In contrast to the single concentration peaks displayed by deposit Types 1, 2 and 3, Type 4 deposits display multiple peaks over a period of elevated shard concentrations whereas Type 5 deposits are characterised by glass shards in multiple consecutive samples, but with no clear pattern or peaks in shard concentrations (Table 2). In most cases, the multiple peaks seen in the Type 4 deposits display similar major element geochemical signatures but they are typically heterogeneous, e.g. the 456-473 cm depth deposit widely dispersed in MD04-2820CQ (Figure 7b; Abbott et al., 2016). This compositional pattern indicates that the entire deposit is an amalgamation of eruptive material from several, closely-timed, volcanic eruptions and that the multiple peaks are the product of secondary transport processes (e.g. bioturbation and bottom current reworking) rather than primary fallout. Alternatively, the glass shards found in Type 4 deposits may have been amalgamated during deposition on the Icelandic ice-sheet and subsequently transported to core sites via iceberg rafting. As with Type 2B deposits, further insights into the mode of deposition may be gained by comparing shard concentration profiles with iceberg rafting proxies. Without a distinct concentration

peak or geochemical evidence that they were sourced from a single eruption, Type 4 deposits typically cannot be utilised as isochronous marker horizons for high-precision correlations. However, they have the potential to be used as regional marine-marine core tie-lines, as suggested for FMAZ III by Abbott et al. (2016).

Type 5 deposits are commonly identified during low-resolution investigations. Only selected deposits were re-evaluated at a high-resolution and for geochemical composition. No distinct concentration peaks were identified, and geochemical analyses revealed heterogeneous populations of shards that were geochemically identical to those of underlying deposits, e.g. NAAZ II. As such, Type 5 deposits are interpreted as a background of glass shards that are deposited at the core sites and dispersed in the sediment column by remobilisation and reworking processes. These background signals vary between sites and may mask and hamper the identification of primary fall events that resulted in deposition of glass shards only in low concentrations. High-resolution analysis coupled with intensive geochemical characterisation may isolate such events and would be appropriate if specific volcanic events were being targeted. However, this was not feasible within our extensive core network.

4.2 Categorising core sequences using the tephra classification scheme

The tephra classification scheme has been employed to categorise the cores according to the presence and dominance of different deposit types. Four core categories have been identified (Figure 8) and range from sites dominated by primary fall deposits (sites marked in green) to sites with deposits affected by secondary processes (sites marked in red). In addition, very few shards were identified in the northernmost (JM04-25PC from the Western Svalbard slope) and southernmost (MD01-2444 from the Iberian Margin) records. Trace amounts (1-2

shards) were identified in some low-resolution samples but none were replicated as significant deposits during high-resolution analysis.

4.2.1 Core dominated by Type 1 deposits

Only two marine sequences exclusively contain Type 1 deposits, MD04-2822 from the Rockall Trough and MD04-2829CQ from the Rosemary Bank (Figure 8). The Type 1 deposits comprise discrete peaks in brown shard concentrations constrained vertically within ~1 cm and both sites have a limited background of brown shards over the zone of interest (e.g. Figure 5a(i)). Shards from the discrete peaks have single homogeneous geochemical populations that can be directly related to single volcanic source regions (Figure 5a(ii)) and hence likely represent isochronous marker horizons. The shard concentrations were low (~5-40 shards per 0.5 g dws in the 25-80 μm fraction) and occasionally replicating these peaks to extract shards for geochemical analysis was challenging. This lack of replication may be a consequence of the uneven distribution of shards within the cores. However, the successful identification of these Type 1 deposits does demonstrate how the approach adopted in this work can be used to trace such low concentration deposits.

4.2.2 Cores containing single occurrences of Type 2A deposits

Two cores, MD95-2010, from the Norwegian Sea, and MD01-2461, from the Porcupine Seabight, each contain just one significant tephra deposit with bell-shaped shard concentration profiles (Figure 6a(i) and b(i)). These deposits were identified because significant numbers of shards were isolated over 10-20 cm intervals in the low-resolution counts. Given their homogeneous geochemical compositions, these are both classified as

Type 2A deposits (Figure 6a(ii) and b(ii)) and are thus thought to be isochronous markers. Evidence of upward reworking within MD01-2461 is seen as a small subsidiary shard peak, positioned 4-5 cm above the highest shard concentrations, with an identical geochemical composition at both depths (Figure 6b). In both cores only trace amounts (<2-3) of shards were present in the rest of the low-resolution samples, apart from ~10 shards identified around NAAZ II in MD95-2010.

4.2.3 Cores containing mixed deposit types

Five of the core sites have been grouped into this category (Figure 8) and contain a range of deposit types. Type 2 deposits dominate and these are typically relatively discrete with high shard concentrations, but the geochemical compositions range between homogeneous (Type 2A) and heterogeneous (Type 2B). Type 4 deposits are also present in some sequences and at most sites the rhyolitic component of NAAZ II is present as a Type 3 deposit. The MD04-2820CQ record is a prime example of this mixed category. It contains a number of Type 2 deposits, with differing geochemical homogeneity, the FMAZ III as a Type 4 deposit and the NAAZ II rhyolitic component as a Type 3 deposit (Abbott et al., 2016). The variability in tephra deposit types means that a careful assessment of individual deposits is required and strongly suggests that the depositional controls at sites in this category varied temporally throughout the last glacial period.

4.2.4 Core dominated by Type 2B and Type 4 deposits

Two cores have been grouped within this category, SU90-24 from the Irminger Basin and M23485-1 from the Iceland Sea (Figure 8). These sites are characterised by multiple glass

concentration peaks within a high background level of shards, e.g. 1,000-10,000s of shards per 0.5 g dws. Peaks in shard concentration are not well-resolved in these records and the distinct contrast between SU90-24 and a Type 1 dominated core (MD04-2822) is shown in Figure 5. For SU90-24, single-shard analyses from some of the concentration peaks have highly heterogeneous geochemical signatures, with a wide range of major oxide values that span several different Icelandic volcanic systems (Figure 5b). Given the shard concentration profiles and compositional results, these deposits are classified as Type 2B and Type 4. M23485-1 is dominated by Type 4 deposits with two major depositional pulses of heterogeneous basaltic and rhyolitic material. Overall, the deposits found in these cores cannot be considered as isochronous horizons.

5. Discussion - Controls on Ash Deposition and Preservation

The core categorisation highlights that a diverse range of tephrostratigraphies were preserved during the last glacial period across the North Atlantic. Geographical clustering of similar core sites suggests that there were both spatial and temporal controls on ash deposition.

Various factors could have controlled the transport and deposition of tephra, including (i) the nature of volcanism inputting tephra into the system, (ii) atmospheric dispersal patterns and distance from eruptive source, (iii) rafting by icebergs and sea-ice and (iv) the rate and nature of sedimentation. Local factors may have also operated at individual cores sites. Through an assessment of these factors we propose that for our core categories common controls operating within different sectors of the North Atlantic can be identified (Figure 9).

5.1 Frequency and composition of Icelandic volcanism

The marine tephra records are ultimately controlled by the nature and frequency of Icelandic eruptions as these provide the primary input of tephra into the North Atlantic. Currently the most well-resolved record of Icelandic eruptions during the glacial period is derived from the Greenland ice-cores (Bourne et al., 2015) as proximal records are relatively limited due to the removal of material by glacial activity and the burial of deposits by subsequent volcanic activity. Within the Greenland ice-cores over 99 tephra or cryptotephra deposits have been identified in this time-period, which is significantly higher than the number identified within our marine tephra framework but could suggest that some of the marine deposits have amalgamated material from multiple eruptions (e.g. FMAZ III in JM11-19PC and MD04-2820CQ; see Figure 7b). Within our core network there is a greater abundance of basaltic horizons in comparison to rhyolitic deposits, which is consistent with the Greenland ice-core records, where 95 % of the deposits are basaltic (Bourne et al., 2015). This dominance of far-travelled basaltic material within distal sites could be due to the increased ice cover during the last glacial period which implies that the horizons were derived from subglacial phreatomagmatic eruptions, which can enhance the explosivity of basaltic eruptions due to the presence of water (Larsen and Eiríksson, 2008). The relative lack of rhyolitic horizons in the ice-cores suggests that the rhyolitic background of shards observed in many of the marine records is most likely due to reworking of material from NAAZ II, rather than resulting from subsequent volcanic activity.

5.2 Atmospheric dispersal patterns and proximity to Iceland

Following a volcanic eruption, the wind-driven dispersal patterns will largely dictate the location of fall deposits. The proximity of a core site to the volcano is important as the grain-size, shard concentration and thickness of airfall deposits decreases exponentially away from

the eruptive source. Atmospheric transport skews this relationship with extended transport of material along transport axes downwind from the eruptive source and this bias is more evident at distal sites (Sparks et al., 1981; Pyle, 1989; Lacasse, 2001).

The four cores solely preserving deposits thought to be transported via primary ashfall (i.e. sites marked green and orange containing Type 1 and Type 2A deposits: MD95-2010, MD04-2829CQ, MD04-2822 and MD01-2461; Figure 9) are located between the south and east of Iceland. This oceanic sector stretches from the south coast of Ireland to the west coast of Norway, with the two green sites containing multiple deposits lying close together towards the SE off the west coast of Scotland (Figure 9). Other sites that preserve a mix of deposit types including some deposited via atmospheric transport, i.e. yellow coded sites, also generally lie to the south and east of Iceland with the exception of MD95-2024 (Figure 9). This clustering of sites suggests that tephra was transported from Iceland via westerly winds, consistent with dominant wind patterns and typical plume heights of Icelandic eruptions.

Modern observations indicate that over Iceland wind direction changes progressively with altitude in the troposphere, with easterlies dominating at ground level shifting to southerly at a low level (1.4 km) and westerlies in the upper troposphere and lower stratosphere between 9-15 km throughout the year (Lacasse, 2001). Above 15 km altitude seasonal variability is observed with strong westerlies during the autumn and winter and relatively weak easterlies during the spring and summer (Lacasse, 2001). The modern atmospheric patterns are utilised as an analogue for dispersal of tephra during the glacial period as the reconstruction of glacial wind patterns is uncertain. Studies do suggest, however, that surface circulation was more intense over the North Atlantic during the last glacial period (e.g. Kutzbach and Wright, 1985; Mayewski et al., 1994;). Plume heights from modern basaltic eruptions similar in

nature to those that occurred during the last glacial period (e.g. Vatnajökull 1996, Hekla 2000, Grímsvötn 2004 and 2011 and Eyjafjallajökull 2010) were typically between ~8-15 km with some reaching 25 km altitude (Gudmundsson et al., 2004; Höskuldsson et al., 2007; Kaminski et al., 2011; Oddsson et al., 2012; Petersen et al., 2012). For older eruptions, Lacasse (2001) deduced from proximal and distal grain sizes that the Saksunarvatn Ash, erupted from Grímsvötn in the early Holocene, produced an eruption column of at least 15 km elevation. Eruptive plume heights together with dominant wind directions suggest that basaltic tephra was mainly atmospherically transported away from Iceland in an easterly direction, which is consistent with our findings.

Southward atmospheric dispersal of some tephras, to core sites such as MD99-2251 and MD04-2820CQ, may be a consequence of modification by more variable surface wind conditions that reflect the weather at the time of an eruption (Lacasse, 2001). A similar scenario was observed for the Eyjafjallajökull 2010 eruption, with weather conditions exerting a strong influence following initial easterly transport of tephra (Davies et al., 2010). Other variable influences such as precipitation, the timing of the eruption, style of volcanism, magma discharge rate and height of eruptive column may have also created differences from the general pattern for individual eruptions. Although our observations indicate some dispersal towards the south, no tephra deposits were preserved in the southernmost site MD01-2444, most likely as a consequence of the long distance between this site and the main Icelandic source.

Preferential atmospheric transport of ash to the east and south of Iceland is also consistent with the identification of tephra fall deposits or cryptotephra deposits from Iceland in terrestrial deposits from sites in northwest Europe (e.g. Lawson et al., 2012) and their absence

to the west and southwest of Iceland (e.g. Greenland – Blockley et al., 2015; eastern North America - Pyne-O'Donnell et al., 2012; Mackay et al., 2016). Cryptotephra is preserved at the most westerly site, MD95-2024. This core is downwind and the second farthest from Iceland with greater peak and background shard concentrations relative to closer and downwind sites such as MD04-2829CQ and MD04-2822. This cryptotephra occurrence conflicts with the expected atmospheric dispersal pattern of tephra and proximity to source, strongly indicating that other processes were at times helping to control tephra delivery to the North Atlantic west of Iceland.

The observation of limited atmospheric dispersal in a northerly direction from Iceland has some conflicts with the observations of Bourne et al. (2015), who inferred direct transport of ash in a north-westerly direction to the Greenland ice-sheet (Figure 9). However, this conflict could be a consequence of marine sites north of Iceland being more dominantly influenced by other controls, such as ice-rafting deposition of tephra (see discussion below), which masked any isochronous primary fall deposits. The distance from source was highly likely to be a dominant control on the non-preservation of tephra at the most northerly site JM04-25PC.

Overall, therefore, while atmospheric transport was the primary mechanism delivering tephra to the sites marked in green and orange on Figure 9 it was only a partial control on the delivery of tephra to the sites marked in yellow. At those locations other controls had an additional influence, leading to the identification of some diachronous deposits.

5.3 Ice-Rafting of Tephra and Ocean Currents

The potential for tephra to have been rafted either by sea-ice or icebergs prior to deposition in the glacial North Atlantic has been highlighted previously and this process can transport material along different trajectories and further from the source than atmospheric dispersal. Three distinct areas that preserve tephra deposited by rafting processes, i.e. deposit Types 2B and 4, have been identified. These areas are the Iceland Sea and Irminger Basin to the north and west of Iceland (core sites M23485-1 and SU90-24), the mid Atlantic (MD95-2024, MD99-2251, GIK23415-9, MD04-2820CQ) and northeast of the Faroe Islands (JM11-19PC). Whilst the Iceland Sea and Irminger Basin were heavily influenced by these processes throughout the 60-25 cal ka BP period, both Type 2A and Type 2B deposits were preserved in the other two areas suggesting that the influence of rafting was temporally variable (Figure 8).

Surface ocean currents have a huge role to play in the trajectory of tephra-bearing sea-ice and icebergs away from Iceland (Bigg et al., 1996) and thus influence the deposition of tephra at core sites during melting. Modern surface ocean currents are illustrated on Figure 9 and are used as an approximate analogue for the glacial period. The North Atlantic Drift (NAD) from the southwest dominates the warm surface ocean currents and splits into the Irminger Current south of Greenland and the North Iceland Irminger Current around Iceland before flowing into the Nordic Seas. Cold currents are dominated by the East Greenland Current flowing down the east coast of Greenland. A distinct feature of the surface circulation is the subpolar gyre, an anti-clockwise ocean surface circulation south of Iceland (Figure 9). These surface ocean currents would have strongly influenced ice-rafting but the source of icebergs and sea-ice extent was also an important factor.

The expanded size of the LGM ice-sheet over Iceland suggests that ice calving margins could have been located all around the island (Figure 9). With the majority of the major volcanic centres located in the south of the island, icebergs from the southward margin may have contained a greater concentration of tephra, however, local atmospheric transport north, east and west of the volcanoes would have contributed material to icebergs calving from all of these margins. The circulation patterns shown in Figure 9 suggests that icebergs from all margins could have been transported in surface ocean currents. Sea-ice reconstructions have shown that its extent over the North Atlantic region varied in time with the DO and Heinrich events (Hoff et al., 2016). It has been suggested that sea-ice retreated abruptly during the warming at the start of interstadials, but spread rapidly from the coast of Greenland during interstadial cooling with perennial sea-ice extending beyond Iceland during cold stadials and reaching a greater extent during Heinrich events (Figure 9; Hoff et al., 2016). This temporal variability in sea-ice coverage and its rafting along similar trajectories to those proposed for icebergs is likely to have played a role in the dispersal of tephra.

Iceberg rafting from the north coast of Iceland was the likely primary control on tephra deposition north and west of Iceland. The M23485-1 site lies close to the northern margin of the LGM Icelandic ice sheet and icebergs calved from this margin could have been entrained within the East Greenland Current and deposited material over the SU90-24 site. In addition, sea-ice rafting may have contributed towards this pattern of tephra deposition as the latter site lies within the stadial perennial ice-sheet limits and would have been covered early in the advances during interstadial cooling phases. Within the mid-Atlantic area, Icelandic icebergs transported in the subpolar gyre are likely to have deposited material at both the MD95-2024 and MD99-2251 sites. The MD04-2820CQ and GIK23415-9 sites lie within the IRD Belt, an area of the North Atlantic within which IRD from the Laurentide Ice Sheet was deposited

during Heinrich events, and may have been influenced by Icelandic icebergs transported in this zone by surface currents (Figure 9). Indeed, glass shards have been found in association with the lithic Heinrich layers (e.g. Obrochta et al., 2014). The influence of sea-ice rafting in the mid-Atlantic would have been temporally variable throughout the glacial period and hence should not be ruled out as a potential process for ash transport and deposition as MD95-2024 and MD99-2251 lie close to the stadial perennial sea ice limit and MD04-2820CQ and GIK23415-9 lie close to the Heinrich event limit (Figure 9). The area to the northeast of the Faroe Islands, the JM11-19PC site, may have been influenced by both rafting processes, with icebergs transported from the north coast of Iceland in the North Iceland Irminger Current and it lies close to the limit of perennial sea-ice during stadial periods. For all sites potentially affected by rafting processes key indicators such as the level of geochemical heterogeneity and shard sizes should be utilised to assess the origin of individual deposits.

The lack of rafted deposits in the MD04-2822 and MD04-2829CQ cores may be due to the Rockall Trough, the main pathway by which the warm North Atlantic surface water flows northward into the Norwegian Sea, effectively isolating them from the influence of Icelandic icebergs. The sites lie close to the stadial perennial sea ice limit so could be susceptible to sea ice rafting. However, the tephrostratigraphic records strongly indicate that this process has not deposited glass at these particular sites. Continuous sea-ice cover can be ruled out as a potential control on the lack of tephra preservation at the northerly JM04-25PC site. The reconstructed sea-ice limits from Hoff et al. (2016) suggest that although the site is the most northerly, sea-ice cover was limited to stadial phases and Heinrich events and was not greater than at other sites, e.g. SU90-24 and M23485-1, which contain significant cryptotephra deposits (glass shard concentrations) (Figure 9).

5.4 Nature and Rate of Sedimentation

Sedimentation rates are a further important control on tephra preservation. They provide information on the nature of sedimentation and slower rates of sedimentation increase the likelihood that the products of separate but closely timed eruptions are amalgamated. Table 1 presents approximate average sedimentation rates for all the sites in the core network between 60-25 cal ka BP. In general, all the sites had relatively high sedimentation rates, a bias created by our prioritisation of sites to include in the network (see Section 2.1).

These high sedimentation rates may indicate that, in addition to sedimentation occurring through pelagic settling, bottom currents were also transporting material to the sites (Rebesco et al., 2014). Thus, the sites incorporated in the network may have an increased susceptibility to secondary deposition of tephra-derived glass shards via bottom current reworking. This process could account for the persistent low background levels of glass shards at most sites (Type 5 deposits) and occasional outlying single shard analyses in the cryptotephra deposits (see Abbott et al., in revision). However, bottom current reworking does not appear to have been a significant control on the nature of these tephra records. The only deposit type that we interpret as being formed and affected by bottom current reworking is Type 3, which can be attributed to the exceptionally high peak shard concentrations in comparison to the other deposit types (Table 2). Almost exclusively, Type 3 deposits are NAAZ II occurrences, unique deposits formed by an event that led to the input of a sufficient concentration of shards into the oceanic system to be reworked and act as a tracer for bottom current activity. As with bioturbation, the lack of evidence of reworking for other deposits does not definitively demonstrate that this process was not occurring, because glass shard tephra

concentrations could have been too low to be detected using our methods (i.e. insufficient to act as an adequate tracer).

There is no clear difference in sedimentation rates between the cores containing only isochronous deposits (i.e. green and orange marked sites) and those dominated by heterogeneous secondary deposits (i.e. red marked sites) with estimated rates of 14-20 cm/ka and 17-19 cm/ka respectively (Table 1; Figure 8). However, in general the sites containing a mix of deposit types (yellow marked sites; Figure 8) have lower sedimentation rates, between 9-11 cm/ka, apart from the MD95-2024 site which had a rate of 22 cm/ka (Table 1). This contrast in sedimentation rates is a general reflection of these cores deriving from the deepest sites in the network, away from terrestrial sediment sources and the higher sedimentation rates observed on continental shelves (Figure 9). The low sedimentation rates may have contributed towards the preferential occurrence of Type 2B and Type 4 deposits at these sites due to the increased likelihood of closely spaced eruptive products being amalgamated. With Icelandic basaltic tephra horizons in the Greenland ice-cores having an average recurrence interval of ~1 per 200 years during this period (Bourne et al., 2015), and 200 years being represented by ~2 cm depth at the sites depicted in yellow (Figure 9) it is highly likely that closely spaced eruptions were mixed. The lower sedimentation rates would also have contributed to slower upward migration of the bioturbation mixing zone, promoting the amalgamation of deposits and vertical elongation of the shard concentration profile for Type 2 deposits. Each deposit must be evaluated individually as these sites may also be heavily influenced by rafting processes, which can produce Type 2B deposits with geochemical heterogeneity. Overall, the lower sedimentation rates and thus temporal resolution at all these sites could account for the lower number of cryptotephra horizons identified within the

marine core network in comparison to the numbers in the Greenland ice-core records (see Abbott et al., in revision for further discussion).

5.5 Local Site Conditions

Based on their proximity to Iceland, atmospheric dispersal patterns and tephra rafting in the North Atlantic, it might be expected that MD95-2010 and MD01-2461 would both contain a number of cryptotephra deposits. Each, however, only contained a single tephra deposit, the FMAZ IV in MD95-2010 and NAAZ II in MD01-2461, strongly suggesting that another factor was limiting the deposition of tephra at these sites. Both sites lie close to the former limits of LGM ice sheets and are amongst the shallowest sites in the network (Figure 9; Table 1). Higher levels of terrigenous sediment deposition might have masked or diluted the glass shard concentrations at these sites, especially if the non-tephric material was coarse-grained and/or dense because the shard concentrations presented in this work are referenced to sediment mass.

5.6 Summary

In general, whilst only a small area of the North Atlantic was disposed to solely preserving isochronous Type 1 and Type 2A deposits, these primary tephra-fall deposits can also be preserved in a wide area to the east and south of Iceland due to prevalent atmospheric dispersal patterns. Only a small area to the north and west of Iceland does not preserve any isochronous primary fall deposits. We suggest that the most significant factor complicating the tephrostratigraphic records is the rafting of tephra within icebergs and sea-ice, which can be constrained spatially but also displays temporal variability, particularly at sites within the

central North Atlantic. In addition, the high frequency of Icelandic volcanic eruptions during the period provides a constraint on the number of tephra. Despite our focus on sites with relatively high sedimentation rates, the rates are potentially still too low to enable individual events to be resolved within the sediment cores.

6. Conclusions

This work provides an integrated methodology for the identification of cryptotephra in North Atlantic marine records alongside a protocol for assessing the integrity of deposits and the influence of primary and secondary transport and depositional processes based on a classification scheme for glass shard deposits. A widespread network of cores was studied and six key glass shard deposit types with common physical characteristics and depositional and transport histories were identified in these records. The deposit types range from valuable ash-fall deposited isochronous horizons, to geochemically heterogeneous deposits with complex histories, to persistent background signals of ash deposition. While the variety of deposit types observed in the glacial North Atlantic reflects the complexity of processes controlling the transport, deposition and post-depositional reworking of tephra and may be unique to this setting, the methodological approach for identification could underpin investigations in other oceanic regions.

A regional analysis of the tephrostratigraphic records has shown that a range of different controls influenced tephra deposition and the deposit types preserved as glass-shard concentrations at different sites within the North Atlantic over the last glacial period. A key area to the southeast of Iceland was sheltered from any ice-rafting influence and only isochronous primary fall deposits have been isolated in these records. However, primary

deposits were also identified in a wide oceanic sector between the south and east of Iceland, which could be the focus of future studies to identify further isochronous horizons or to extend the distribution of those identified within this work. The wider significance of the isochronous horizons identified in this work is discussed in Abbott et al. (in revision), which defines a framework of marine tephra horizons for the 60-25 cal ka BP period in the North Atlantic region.

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Figures Captions

Figure 1: Flow chart of the transportation and depositional processes that could have affected tephra within the glacial North Atlantic prior to preservation in marine sediments. Adapted from Griggs et al. (2014).

Figure 2: Network of North Atlantic marine cores studied within this work and ice-cores mentioned within the text.

Figure 3: Flow chart of the consistent methodology utilised to determine the glass shard content of cores within the marine network and to extract and prepare such shards for major element geochemical analysis. NaOH = sodium hydroxide. SPT = sodium polytungstate.

Figure 4: Example of the construction of a tephrostratigraphy using the MD99-2251 core. (a) Low-resolution brown glass shard concentration profiles split into three grain-size fractions. Blue bars denote depth intervals reinvestigated at a 1 cm sampling resolution in the core. (b) Integrated high and low resolution brown shard counts for the MD99-2251 core. Shard counts have been truncated for clarity. Shard counts in the 1686-1687 cm sample (*) are 4991, 1862 and 507 shards per 0.5 g dws in the 25-80, 80-125 and >125 μm grain-size fractions, respectively. The shard counts for the 25-80 μm grain-size fraction from the 1904-1905 cm sample (**) are 3776 shards per 0.5 g dws. Red bars denote samples depths from which glass shards were subsequently extracted for compositional characterisation by EPMA.

Figure 5: Comparison of (i) tephrostratigraphic records and (ii) compositional characterisations of glass shard deposits from the (a) MD04-2822 and (b) SU90-24 marine

sequences. Brown shard counts for the 25-80 μm grain-size fraction from 470-500 cm in SU90-24 have been truncated for clarity. Shard counts exceed 40,000 shards per 0.5 g dws, but, two peaks could be identified at 480-481 cm and 486-487 cm. Percentage abundance of *Neogloboquadrina pachyderma* (sinistral) record for MD04-2822 from Hibbert et al. (2010). Magnetic susceptibility record for SU90-24 from Elliot et al. (2001). Geochemical fields for Icelandic source volcanoes are based on normalised whole rock and glass shard analyses utilised in Bourne et al. (2015) and references within and additional data for the Kverkfjöll volcano from Gudmundsdóttir et al. (2016). Within MD04-2822 additional discrete peaks can be observed, e.g. at 1731-1732 cm and 1965-1966 cm. However, it was not possible to acquire sufficient material for geochemical characterisation. All geochemical data plotted on a normalised anhydrous basis.

Figure 6: Examples of shard concentration profiles and geochemical characterisations for Type 2A tephra deposits from two North Atlantic marine records within the network. (a) MD95-2010 (i) 910-920 cm high-resolution tephrostratigraphy of brown glass shards, (ii) compositional variation diagrams of analyses from glass shards extracted from the 915-916 cm depth sample. Chemical classification and nomenclature for total alkalis versus silica plot after Le Maitre et al. (1989) and division line to separate alkaline and sub-alkaline material from MacDonald and Katsura (1964). Geochemical fields for Icelandic tholeiitic volcanic systems defined using normalised whole rock and glass shard analyses from Jakobsson et al. (2008) (Reykjanes), Höskuldsson et al. (2006) and Óladóttir et al. (2011) (Kverkfjöll) and Jakobsson (1979), Haflidason et al. (2000) and Óladóttir et al. (2011) (Grímsvötn and Veidivötn-Bardabunga). (b) MD01-2461 (i) 940-950 cm high-resolution tephrostratigraphy of colourless glass shards (ii) total alkalis versus silica plot of analyses from glass shards extracted from the 947-948 cm depth sample. Normalised compositional fields for the

Icelandic rock suites derived from whole rock analyses in Jakobsson et al. (2008). All geochemical data plotted on a normalised anhydrous basis.

Figure 7: Examples of shard concentration profiles and geochemical characterisations for a (a) Type 3 and a (b) Type 4 deposits from two North Atlantic marine records within the network. (a) MD99-2251 (i) 1950-2030 cm tephrostratigraphy of colourless glass shards integrating low and high-resolution shard counts (ii) compositional variation diagrams comparing characterisations of colourless glass shards from 1974-1979 cm and 2014-2015 cm depth. (b) MD04-2820CQ (i) 450-480 cm high-resolution tephrostratigraphy of brown glass shards (ii) compositional variation diagrams comparing characterisations from four shard peaks within the Type 4 deposit. Data from Abbott et al. (2016). Chemical classification and nomenclature for total alkalis versus silica plot after Le Maitre et al. (1989) and division line to separate alkaline and sub-alkaline material from MacDonald and Katsura (1964). All geochemical data plotted on a normalised anhydrous basis.

Figure 8: Classification of core sites within the marine core network. See Section 4.2 for details of classes.

Figure 9: Primary controls and influences on the deposition of tephra within the glacial North Atlantic Ocean. Ocean surface currents and names from Voelker and Haflidason (2015) and Rasmussen et al. (2016). Currents: IC = Irminger Current; NIIC = North Iceland Irminger Current; EGC = East Greenland Current; EIC = East Iceland Current; NAD = North Atlantic Drift; SPG = Subpolar Gyre. Last Glacial Maximum (LGM) ice limits from Dyke et al. (2002), Funder et al. (2011) and Hughes et al. (2016). Perennial sea ice limits from Hoff et al. (2016). Core classification from Figure 7.

Supplementary Information

Table S1: Original major oxide concentrations of shards from tephra deposits in the MD04-2822 core. Deposits analysed are from the depths of (i) 1836-1837 cm (ii) 2004-2005 cm and (iii) 2017-2018 cm.

Table S2: Original major oxide concentrations of shards from tephra deposits in the SU90-24 core. Deposits analysed are from the depths of (i) 340-342 cm (ii) 420-422 cm (iii) 480-481 cm and (iv) 486-487 cm.

Table S3: Original major oxide concentrations of shards from the MD95-2010 915-916 cm tephra deposit.

Table S4: Original major oxide concentrations of shards from MD01-2461 related to the rhyolitic component of North Atlantic Ash Zone II (II-RHY-1). Deposits analyses are at (i) 942-943 cm and (ii) 2014-2015 cm depth.

Table S5: Original major oxide concentrations of shards from MD99-2251 related to the rhyolitic component of North Atlantic Ash Zone II (II-RHY-1). Deposits analyses are at (i) 1974-1975 cm and (ii) 947-948 cm depth.

Table S6a: Original secondary standard analyses of the BCR2g standard made throughout analytical periods during which sample glass shard analyses presented in this work were analysed.

Table S6b: Original secondary standard analyses of the Lipari standard made throughout analytical periods during which sample glass shard analyses presented in this work were analysed.

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Table 1: Details of the North Atlantic marine core network investigated in this study. Approximate sedimentation rates cover the 60-25 cal ka BP period for the cores, except for MD04-2829CQ which covers the 41-25 cal ka BP period, and were calculated using existing age-depth models for the sequences or approximated based on ages for event markers e.g. Heinrich events and North Atlantic Ash Zone II.

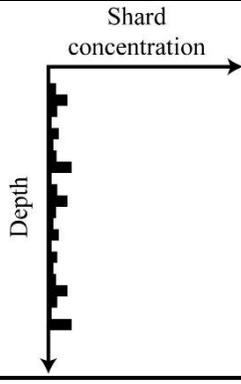
Core	Location	Lat/Long	Water depth	Approx. average sedimentation rate (cm/ka)	Example references
JM04-25PC	Western Svalbard slope	77° 28' N, 09° 30' E	1880 m	10	Jessen et al. (2015)
M23485-1	Iceland Sea	76° 54.9' N, 17° 52.4' W	1120 m	17	-
MD95-2010	Norwegian Sea	66° 41.05' N, 04° 33.97' E	1226 m	16	Dokken and Jansen (1999)
JM11-19PC	North Faroe Slope	62° 49' N, 03° 52' W	1179 m	11	Ezat et al. (2014); Griggs et al. (2014)
SU90-24	Irminger Basin	62° 40' N, 37° 22' W	2100 m	19	Elliot et al. (1998, 2001)
MD04-2829CQ	Rosemary Bank	58° 56.93' N, 09° 34.30' W	1743 m	20	Hall et al. (2011)
MD04-2822	Rockall Trough	56° 50.54' N, 11° 22.96' W	2344 m	14	Hibbert et al. (2010)
MD99-2251	Gardar Drift	57° 26' N, 27° 54' W	2620 m	11	-
MD95-2024	Labrador Sea	50° 12.40' N, 45° 41.22' W	3539 m	22	Stoner et al. (2000)
GIK23415-9	Northern North Atlantic	53° 10.7' N, 19° 08.7' W	2472 m	9	Weinelt et al. (2003)
MD01-2461	Porcupine Seabight	51° 45' N, 12° 55' W	1153 m	13	Peck et al. (2006, 2008)
MD04-2820CQ	Goban Spur	49° 05.29' N, 13° 25.90' W	3658 m	11	Abbott et al. (2016)
MD01-2444	Iberian Margin	37° 33.68' N, 10° 08.53' W	2637 m	23	Martrat et al. (2007)

Table 2: Summary of the shard profiles, characteristics, transportation and deposition processes of cryptotephra deposit types common to North Atlantic marine sequences between 60-25 cal ka BP. ★ = position of the isochron for deposit type.

Deposit type	Typical shard profile	Deposit type characteristics	Transport and deposition processes
TYPE 1 Low concentration peak		<ul style="list-style-type: none"> -Well constrained shard concentration peak -Low shard concentrations (< 50 per 0.5 g dws) -Shards generally 25-80 μm in diameter -Homogenous geochemical composition 	<ul style="list-style-type: none"> -Single depositional event -Sourced from a single volcanic eruption -Potentially limited post-depositional reworking -Most likely primary airfall deposition
TYPE 2 High concentration peak		<ul style="list-style-type: none"> -Distinct peak in shard concentration -High shard concentrations (100s-1,000s per 0.5 g dws) -Upward and downward spanning up to 10 cm -Homogenous (Type 2A) or heterogeneous (Type 2B) geochemical composition 	<ul style="list-style-type: none"> -Analysis of geochemistry, shard sizes and IRD required -Bioturbative reworking -Single depositional event -Transport via primary airfall, sea-ice and iceberg rafting possible
TYPE 3 High concentration peak; gradational upward tail		<ul style="list-style-type: none"> -Flat bottomed profile with a clear gradational upward tail -Very high shard concentrations (100,000s-1,000,000 per 0.5 g dws) -Deposit spread up to 100 cm -Homogenous composition of shards in peak 	<ul style="list-style-type: none"> -Single depositional and volcanic event -Reworking via secondary deposition and bioturbation -Transport via primary airfall or sea-ice rafting -Useful isochron
TYPE 4 Diffuse distribution; multiple peaks		<ul style="list-style-type: none"> -High shard concentrations (1,000s-1,000,000s per 0.5 g dws) -Multiple peaks in concentration in a period of elevated shard concentrations -Deposit spread of 10s of cms -Heterogeneous geochemical composition common between peaks 	<ul style="list-style-type: none"> -Deposition of multiple closely spaced eruptions or deposition via iceberg rafting -Comparison to Greenland tephra framework and IRD records required -Potential as regional marine-marine tie-lines

TYPE 5

Background of consistent concentration



- Consistent deposition of shards with limited variability in concentrations between samples
- Wide variability of deposit spreads
- Heterogeneous or geochemically related to underlying deposits

- Background signal of glass shards
- Shards reworked and remobilised in the oceanic system
- Potential masking of low concentration glass shard deposits

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Research Highlights

- Methodology for the identification and assessment of marine cryptotephra presented
- Deposit type scheme defined based on glass shard profiles and chemical signatures
- Physical characteristics of deposit types indicative of common transport histories
- Ice rafting the most significant factor complicating tephrostratigraphic records
- Wide sector to the south and east of Iceland a repository for isochronous horizons

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