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Tracing marine cryptotephras in the North Atlantic during the Last Glacial Period:
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      Improving the North Atlantic marine tephrostratigraphic framework
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19
      Abstract
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      Tephrochronology is increasingly being recognised as a key tool for the correlation of
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      disparate palaeoclimatic archives, underpinning chronological models and facilitating
      climatically independent comparisons of climate proxies. Tephra frameworks integrating both
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      distal and proximal tephra occurrences are essential to these investigations providing key
      details on their spatial distributions, geochemical signatures, eruptive sources as well as any
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      available chronological and/or stratigraphic information. Frameworks also help to avoid mis-
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correlation of horizons and provide important information on volcanic history. Here we

present a comprehensive chronostratigraphic framework of 14 tephra horizons from North

Atlantic marine sequences spanning 60-25 cal ka BP. Horizons previously discovered as

visible or coarse-grained deposits have been combined with 11 newly recognised volcanic

assessed using their physical characteristics. All horizons originated from Iceland with the

methods to a wide network of marine sequences. Their isochronous integrity has been

vast majority having a basaltic composition sourced from the Grímsvötn, Kverkfjöll,

deposits, identified through the application of cryptotephra identification and characterisation

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Hekla/Vatnafjöll and Katla volcanic systems. New occurrences, improved stratigraphic 35 36 placements and a refinement of the geochemical signature of the NAAZ II are reported and the range of the FMAZ IV has been extended. In addition, several significant geochemical 37 populations that further investigations could show to be isochronous are reported. This tephra 38 framework provides the foundation for the correlation and synchronisation of these marine 39 40 records to the Greenland ice-cores and European terrestrial records to investigate the phasing, rate, timing and mechanisms controlling the rapid climate changes that characterised the last 41 42 glacial period.

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Keywords: Quaternary; palaeoceanography; tephrochronology; North Atlantic; tephra
framework; marine cores

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47 1. Introduction

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Tephrochronology, the use of volcanic ash deposits as isochronous tie-lines between 49 50 disparate palaeoclimatic records, is increasingly being utilised as a key geochronological tool for reconstructing the timing and phasing of past climatic events (e.g. Lowe, 2011; Lowe et 51 52 al., 2012; Lane et al., 2013; Davies, 2015). This upsurge is directly linked to advances in cryptotephra analysis, which has dramatically increased the number of potential tie-lines and 53 54 led to the compilation of regional tephra frameworks (e.g. Lowe et al., 2008; Tryon et al., 2009; Zanchetta et al., 2011; Davies et al., 2012; Abbott and Davies, 2012; Lowe et al., 55 56 2015). Tephrostratigraphical frameworks typically include a compilation of key information relating to the tephra horizons within them, including their spatial extent, based on 57 preservation within palaeoclimate records, glass shard concentrations, glass shard 58 composition and eruptive source alongside chronological and stratigraphic information (e.g. 59 60 Lowe et al., 2008; Davies et al., 2014; Bourne et al., 2015; Matthews et al., 2015). The most comprehensive frameworks include both distal and proximal tephra findings, visible and 61 cryptotephra occurrences and combine newly discovered data with previously published 62 deposits. Integrating all this information can provide valuable frameworks for the volcanic 63 64 history of a region and provide key reference tools for future studies. Distal archives are often more complete than proximal records, which are prone to removal or burial of deposits, 65 although proximal archives can often record more information regarding eruptions, such as 66 their full geochemical evolution. In addition, developing the most comprehensive tephra 67 68 frameworks will help to reduce instances of mis-correlation which can occur if volcanic

regions produce multiple, closely-timed eruptions with similar geochemical compositions(e.g. Lowe, 2011; Bourne et al., 2013).

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For the North Atlantic region, various detailed frameworks spanning a range of time-intervals 72 73 are currently available. For example, Gudmundsdóttir et al. (2016) provides a proximal 74 framework of Icelandic eruptions during the Holocene, Blockley et al. (2014) summarises the 75 European tephra stratigraphy over the last glacial cycle and Davies et al. (2014) provides an integrated framework of MIS 5 tephras in Greenland ice-cores and North Atlantic marine 76 77 records. The tephra framework for the Greenland ice-cores has significantly expanded in recent years (e.g. Mortensen et al., 2005; Abbott and Davies, 2012; Davies et al., 2014), in 78 particular over the MIS 2-3 period (Bourne et al., 2015), highlighting the value of exploring 79 these distal archives. In comparison, however, only a limited number of tephra horizons have 80 been identified in North Atlantic marine records spanning MIS 2-3 (see Haflidason et al., 81 2000; Wastegård et al., 2006; Section 2). This relative paucity is despite considerable 82 advances in distal tephrochronology and the high potential for a tephra framework from these 83 84 sequences to be used to establish correlations to the Greenland ice-cores and European terrestrial records. Such correlations could help answer key questions regarding the relative 85 86 timing of atmospheric and oceanic changes associated with the rapid climatic events, that punctuated the region during the last glacial period (e.g. NGRIP Members, 2004; Bond et al., 87 88 1993; Martrat et al., 2007; Hall et al., 2011; Zumaque et al., 2012; Henry et al., 2016). 89

90 Here we present a tephra framework for North Atlantic marine records spanning MIS 2-3, which is underpinned by our investigations of an extensive core network (Figure 1) using 91 92 recently developed cryptotephra identification methods (Abbott et al., in revision). Prior studies are also reviewed (Section 2) and previously identified isochronous horizons are 93 94 integrated with our new cryptotephra discoveries. This integration represents the most concerted attempt to improve the tephra framework for the North Atlantic, and overall a 95 framework of 14 marine tephra or cryptotephra horizons from between 60-25 cal ka BP has 96 been defined (Figure 2). 97

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99 2. Prior North Atlantic Tephra Investigations between 25-60 ka BP

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101 It was highlighted earlier that tephra frameworks should integrate all isochronous tephra
102 deposits from a region, so the framework presented in this work integrates our new

discoveries alongside previously published data from multiple cores sites from the North 103 Atlantic (green sites on Figure 1). Within these prior tephrochronological studies of the MIS 104 2-3 period, several isochronous tephra horizons have been identified, i.e. North Atlantic Ash 105 Zone II (NAAZ II), Faroe Marine Ash Zone (FMAZ) II and FMAZ IV. Reviewing the 106 literature does, however, highlight some of the challenges associated with determining the 107 108 isochronous nature of deposits and the limitations of earlier studies that only focused on the coarse fraction (>150 µm) of the marine sediments. These were the major factors driving the 109 development of a procedure for isolating fine-grained cryptotephras (down to 25 µm 110 111 diameter) and interpreting transportation and depositional processes (e.g. Abbott et al., 2011, in revision; Davies et al., 2014; Griggs et al., 2014). This is essential to determine the 112 isochronous nature of fine-grained, cryptotephra deposits for which macro-sedimentary 113 evidence cannot be utilised to determine the relative influence of primary and secondary 114 processes. These methods were utilised by Abbott et al. (2016) to identify three previously 115 116 undocumented MIS 2-3 volcanic events within a core retrieved from the Goban Spur (see Section 4 for details) and are more widely applied in this study. 117

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119 The first MIS 3 tephra deposit to be recognised in the North Atlantic was NAAZ II, initially

identified by Bramlette and Bradley (1941) and later described by Ruddiman and Glover

121 (1972). NAAZ II is a complex ash zone composed of the products of several Icelandic

eruptions (see Section 4.1.1) with rhyolitic material from one eruption (II-RHY-1) the most

123 widespread, being traced into multiple marine cores and the Greenland ice-cores (e.g.

124 Kvamme et al., 1989; Grönvold et al., 1995; Lacasse et al., 1996; Zielinski et al., 1997;

Haflidason et al., 2000; Austin et al., 2004; Svensson et al., 2008). The widespread nature of

126 II-RHY-1 gives rise to a key tie-line between North Atlantic marine records and the

127 Greenland ice-cores within the North Atlantic tephra framework (Austin and Abbott, 2010).

128

The FMAZs comprise a series of ash zones identified in cores around the Faroe Islands 129 130 region, and three, II, III and IV, were deposited during MIS 2-3. Two of these, FMAZ II and IV, have isochronous characteristics and are integrated within the framework (Figures 1 and 131 2; Rasmussen et al., 2003; Wastegård et al., 2006; Wastegård and Rasmussen, 2014; Griggs 132 et al., 2014). FMAZ II was described by Wastegård et al. (2006) as a visible horizon and was 133 suggested to be a widespread primary fall deposit. The FMAZ II was subsequently traced into 134 the NGRIP ice-core by Davies et al. (2008) (NGRIP 1848 m; $26,740 \pm 390$ yr b2k), providing 135 a clear demonstration of the high potential for ice-marine correlations between the Greenland 136

137 ice-cores and North Atlantic marine sequences during the 60-25 cal ka BP period. FMAZ IV

138 was first described by Wastegård and Rasmussen (2014) as a layer up to 20 cm thick

deposited shortly after warming related to Dansgaard-Oeschger (DO) event 12. Due to its

140 homogeneous composition and micro-sedimentary features (Griggs et al., 2014, 2015) it has

141 been interpreted as a primary ashfall deposit.

142

FMAZ III, identified as a thick relatively dispersed zone of tephra spread over ~20 cm depth 143 in the Faroes cores, was also thought to have a correlative in the NGRIP core (NGRIP 144 145 2066.95 m; 38,122 ± 723 yr b2k; Davies et al., 2010). However, Bourne et al. (2013) subsequently identified a series of closely-spaced tephra horizons in the NGRIP and NEEM 146 ice-cores around NGRIP 2066.95 m, many with geochemical compositions that fall within 147 the wide geochemical envelope of FMAZ III. This highlighted the complexity of the period 148 and demonstrated that the suggested correlation was inappropriate and did not represent an 149 150 ice-marine tie-line (Bourne et al., 2013). Bourne et al. (2013) and Griggs et al. (2014) both suggested that FMAZ III formed through the amalgamation of several separate tephra-fall 151 152 events and low sedimentation rates at the core sites so the diachronous deposits are not incorporated in the marine tephra framework. 153

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Early studies of North Atlantic tephra mainly focused on investigating visible tephra horizons 155 156 or glass shards present within the coarse fraction of the sediment (i.e. >150 µm diameter). This may have created a bias towards the identification of horizons from large scale eruptions 157 and/or horizons not deposited via primary ash-fallout (Brendryen et al., 2010; Abbott et al., 158 2011). The study of Lackschewitz and Wallrabe-Adams (1997) highlights the limitation of 159 this approach. Several ash zones above NAAZ II were identified within and correlated 160 between a series of cores from the Reykjanes Ridge, however, most of these deposits have 161 heterogeneous geochemical compositions and in general coincide with distinct peaks in ice-162 rafted debris (IRD). Based on these factors Lackschewitz and Wallrabe-Adams (1997) 163 164 concluded that this material was transported to the sites via iceberg rafting. This process could have significantly delayed the deposition of these deposits and, hence, they do not 165 166 represent isochronous marker horizons and are not incorporated in the marine tephra framework. The only deposit with isochronous characteristics was the X peak, a discrete high 167 concentration peak within VZ 1 in the SO82-5 core, with a homogeneous glass composition 168 and no coeval IRD peak. This horizon was subsequently correlated to FMAZ II by Wastegård 169 170 et al. (2006) (Figure 2).

Voelker and Haflidason (2015) utilised the coarse sediment fraction to define a high-172 resolution tephrostratigraphy for the last 86 ka from the southern Greenland Sea PS2644 core. 173 This sequence was interpreted as containing a record of 68 volcanic events between ~60-25 174 cal ka BP based on the geochemical analysis of glass shards from 28 depths in the core. The 175 176 volcanic events, however, are sometimes defined based on a limited number of geochemical analyses of deposits with multiple glass-based geochemical populations/events often 177 identified at the same depth. According to protocols for assessing deposits this heterogeneity 178 179 could be indicative of deposition via iceberg rafting and/or secondary depositional processes (Abbott et al., in revision), however, while these processes were acknowledged a distinction 180 between tephra deposited via primary or secondary process is often not made. This may have 181 led to the overreporting of the number of isochronous deposits present so the deposits from 182 these volcanic events are not incorporated into the North Atlantic tephra framework presented 183 184 here. However, it is important to note these findings as a reappraisal of these deposits together with IRD evidence may well reveal the presence of dominant populations and 185 186 valuable isochrons in the future.

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188 3. Methodology

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190 *3.1 Detecting, characterising and correlating cryptotephra deposits*

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192 A widespread network of North Atlantic cores was investigated (Figure 1) and we applied the consistent methodological approach for cryptotephra identification outlined in Abbott et al. 193 194 (in revision). Following preliminary low-resolution analysis, high-resolution glass shard concentration profiles were gained from the core deposits. The major element composition of 195 196 peaks in glass shard concentrations were characterised using electron-probe micro-analysis (EPMA) with at least 20-40 individual shards from each deposit analysed (see Abbott et al., 197 198 in revision for full description). For all analysis and data comparison, the major element data were normalised to an anhydrous basis, i.e. 100 % total oxides, however, the raw 199 200 geochemical data are provided in the Supplementary Data alongside secondary standard analyses (Table S12). Potential sources for geochemical populations and tephra or 201 cryptotephra horizons were explored through graphical comparison of the composition of 202 individual shards with glass and whole rock analyses from proximal Holocene Icelandic 203 204 deposits from the three different rock suites and specific volcanic systems. We acknowledge

that some centres may have geochemically evolved or not been productive during the lastglacial period, therefore, the potential sources proposed here may need to be revised.

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Potential cross-correlations between all the isochronous horizons and significant glass shard 208 209 derived geochemical populations in cores within the network and other marine records were 210 explored using statistical comparisons of their average geochemical signature and graphical comparisons on bivariate plots. The similarity coefficient function (SC) of Borchardt et al. 211 (1972) was utilised to construct a matrix for all these comparisons (Table S13). Twenty-five 212 213 of the comparisons returned SC values greater than 0.97, which implies there are strong 214 similarities in the geochemical signatures and further assessment was required to determine if they are correlatives. A combination of three main factors were used to rule out most of these 215 comparisons as potential correlatives: large stratigraphic discrepancies, subtle geochemical 216 217 differences, and occurrence at different depths in the same core sequence. Despite the 218 majority being ruled out, upon further assessment two of the comparisons with high SC values were found to have very strong geochemical similarities and consistent stratigraphic 219 220 positions and are suggested as correlatives between marine sequences in the network (see Section 4). 221

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223 *3.2 Assessing the isochronous nature of cryptotephra deposits*

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Several of the deposits reported here have been described in Abbott et al. (in revision) as 225 226 illustrative examples for assessing the dominant controls on tephra deposition in the North Atlantic region. We synthesise these results in a framework of tephra deposits that represent 227 isochronous marker horizons identified using protocols set out in Griggs et al. (2014) and 228 Abbott et al. (in revision). The key characteristics used to define isochronous horizons are: (i) 229 230 a clear peak in the shard concentration profile that can be used as the isochron position and (ii) a homogeneous geochemical population or distinct trend in glass shard analyses 231 232 indicative of material deriving from a single volcanic eruption. Abbott et al. (in revision) outlines a tephra deposit type scheme that uses glass shard concentration profiles and 233 geochemical homogeneity/heterogeneity to identify six North Atlantic marine tephra deposits 234 types with common modes of tephra delivery and post-depositional reworking. Here that 235 scheme is utilised to aid the assessment of the deposits identified in the marine records 236 Although Type 1 and 3 deposits are typically characterised by single homogeneous 237 populations there is greater variability and complexity in the geochemical signatures of Type 238

2 deposits. For the latter a larger number, typically >30 but on occasions up to 60, of single-239 grain major element analyses were acquired. These were graphically assessed to explore the 240 relative homogeneity or heterogeneity of deposits, define homogeneous populations that may 241 have derived from single eruptions, quantify their relative dominance within the deposits and 242 categorise them as Type 2A or Type 2B deposits. Outliers were defined as analyses that were 243 not consistently associated with a defined population. For some heterogeneous deposits 244 where populations were not identifiable analyses were grouped based on affinities to the 245 Icelandic rock suites (see Supplementary Figures). 246

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- 248 *3.3 Age and stratigraphic constraints*
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The timing of deposition for each tephra deposit is given based on the available 250 climatostratigraphy for the specific core within which the horizons were isolated (Table 1). 251 For some records, there is strong stratigraphic control based on proxy records from the cores 252 that record the DO events which characterised the North Atlantic region during the last 253 254 glacial period, e.g. MD04-2822 and MD04-2829CQ. However, for other cores, e.g. MD99-2251 and GIK23415-9, the stratigraphic frameworks are not as distinct with deposits from the 255 256 Heinrich events providing the best stratigraphic control. Due to uncertainties in the relative timing of closely spaced horizons not identified in the same core sequence the stratigraphic 257 relationships presented in Figure 2 should be treated with caution, e.g. the cluster of horizons 258 that have been identified in various cores around the H4 event (Figure 2). Further 259 260 investigations of these horizons, such as their tracing into other sequences, may help to refine the sequence of the volcanic events in the future. 261

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263 4. North Atlantic Tephra Framework

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An improved marine tephra framework for the North Atlantic between 60-25 cal kyr BP is presented in Figure 2 and Table 1. Overall, a framework of 14 isochronous horizons can be defined, including 8 new isochronous horizons presented for the first time, 3 cryptotephra deposits identified in MD04-2820CQ by Abbott et al. (2016) and 3 previously published deposits (NAAZ II, FMAZ IV and FMAZ II). This new framework represents a significant increase in the number of tephra marker horizons that could be utilised for the correlation of records during this period.

With the exception of NAAZ II (II-RHY-1) and MD04-2820CQ 497-498 cm, all tephras in 273 the framework are basaltic in composition and originated from Iceland, specifically from the 274 Grímsvötn, Kverkfjöll, Hekla/Vatnafjöll and Katla volcanic systems (Table 1). The most 275 widespread isochronous horizon in the framework is the NAAZ II (II-RHY-1) (Figures 3 and 276 4). The wide distribution and importance of this horizon had been established in prior studies, 277 however, here we have isolated it in more sequences, gained greater control on the timing of 278 279 deposition, with peaks in shard concentration determined at a 1 cm resolution, and provided an improved glass geochemical signature for the horizon (Section 4.1.1). The geographical 280 281 range of the previously identified FMAZ IV can be expanded, to a limited extent, from the Faroe Islands region to the Norwegian Sea following its identification in MD95-2010 (Figure 282 5; Section 4.1.2). 283

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Within our network only two cores, MD04-2822 and MD04-2829CQ, exclusively preserved 285 isochronous Type 1 deposits (Figures 6a and 6b). New isochronous horizons were also 286 identified in two further cores, MD99-2251 and GIK23415-9, alongside other deposits 287 without clear isochronous characteristics, i.e. Type 2B and Type 4 deposits (Figures 7a and 288 8a), which can be attributed to temporal variability in the processes controlling tephra 289 290 deposition at these sites (see Abbott et al., in revision). Further details regarding all the isochronous horizons are provided in Section 4.1 in chronological order from the oldest to the 291 youngest horizon. 292

293

The Type 2B and Type 4 horizons are not overlooked though as analysis showed that within many of these deposits significant homogeneous geochemical populations could be isolated (Figures 7b and 8b; Table 1). These populations are presented alongside the framework of isochronous horizons as their geochemical homogeneity suggests that they were derived from single volcanic events, but, at present, questions remain over their depositional origin and isochronous nature. Further investigations, however, may permit their integration into the regional tephra framework and this is discussed further in Section 4.2.

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302 *4.1 Isochronous horizons*

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304 *4.1.1 NAAZ II*

306 NAAZ II is a crucial deposit within the North Atlantic marine tephra framework and it has

- 307 been identified at nine sites within our network as a clear peak in rhyolitic material and at 6
- 308 sites basaltic/intermediate material was also present. Based on occurrences of NAAZ II in
- 309 several North Atlantic sites, this ash zone was defined as being composed of five
- 310 geochemical populations, one rhyolitic (II-RHY-1) and four basaltic (II-THOL-1, II-THOL-
- 311 2, II-THOL-3 and II-TAB-1) by Kvamme et al. (1989).
- 312

Shards from the peaks in rhyolitic material at the 9 sites have a consistent homogeneous 313 314 transitional alkali rhyolitic composition (Figure 3a(i) and 4b; Table S2). In comparison to prior characterisations of NAAZ II from several North Atlantic marine cores, strong 315 similarities can be observed for some oxides, e.g. FeO and CaO (Figure 3bi) but some offsets 316 are apparent for other oxides, e.g. Na₂O and SiO₂ (Figure 3bii). These differences are 317 reflected in similarity coefficient comparisons (Table S2) and are consistent with sodium loss 318 319 affecting the older EPMA analyses (Hunt and Hill, 1993; Kuehn et al., 2011), particularly for the analyses from Kvamme et al. (1989), and are highly unlikely to indicate a different source 320 321 for the material. Therefore, the nine deposits in this network can be correlated to the II-RHY-1 component of NAAZ II. These new analyses provide an up-to-date composition for this 322 323 component and highlight that data quality must be considered when assessing correlations between datasets, especially for rhyolitic material. 324

325

A peak in brown shards was isolated in direct association with the II-RHY-1 peak at 6 sites 326 327 (Figure 4b; e.g. in MD99-2251 (Figure 4a)). Compositional analyses revealed a range of signatures with basaltic and intermediate material present (Figure 3a(ii)). Shards related to 328 three of the basaltic populations of Kvamme et al. (1989) have been identified, but no shards 329 related to the II-THOL-3 population were isolated (Figure 3c). Glass shards with an 330 intermediate trachyandesite to trachydacite composition have been identified (Figure 3a(ii)) 331 and grouped as a new population, which we name II-INT-1. Some material with an 332 333 intermediate composition was found in association with the proximal Icelandic deposit correlated to NAAZ II, the Thorsmörk ignimbrite (Jørgensen, 1980). However, this is less 334 335 evolved than the material in these marine deposits with SiO₂ values of 56-58 % and is unlikely to be directly related. This additional intermediate population suggests that the 336 basaltic material associated with NAAZ II derives from more individual eruptions than 337 previously thought. This assertion is also supported by differences in the composition of 338 339 material from this study attributed to the populations of Kvamme et al. (1989) which may

- indicate they grouped material from multiple eruptions as single populations. For example,
- shards from M23485-1 and GIK23415-9 display geochemical differences, e.g. Figure 3cii,
- despite all falling into the II-THOL-2 field of Kvamme et al. (1989). At three of the sites the
- 343 brown shards can be grouped as single populations: homogeneous populations within the II-
- THOL-2 geochemical field in M23485-1 and JM11-19PC and only shards from the
- intermediate population are present in MD01-2461 (Figure 4c). The remaining three sites
- 346 preserve a mix of populations. MD04-2820CQ preserves three populations (II-THOL-1, II-
- THOL-2 and II-INT-1), each exceeding 24% of the shards present. GIK23415-9 and MD99-
- 2251 are dominated by the II-THOL-1 and II-TAB-1 populations, respectively.
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350 The contrast between the homogeneity of the rhyolitic material at all sites and the

- 351 heterogeneity and inconsistent signatures of the basaltic/intermediate material may indicate
- that despite coeval deposition the two components were transported differentially. It has been
- 353 suggested that NAAZ II was primarily transported from Iceland via sea-ice rafting and
- primary airfall (e.g. Ruddiman and Glover, 1972; Austin et al., 2004; Wastegård et al., 2006).
- 355 Sea-ice rafting may have contributed towards the relatively higher rhyolitic shard
- 356 concentrations at sites to the south and west of Iceland. The geochemical homogeneity and
- distinct peak with an upward tail in rhyolitic shard concentrations (i.e. Type 3 deposits; e.g.
- Figure 4a(i)), observed at all sites is consistent with these transport processes and supports
- the isochronous nature of the II-RHY-1 component.
- 360

The heterogeneity of the basaltic material and relative discreteness of the concentration 361 peaks, e.g. Figure 4a(ii), are consistent with transport via iceberg rafting and the between-site 362 contrasts in geochemical signatures highlights that icebergs calved from different margins of 363 the Icelandic ice sheet could have transported and deposited material at the core sites. The 364 absence of basaltic material associated with the rhyolitic peaks in the MD04-2822 and 365 MD95-2010 sites is consistent with the findings of Abbott et al. (in revision) that ice rafting 366 367 did not transport tephra to these sites during the last glacial period. Transportation via iceberg rafting can delay the deposition of tephra: therefore the peaks in basaltic material related to 368 369 NAAZ II should not be utilised as isochronous markers. However, based on their dominance as homogeneous populations at some sites, II-THOL-2, II-TAB-1 and II-INT-1 are regarded 370 as significant geochemical populations (Table 1). It cannot be ruled out that one or more of 371 the basaltic populations were deposited coevally via primary fallout with the rhyolitic 372 373 material, particularly at sites only containing one population. However, it is unlikely that this

process deposited all of the basaltic populations with subsequent amalgamation in the
sediment column, as shard concentrations profiles for that type of deposit (Type 4) typically
have a greater vertical spread within sequences and display multiple concentration peaks.

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The coeval deposition of the two shard types may indicate that the volcanic eruption that produced the rhyolitic tephra horizon triggered an ice-rafting event which deposited the basaltic material, but the resolution of the marine records under investigation here is insufficient to resolve this temporal phasing.

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4.1.2 FMAZ IV – MD95-2010 915-916 cm

384

FMAZ IV was identified in the MD95-2010 core from the Norwegian Sea as a discrete 385 deposit at 915-916 cm depth (Figure 5a). This deposit has a homogeneous basaltic glass 386 composition with affinities to the Icelandic tholeiitic rock suite and the products of the 387 Grímsvötn volcanic system. The glass composition of MD95-2010 915-916 cm is identical to 388 389 the characterisation of the JM11-19PC 542-543 cm deposit of Griggs et al. (2014) (Figure 5b; SC - 0.985), previously correlated to the FMAZ IV of Wastegård and Rasmussen (2014). 390 391 According to the age model and stratigraphy for MD95-2010 from Dokken and Jansen (1999), this layer has an age of ~44.45 cal ka BP and was deposited during the DO-12 event 392 based on the magnetic susceptibility record. This stratigraphic position and age estimate are 393 consistent with the work of Wastegård and Rasmussen (2014). This horizon has previously 394 395 not been identified outside the Faroe Islands region and, therefore, this discovery expands its geographical range in a northeasterly direction to the Nordic Sea. 396

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398 *4.1.3 MD04-2820CQ 524-525 cm*

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MD04-2820CQ 524-525 cm has previously been described by Abbott et al. (2016) where it
was identified as a clear peak in shard concentrations spanning ~6 cm depth. Geochemical
analyses of shards from this deposit form a homogeneous tholeiitic basaltic population
sourced from either the Grímsvötn or Kverkfjöll Icelandic volcanic systems. These
characteristics allow the deposit to be defined as Type 2A and, allied with a lack of direct
covariance with IRD, this deposit is thought to have been deposited via primary fallout
despite occurring during a period of elevated IRD concentrations (Abbott et al., 2016).

410 High-resolution analysis of MD04-2822 showed a well-constrained peak in brown glass

shards in all grain-size fractions at 2017-2018 cm depth (Figure 6a). According to the core

stratigraphy, this horizon was deposited during a stadial period prior to the warming

transition into DO-9 (Figure 6a). Shards have a homogeneous basaltic composition with

affinities to the Icelandic tholeiitic rock suite and the products of the Grímsvötn volcanicsystem (Figure 6c).

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417 *4.1.5 MD04-2820CQ 497-498 cm*

418

MD04-2820CQ 497-498 cm was identified as a small peak in colourless glass shards, during 419 a period of consistently elevated shard concentrations, deposited prior to DO-9 (Abbott et al., 420 2016). Shards from the peak have a transitional alkali rhyolitic composition and form a single 421 population with affinities to a number of distal tephra deposits previously attributed to the 422 423 Katla volcanic system (Abbott et al., 2016). This horizon is notable as it is the only other rhyolitic horizon within the marine tephra framework apart from the rhyolitic component of 424 425 NAAZ II (Table 1). Due to its homogeneity and the prevalence of shards in the 25-80 µm fraction, this deposit was interpreted as an isochronous horizon deposited via primary ashfall 426 427 (Abbott et al., 2016).

428

429 4.1.6 MD04-2820CQ 487-488 cm

430

431 Deposited just prior to Heinrich Event 4, MD04-2820CQ 487-488 cm was identified as a clear peak in brown glass shard concentrations across all grain size fractions spread over ~3 432 cm depth (Abbott et al., 2016). While some transitional alkali outliers are present within 433 shard analyses from this deposit, the vast majority of shards (~85 %) form a homogeneous 434 geochemical population with a tholeiitic basaltic composition and affinities to the Grímsvötn 435 volcanic system (Abbott et al., 2016). This homogeneous composition and a lack of 436 437 covariance of shard concentrations with IRD suggests it was not deposited via iceberg rafting. Deposition is likely to have occurred via primary fall, however, the high proportion of shards 438 in the coarser grain-size fractions (80-125 μ m and >125 μ m) in comparison to the 25-80 μ m 439 fraction may also indicate transport via sea-ice rafting. Neither transport process would 440

- 441 impart a significant temporal delay in deposition, therefore, MD04-2820CQ 487-488 cm is
 442 viewed as an isochronous deposit (Abbott et al., 2016).
- 443

444 4.1.7 MD04-2829CQ 934-935 cm and 930-931 cm

445

Two distinct and closely spaced peaks in brown glass shards were isolated in MD04-2829CQ 446 with concentrations of ~35 shards per 0.5 g dws in the 25-80 µm grain-size fraction (Figure 447 6b). Only a limited number of shards were isolated in one of the three samples between these 448 449 peaks. The stratigraphy for MD04-2829CQ indicates that these horizons were deposited during and just after the rapid warming into DO-8 (Figure 6b; Hall et al., 2011). Shards from 450 both peaks were geochemically analysed and the analyses revealed two homogeneous 451 basaltic populations with affinities to the Icelandic tholeiitic rock suite and the products of the 452 Grímsvötn volcanic system. However, there are distinct differences in Al₂O₃, FeO, CaO and 453 MgO between the two deposits (Figure 6c). These differences show that despite being 454 separated by only 3 cm of sediment the horizons were produced by two separate volcanic 455 eruptions and, coupled with their other characteristics, can both be considered as valuable 456 isochronous marker horizons. 457

458

459 4.1.8 MD04-2822 2004-2005 cm

460

High-resolution shard counts identified brown shards within the 25-80 and >125 µm grain-461 size fractions in the 2004-2005 cm sample of MD04-2822 (Figure 6a). While the shard 462 concentrations are low the peaks are discrete as no further shards were identified in adjacent 463 464 samples. Based on the stratigraphy of the core this material was deposited shortly after the warming transition into DO-8 (Figure 6a; Hibbert et al., 2010). Geochemical analysis shows 465 that shards from the deposit have a homogeneous transitional alkali basaltic composition 466 (Figure 6c). The shards are characterised by high TiO₂ values of ~4.65 % wt and comparisons 467 to proximal Icelandic deposits demonstrate that the deposit was most likely sourced from the 468 Katla volcanic system (Figure 6c). The geochemical composition of the material in this peak 469 is markedly distinct from the material in the underlying MD04-2822 2017-2018 cm horizon, 470 indicating that they represent two discrete eruption events. 471

472

473 *4.1.9 MD99-2251 1680-1681 cm*

- The highest brown shard concentrations in MD99-2251 were identified as a peak centred
 around 1680-1681 cm depth (Figure 7a). Overall, high shard concentrations associated with
- this peak cover approximately 10 cm depth, typical of a Type 2 deposit, and glass shards
- 478 from the main peak and a secondary peak at 1683-1684 cm were geochemically analysed.
- 479

480 Shards from 1680-1681 cm form a clear near-homogeneous population, with 76 % of the analyses in this population (Figure 7b). High TiO₂ concentrations in excess of 4.4 % wt 481 strongly indicate an origin from the Katla volcanic system (Figure 7b). Within the remaining 482 483 25 % of shards a minor population (6 %) of tholeiitic material, most likely sourced from the Kverkfjöll volcanic system, was also identified alongside several outlying shards (Figure 7b). 484 The significant dominance of a single homogeneous population in the 1680-1681 cm peak, 485 suggests that this material was deposited via primary ashfall and that this tephra deposit 486 represents an isochronous marker horizon despite being deposited during a period of elevated 487 488 IRD concentrations associated with Heinrich Event 3 (Figure 7a).

489

The glass-derived geochemical signature of material from the underlying 1683-1684 cm peak is the same as that of the major 1680-1681 cm peak suggesting that this does not represent an earlier and separate depositional event but instead represents downward reworking of material from the main concentration of glass. The slight deviation of the shard concentration profile from a gradational downward tail could imply that any reworking processes were not uniform across the core. Such variability was observed by Griggs et al. (2015) in 3D reconstructions of the structure of tephra deposits gained using X-ray microtomography.

498 *4.1.10 MD04-2829CQ 800-801 cm*

499

500 The highest shard concentrations in core MD04-2829CQ were identified at 800-801 cm, with increases observed in all grain-size fractions (Figure 6b). This deposit is very discrete with 501 limited shards identified in adjacent samples. Stratigraphic constraints indicate that this 502 horizon was deposited in the cold period prior to DO-4 (Figure 6b; Hall et al., 2011). 503 504 Compositional analysis of individual shards shows that all material has a tholeiitic basaltic composition and can be grouped into two homogeneous populations, with clear bimodality 505 observed for some oxides, including TiO₂, FeO, CaO and MgO (Figure 6c). Analyses 506 grouped into population THOL-1 were only derived from shards from the 25-80 µm grain-507 508 size fraction, whereas the majority of analyses in population THOL-2 are from shards >80

µm in diameter. Based on comparisons to proximal Icelandic deposits, THOL-1 has a close
affinity to products of Grímsvötn while THOL-2 is most likely derived from the Kverkfjöll
volcanic system (Figure 6b; e.g. Óladóttir et al., 2011). This implies that the deposit was
formed from the deposition of material from two coeval eruptions of these volcanic centres. *4.1.11 MD04-2822 1836-1837 cm - GIK23415-9 225-226 cm*

515

Within the MD04-2822 record the largest peak in brown shards was identified at 1836-1837 516 517 cm depth with >40 shards per 0.5 g of dws present in the 25-80 μ m fraction (Figure 6a). The material is stratigraphically well constrained with only 2 shards present in the underlying 518 sample. According to the stratigraphy this material was deposited during the cold stadial 519 period shortly before the transition into DO-4 (Hibbert et al., 2010). Compositional analysis 520 of glass shows that material from this peak has a transitional alkali basaltic composition and 521 forms a homogeneous geochemical population (Figure 6c). Comparisons to proximal 522 Icelandic deposits indicate that the horizon was sourced from either the Katla or 523 524 Hekla/Vatnafjöll volcanic system (Figure 6c).

525

526 A discrete peak in shard concentrations, restricted to 1 cm and with the characteristics of a Type 1 deposit, was also isolated between 225-226 cm in GIK23415-9 (Figure 8a). 527 Geochemical analysis of the shards from this deposit shows that all have a transitional alkali 528 composition (Figure 8b). Within the analyses bimodality can be observed for some oxides, 529 530 most notably TiO_2 , and they can be split into two homogeneous populations. A dominant population (TAB-1) of 70 % of the shards with low TiO₂ values and a smaller population 531 532 (TAB-2) of 15 % of the analysed shards with TiO₂ values $\sim 0.35\%$ wt higher. TiO₂ values have been identified as one of the primary oxides that can be used to discriminate between 533 Icelandic basaltic eruptions from the last glacial period (e.g. Bourne et al., 2013, 2015). The 534 remaining 15 % of analyses are classified as outliers. Comparisons to proximal deposits show 535 that the populations have similarities to the products of both the Katla and Hekla/Vatnafjöll 536 volcanic systems (Figure 8b). GIK23415-9 225-226 cm was deposited during Heinrich Event 537 538 3 which could suggest it was deposited via iceberg rafting. However, the relative dominance of the TAB-1 population and a lack of direct covariance of shard concentrations with IRD, 539 with the discrete shard peak contrasting with elevated IRD concentrations for ~25 cm of core 540 depth, do not support this interpretation. These indicators provide support for primary ashfall 541

deposition of glass shards from either a single chemically bimodal eruption or two eruptionevents very close in time.

544

Statistical analysis (SC of 0.987) and graphical comparisons support a correlation between 545 MD04-2822 1836-1837 cm and GIK23415-9 225-226 cm (TAB-1) (Table S13; Figure 9a). In 546 547 addition, there is a consistency in the stratigraphic position of the two horizons. MD04-2822 1836-1837 cm was deposited between DO events 5 and 4 (Figure 6a), while GIK23415-9 548 225-226 cm was deposited at the end of Heinrich Event 3 (Figure 8a), which, based on a 549 550 comparison of ages for the Heinrich Events from Sanchez Goñi and Harrison (2010) and the Greenland ice-core chronology presented in Seierstad et al. (2014), occurred after Greenland 551 Interstadial (GI) 5, the ice counterpart to DO-5. Based on the available information, we assert 552 that these two deposits are the products of the same volcanic event and form a tie-line 553 between the two relatively closely spaced sequences (Figure 2). 554

555

556 4.1.12 GIK23415-9 173-174 cm

557

A peak in basaltic glass shard concentrations was identified in the GIK23415-9 core at a 558 559 depth of 173-174 cm, following Heinrich Event 2 (Figure 8a). The shard concentration profile of this deposit is akin to a Type 1 deposit with a relatively discrete peak in shard 560 561 concentrations restricted to ~1 cm (Figure 8a). Geochemical analysis of shards from this deposit show one clear homogeneous population, composed of 60 % of the analysed shards, 562 with a basaltic tholeiitic composition and an affinity to the Kverkfjöll volcanic system 563 (Figure 8b). The remaining 40 % are heterogeneous and can be regarded as outliers (Figure 564 S8). Although the overall homogeneity of the deposit is not as distinct as most Type 1 565 deposits, the occurrence of a homogeneous population deposited during a period of low IRD 566 input does suggest that primary fall occurred to form an isochronous deposit. The outlying 567 shards may derive from a low background of IRD input of ice-rafted shards during this 568 period. In addition, the use of the percentage abundance of populations to assess this deposit 569 has some limitations as only a low number of analyses, 15, were gained from shards within 570 this deposit. 571

572

573 4.2 Significant geochemical populations and possible isochrons

In addition to the isochronous deposits outlined in Section 4.1, six tephra deposits in the 575 MD99-2251 core and four in the GIK23415-9 sequence were assessed as having non-576 isochronous characteristics and have been classified as Type 2B or Type 4 deposits (Figures 577 7a and 8a). The main criterion underpinning this assessment was the geochemical 578 heterogeneity of the deposits, indicative of the amalgamation of material from a number of 579 580 volcanic eruptions. However, while only three deposits, MD99-2251 1654-1655 cm and 1796-1797 cm and GIK23415-9 193-194 cm, have fully heterogeneous compositions the 581 other deposits contain 16 significant homogeneous geochemical populations, in total, within 582 583 their overall heterogeneity (Figure 2; Table 1). The significant geochemical populations may relate to single volcanic eruptions, but due to their occurrence within heterogeneous deposits 584 further investigations are required to determine if they were deposited isochronously or 585 otherwise. The full glass-based geochemical signatures of all MD99-2251 and GIK23415-9 586 deposits and the populations identified within them are summarised in Figures S1-S14 and 587 588 Tables S8 and S10.

589

The 16 populations all have a basaltic composition and were sourced from Iceland. In addition to the volcanoes which deposited isochronous horizons in the North Atlantic region, i.e. Grímsvötn, Kverkfjöll, Hekla/Vatnafjöll and Katla, homogeneous glass shard populations with geochemical similarities to the products of the Veidivötn-Bardarbunga and Vestmannaeyjar volcanic systems were identified (Table 1; Figures 7a and 7b). Their relative dominance within the deposits is variable, ranging from ~10 to 60 % of the total single-shard analyses used to characterise the deposits (Tables S8 and S10).

597

Co-variance of shard concentration profiles with IRD records was another variable used to 598 assess the isochronous nature of the deposits (Abbott et al., in revision). Some of the deposits 599 600 with heterogeneous signatures were deposited during periods of elevated or rising IRD concentration, which could indicate transport via iceberg rafting and a significant temporal 601 602 delay between eruption and deposition. However, iceberg rafting is not the only process that can amalgamate the products of multiple eruptions. For example, for some deposits post-603 604 depositional mixing in the sediment column of the products of several closely-timed eruptions cannot be ruled out as some were isolated within periods of limited IRD deposition. 605 In this later scenario, deposition would have been via primary ashfall with no temporal delay, 606 however, determining the isochron position is challenging as complexity is often observed in 607 608 the shard concentration profiles. Primary fallout could also have occurred during a period of

ice-rafting deposition resulting in the incorporation of a homogeneous ashfall populationwithin a heterogeneous background rafted signal.

611

These differing scenarios and the uncertainty in the depositional processes implies that 612 further investigations are required to assess whether these populations are isochronous. 613 614 Consequentially we have reported the significant geochemical populations, but we have not incorporated them within the regional tephra framework until further evidence is gained. 615 Such evidence may include their identification in other North Atlantic marine cores and/or 616 617 the Greenland ice-core tephra framework in a similar stratigraphic position. In addition, for some records the covariance with IRD could not be fully explored because of the lower 618 resolution in this dataset relative to the shard concentration profiles. Improved high-619 resolution IRD records would be highly advantageous for further assessing depositional 620 processes. An example of how tracing these populations into other records could provide 621 622 further insights into their isochronous nature is provided within our work.

623

The assessment of potential correlations (Table S13) highlighted a strong similarity between 624 the glass-based geochemical signature of FMAZ II and the THOL-1 population in the 625 GIK23415-9 202-203 cm deposit (Figure 7; Table 1). The SC comparison returned a high 626 coefficient of 0.990, demonstrating that the signatures were nearly identical, and this 627 observation is corroborated by graphical comparisons (Figure 9b). Stratigraphically, FMAZ II 628 has been identified between Heinrich Events 3 and 2 in marine records and was deposited 629 prior to an increase in IRD concentrations in the ENAM93-21 core (Rasmussen et al., 2003) 630 and after GI-3 in the Greenland ice-core stratigraphy (Davies et al., 2010). GIK23415-9 202-631 632 203 cm was deposited during a period of increasing IRD concentrations related to the start of Heinrich Event 2 (Figure 8a). These stratigraphic juxtapositions are consistent and, coupled 633 with the strong geochemical similarities, could imply isochronous deposition from the same 634 volcanic event. GIK23415-9 202-203 cm (THOL-1) is one of 4 homogeneous geochemical 635 populations within the deposit and, due to their co-occurrence, it was interpreted as being 636 deposited by iceberg rafting. The proposed correlation does not contradict this interpretation 637 but could demonstrate that GIK23415-9 202-203 cm (THOL-1) was deposited via primary 638 ashfall during a period when tephra from other events was rafted by icebergs. Overall, this 639 potential correlation highlights the complexity of some deposits, but demonstrates how these 640 significant glass geochemical populations are important to consider as potential isochronous 641 642 markers.

644 5. Discussion

645

646

5.1 Future application of the North Atlantic marine tephra framework

647

648 The North Atlantic marine tephra framework between MIS 2-3 has been significantly 649 improved through the most extensive application of cryptotephra methods, comprehensive compositional analysis and rigorous and defined protocols to assess the isochronous nature of 650 each deposit. For a long period only a limited number of horizons had been identified in this 651 time period (Haflidason et al., 2000; Wastegård et al., 2006). Now this framework includes 652 14 isochronous horizons that have considerable promise for correlating and synchronising 653 palaeoclimatic records. There is also potential to add further isochronous markers given the 654 significant geochemical populations identified in heterogeneous deposits also reported in this 655 656 study.

657

658 NAAZ II remains a dominant tephra within this framework and our work has identified it in 659 numerous additional cores with greater control on the timing of deposition derived from highresolution shard counts and an improved geochemical signature for glass shards associated 660 661 with the widespread rhyolitic component (II-RHY-1). This tephra, with an age of $55,380 \pm$ 1184 yr b2k in the Greenland ice-core records (Svensson et al., 2008), represents a key 662 663 marker horizon for the period providing an isochronous tie-line linking numerous widespread marine cores and the Greenland ice-core records beyond the radiocarbon window. The 664 665 distribution of the FMAZ IV has been extended from the Faroe Islands region into the Nordic 666 Seas and has the potential to be a key tie-line for DO 12. However, despite being found 667 previously in several North Atlantic cores and the NGRIP ice-core (see summary map in Davies et al., 2012), the FMAZ II was only found in one additional core, GIK23415-9. 668 Furthermore, most of the new cryptotephras are single-core occurrences, highlighting 669 670 challenges with cryptotephra tracing within the North Atlantic Ocean. The limited tracing of horizons may reflect the difficulties of detecting and isolating deposits that often only contain 671 a low concentration of shards, but could also indicate the relatively constrained dispersal of 672 the basaltic eruptions depositing material over the North Atlantic. Only one correlation has 673 674 been made between newly identified isochronous horizons in the framework, MD04-2822 1836-1837 cm and GIK23415-9 225-226 cm (Section 4.1.11; Figure 2). These cores are 675 676 relatively closely spaced, supporting the suggestion of limited basaltic ash dispersal.

Assessing potential correlations between the records highlighted that while a range of factors 678 demonstrated that there are few direct correlations, many of the horizons have similar major 679 element geochemical signatures, especially eruptives from the Grímsvötn and Katla volcanic 680 systems (Table 1). This conclusion corroborates the findings of Bourne et al. (2015) who 681 682 observed similar repetition of major element glass geochemical signatures from these systems in tephra horizons in the Greenland ice-cores. This repetition is particularly notable for the 683 period around H4 as a cluster of six closely spaced horizons has been identified in the marine 684 685 cores (Figure 2). Of these, five horizons have similar tholeiitic glass major element basaltic compositions and are thought to be derived from the Grímsvötn volcanic system. However, 686 subtle differences in geochemical signatures show they represent individual events, which 687 may be further emphasised through trace element analysis (Lowe et al., 2017). 688

689

690 The observations that the new cryptotephras in the North Atlantic region may have limited dispersal and geochemical similarities do provide challenges for future correlation. There is, 691 692 however, the potential to constraint a number of rapid climate events, such as H4 and DO 8 and H3 as clusters of isochronous horizons are present around those events. Further 693 694 investigations should initially focus on sites close to those preserving the isochronous 695 horizons in this framework and/or re-evaluate previously explored sites (e.g. green sites on Figure 1), with adaptions to the methodological approach discussed in Section 5.3. It is 696 imperative that potential correlations are rigorously assessed as correlating horizons or 697 populations with close, but not identical, major element glass geochemical signatures, could 698 699 lead to the establishment of incorrect tie-lines between records. Trace element analysis of the 700 glass shards may aid this assessment as the additional signature may show greater differences between tephras (Lowe et al., 2017). Other supporting evidence such as broad stratigraphic 701 702 constraints and independent age estimates can also be used to support and test correlations. A detailed assessment of possible correlations to the Greenland ice-cores will be discussed in a 703 704 forthcoming publication whereby trace element signatures are also employed to assess and 705 support correlations.

706

707 *5.2 Reconstructing Icelandic volcanic history*

708

This framework adds to our understanding of the volcanic history of Iceland during the lastglacial period between 60-25 cal kyr BP. The dominance of basaltic over rhyolitic horizons

and the high productivity of the Grímsvötn/Kverkfjöll and Katla volcanic systems around 711 Heinrich Event 4 and Henrich Event 3, respectively, is consistent with the Greenland ice-core 712 tephra framework for the same period (Bourne et al., 2015). The dominance of basaltic 713 horizons in both sets of archives strongly suggests that differential dispersal of the products 714 of rhyolitic eruptions was not occurring and their paucity reflects a relatively lower frequency 715 716 of Icelandic rhyolitic eruptions during this period. Basaltic horizons potentially sourced from other volcanic centres were observed, including the Veidivötn-Bardabunga and 717 Vestmannaeyjar volcanic systems. There are very few or no tephras in the Greenland 718 719 framework with glass-based geochemical similarities to those horizons, potentially due to a bias in dispersal direction, a low number of eruptions from these sources and/or the nature of 720 volcanic eruptions from these systems. This observation shows that a more complete 721 reconstruction of Icelandic volcanism will be gained by integrating the two frameworks. 722 There is, however, a notable difference between the number of tephra deposits identified 723 between the marine and ice-core records. With 99 volcanic events recorded in the Greenland 724 records in contrast to 33 events in the marine archives, if the homogeneous populations are 725 726 assumed to derive from individual volcanic events. The lower resolution of the marine records, the potential for the amalgamation of airfall deposits, post-depositional reworking 727 728 processes and the masking of low concentration glass shard deposits (see below) are the most 729 likely causes of this disparity.

730

731 *5.3 Improving the marine tephra framework*

732

This work has demonstrated the potential of identifying isochronous cryptotephras in North 733 734 Atlantic marine records of the last glacial period. However, the methodology employed to identify cryptotephras in this work most likely created a bias towards the identification of 735 736 horizons depositing a high concentration of glass shards at core sites. As discussed by Timms et al. (2017), the process of completing low-resolution scans prior to a subjective peak 737 738 selection for high-resolution (1 cm) analysis may introduce a bias as low concentration or discrete peaks might not have sufficient shard concentrations to be observed in the low-739 740 resolution record. The background of shards that is prevalent at some marine sites could mask individual eruptions that deposited a low concentration of shards. In the ice-core records, 741 tephra events have been defined on the basis of as few as 3 shards (Bourne et al., 2015). 742 Detecting deposits of this kind would be particularly challenging in the marine environment 743 as they could be dismissed as "background" concentrations or hidden with the upward or 744

downward tail of a deposit or within an ash-rich deposit. We have attempted to explore the
presence of such horizons in this study but agree with Timms et al. (2017) who advocate the
use of more high-resolution shard concentration and glass-based chemical analyses to
improve tephrostratigraphies, while acknowledging that this may be limited by sediment
availability, time and financial considerations.

750

751 The marine tephra framework presented in this study should not be viewed as complete. However, by focusing on maximising the number and geographical range of sequences an 752 753 initial framework has been produced that is a significant step towards a more comprehensive tephra-based synchronisation of North Atlantic marine records. Coupling the success of the 754 methodology, the initial framework presented here and the insights into the spatial controls 755 on tephra deposition discussed in Abbott et al. (in revision), there is huge potential to add to 756 and refine the marine tephra framework. This can be achieved through focusing on new cores 757 from areas with a high potential to preserve isochronous horizons and reassessing previously 758 investigated cores at a high-resolution over key intervals during which isochronous horizons 759 760 were identified in this work. In addition, innovative techniques for the identification and quantification of tephra that are currently being developed, for example X-ray fluorescence 761 762 core scanning (e.g. Kolling and Bauch, 2017), hyperspectral core imaging (e.g. Aymerich et al., 2016) and automated flow cytometry and microscopy (e.g. D'Anjou et al., 2014), could 763 764 be tested and incorporated into the methodological approach if appropriate.

765

766 6. Conclusions

767

A consistent methodology for the identification and characterisation of marine cryptotephras 768 and the rigorous assessment of the influence of transportation and deposition processes on 769 770 tephra deposits were used to build an enhanced North Atlantic marine tephra framework. Eleven isochronous deposits were identified in a wide network of marine sequences and have 771 been integrated with prior data to create a marine tephra framework for the MIS 2-3 period. 772 773 Key information for each deposit, such as their spatial extent, geochemical signature, eruptive source and timing of deposition, is synthesised. A number of significant geochemical 774 populations are also reported that require further work to assess whether they originate from 775 single volcanic eruptions and were deposited isochronously via primary tephra fallout. 776 777

There is considerable potential to improve this framework by tracing the deposits into other 778 marine sequences, by identifying new deposits and/or gaining trace element characterisations 779 to aid the differentiation of closely-spaced horizons. Combining this framework with 780 knowledge of the processes controlling the deposition of tephra in the North Atlantic and the 781 identification of key areas where isochronous horizons are preserved provided in Abbott et al. 782 783 (in revision) these future investigations could be highly focussed, both temporally and spatially. The full potential of this framework will only be realised if attempts are made to 784 785 trace these horizons into other archives such as the Greenland ice-cores and terrestrial 786 records. If successful they can act as time-synchronous tie-lines to correlate and synchronise these palaeoclimatic records, providing insights into the phasing, rate, timing and 787 mechanisms forcing the rapid climate changes that characterised this period. 788

789

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791

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814 Figures

815

- **Figure 1:** Location map of cores within the marine network (red) and other cores referred to
- in the text (green). Location (1) includes cores SO82-2, SO82-5, LO09-23, LO09-21, SO82-7
- and SO82-4 described in Lackschewitz and Wallrabe-Adams (1997). Location (2) includes
- cores ENAM93-21 and ENAM93-20 and location (3) includes cores LINK16, LINK17,
- LINK15 and LINK04 described in Rasmussen et al. (2003), Wastegård et al. (2006) and
- 821 Wastegård and Rasmussen (2014).

822

Figure 2: Schematic representation of the improved marine tephra framework for the North Atlantic between 60-25 cal kyr BP. Ages and the stratigraphic relationship of tephra horizons between cores are approximate should be treated with caution, see text for details. The ages utilised are based on either existing age models for sequences or estimates based on stratigraphic positions. Heinrich Events 2-5 are included as stratigraphic markers and their ages are based on those given in Sanchez Goñi and Harrison (2010).

829

Figure 3: (a) Total alkali v silica plot focusing on (i) rhyolitic material and (ii) basaltic and

831 intermediate composition glass material from NAAZ II deposits in the marine network. (b)

832 Comparison of new characterisations of NAAZ II rhyolitic glass to characterisations from

- prior studies. Geochemical fields based on analyses of glass from deposits in cores V23-23,
- 834 V27-114, V23-82, V23-81 and V23-42 (Kvamme et al., 1989), MD95-2006 (Austin et al.,
- 835 2004), ENAM93-20, ENAM33 and EW9302-2JPC (Wastegård et al., 2006) and MD99-2289
- 836 (Brendryen et al., 2011). (c) Comparison of basaltic glass from newly characterised NAAZ II
- deposits to basaltic NAAZ II glass-based populations defined by Kvamme et al. (1989). All

838 geochemical data plotted on a normalised anhydrous basis.

- 839
- **Figure 4:** (a) Tephrostratigraphy of MD99-2251 between 1950-2030 cm covering the depth

interval of NAAZ II. (i) Rhyolitic glass shards in the 25-80 µm grain-size fraction. (ii)

- Basaltic glass shards in the 25-80 µm grain-size fraction. (b) Peak concentrations of
- 843 colourless (rhyolitic) and brown (basaltic) glass shards in tephra and cryptotephra deposits
- related to North Atlantic Ash Zone II. (c) Relative proportion of geochemical populations
- 845 within analyses of basaltic glass tephra shards from NAAZ II deposits at six sites within the
- 846 marine core network. Shard analyses not linked to the previously published populations or II-
- 847 INT-1 were classified as uncorrelated.

Figure 5: (a) High-resolution concentration profiles of brown glass shards between 910-920
cm in MD95-2010. (b) Comparison of the glass shard composition of MD95-2010 915-916
cm to the glass-based characterisation of FMAZ IV (JM11-19PC 542-543 cm) from Griggs et
al. (2014). All geochemical data plotted on a normalised anhydrous basis.

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Figure 6: (a) (i) Percentage abundance of *Neogloboquadrina pachyderma* (sinistral) and (ii) 854 brown glass shard tephrostratigraphy incorporating 5 cm and 1 cm counts for the MD04-2822 855 856 core. (b) (i) Percentage abundance of Neogloboquadrina pachyderma (sinistral) and (ii) 857 brown glass shard tephrostratigraphy incorporating 5 cm and 1 cm counts for the MD04-2829CQ core. Foram abundances and Dansgaard-Oeschger event numbering for MD04-2822 858 and MD04-2829CQ from Hibbert et al. (2010) and Hall et al. (2011) respectively. (c) 859 Geochemical characterisations of glass shards from Type 1 tephra deposits in the MD04-2822 860 861 and MD04-2829CQ cores. (i) inset of total alkali vs. silica plot. Division line to separate alkaline and sub-alkaline material from MacDonald and Katsura (1964). Chemical 862 classification and nomenclature after Le Maitre et al. (1989). (ii) FeO/TiO₂ vs. SiO₂ and (iii) 863 TiO₂ vs. Al₂O₃ compositional variations diagrams comparing the glass shard composition of 864 865 MD04-2822 and MD04-2829CQ deposits to characterisations of proximal Icelandic material. 866 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 867 for the Kverkfjöll volcano from Gudmundsdóttir et al. (2016). All geochemical data plotted 868 869 on a normalised anhydrous basis.

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871 Figure 7: (a) (i) Percentage abundance of *Neogloboquadrina pachyderma* (sinistral), (ii) ash free IRD concentration and (iii) tephrostratigraphic record of the MD99-2251 marine core. 872 Glass shard counts have been truncated for clarity. Shard counts in the 1686-1687 cm sample 873 (*) are 4991, 1862 and 507 shards per 0.5 g dws in the 25-80, 80-125 and >125 µm grain-size 874 875 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 876 877 which glass shards were subsequently extracted for compositional characterisation. (b) Composition of significant geochemical populations identified in glass analyses of tephra 878 deposits within the MD99-2251 core. (i) inset of total alkali vs. silica plot. Division line to 879 separate alkaline and sub-alkaline material from MacDonald and Katsura (1964). Chemical 880 classification and nomenclature after Le Maitre et al. (1989). (ii) FeO/TiO₂ vs. SiO₂ and (iii) 881

- 882 TiO₂ vs. Al₂O₃ compositional variations diagrams comparing significant glass shard based
- geochemical populations from the MD99-2251 deposits to characterisations of proximal
- 884 Icelandic material. 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.
- 887 (2016). All geochemical data plotted on a normalised anhydrous basis.
- 888
- **Figure 8:** (a) (i) Percentage abundance of *Neogloboquadrina pachyderma* (sinistral), (ii) percentage IRD (>150 μ m fraction) and (iii) tephrostratigraphic record of the GIK23415-9 marine core. *Np*(s) and IRD data from Vogelsang et al. (2004) and Weinelt (2004), respectively. Labels for Heinrich Events from Weinelt et al. (2003) and Lu et al. (2007).
- 893 Shard counts have been truncated for clarity. Shard counts in the 193-194 cm sample are
- 5131 and 280 shards per 0.5 g dws in the 25-80 and >125 μ m grain-size fractions
- respectively. Red bars denote samples depths from which glass shards were subsequently
- 896 extracted for compositional characterisation. (b) Composition of significant glass-based
- geochemical populations identified in tephra deposits within the GIK23415-9 core. (i) inset
- of total alkali vs. silica plot. Division line to separate alkaline and sub-alkaline material from
- 899 MacDonald and Katsura (1964). Chemical classification and nomenclature after Le Maitre et
- al. (1989). (ii) FeO/TiO₂ vs. SiO₂ and (iii) TiO₂ vs. Al₂O₃ compositional variations diagrams
- 901 comparing significant glass-based geochemical populations from the GIK23415-9 deposits to
- 902 characterisations of proximal Icelandic material. 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
- 905 Gudmundsdóttir et al. (2016). All geochemical data plotted on a normalised anhydrous basis.906
- Figure 9: (a) Comparison of the glass analyses of the MD04-2822 1836-1837 cm tephra
 horizon and the GIK23415-9 225-226 cm (TAB-2) geochemical population. (b) Comparison
 of the glass analyses of the FMAZ II tephra horizon (JM11-19PC 202-203 cm from Griggs et
 al. (2014)) and that of the GIK23415-9 202-203 cm (THOL-1) geochemical population. All
 geochemical data plotted on a normalised anhydrous basis.

912	Supplementary Information
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914	Supplementary Figures
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916	Figures S1-S13: Graphical analysis of geochemical populations identified within single-
917	shard major element glass analyses from tephra deposits within the MD99-2251 (S1-S7) and
918	GIK23415-9 (S8-S13) cores.
919	
920	Supplementary Data
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922	Table S1: Original major oxide concentrations of glass shards from deposits related to the
923	rhyolitic component of North Atlantic Ash Zone II (II-RHY-1). Deposits analysed are (i)
924	MD04-2822 2168-2169 cm (ii) MD95-2024 1445-1446 cm (iii) MD99-2251 1974-1975 cm
925	(supplementary peak) (iv) MD99-2251 2014-2015 cm (main peak) (v) M23485-1 622-623
926	cm (vi) GIK23415-9 429-430 cm (vii) MD01-2461 942-943 cm (supplementary peak) (viii)
927	MD01-2461 947-948 cm (main peak) (ix) MD04-2820CQ 610-611 cm (x) JM11-19PC 618-
928	623 cm (xi) MD95-2010 996-1000 cm.
929	
930	Table S2: Similarity coefficient comparisons of average concentrations of glass analyses of
931	the II-RHY-1 component in deposits from cores analysed within this work and by Kvamme et
932	al. (1989), Austin et al. (2004), Wastegård et al. (2006) and Brendryen et al. (2011).
933	
934	Table S3: Original major oxide concentrations of glass shards from basaltic and intermediate
935	shards directly associated with deposits of the rhyolitic component of North Atlantic Ash
936	Zone II (II-RHY-1). Deposits analysed are (i) MD99-2251 2014-2015 cm (ii) M23485-1 622-
937	623 cm (iii) GIK23415-9 429-430 cm (iv) MD01-2461 947-948 cm (v) MD04-2820CQ 610-
938	611 cm (vi) JM11-19PC 618-623 cm.
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940	Table S4: Original major oxide concentrations of glass shards from the MD95-2010 915-916
941	cm tephra deposit.
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943	Table S5: Original major oxide concentrations of glass shards from tephra deposits in the
944	MD04-2822 core. Deposits analysed are from the depths of (i) 1836-1837 cm (ii) 2004-2005
945	cm and (iii) 2017-2018 cm.

946 Table S6: Original major oxide concentrations of glass shards from tephra deposits in the 947 MD04-2829CQ core. Deposits analysed are from the depths of (i) 800-801 cm (ii) 930-931 948 949 cm and (iii) 934-935 cm. 950 951 **Table S7:** Original major oxide concentrations of glass shards from tephra deposits in the 952 MD04-2820CQ core. Deposits analysed are from the depths of (i) 487-488 cm (ii) 497-498 953 cm and (iii) 524-525 cm. 954 Table S8: Original major oxide concentrations of glass shards from tephra deposits in the 955 MD99-2251 core. Deposits analysed are from the depths of (i) 1654-1655 cm (ii) 1680-1681 956 cm (iii) 1683-1684 cm (iv) 1713-1714 cm (v) 1772-1773 cm (vi) 1796-1797 cm (vii) 1812-957 1813 cm and (viii) 1904-1905 cm. 958 959 Table S9: Analysis of glass-based geochemical populations present within tephra deposits 960 961 identified in the MD99-2251 marine core. n = total number of analyses from deposits. Veid.-Bárd. = Veidivötn-Bárdarbunga. 962 963 Table S10: Original major oxide concentrations of glass shards from tephra deposits in the 964 965 GIK23415-9 core. Deposits analysed are from the depths of (i) 173-174 cm (ii) 193-194 cm (iii) 202-203 cm (iv) 225-226 cm (v) 302-303 cm (vi) 305-306 cm and (vii) 375-376 cm. 966 967
Table S11: Analysis of glass-based geochemical populations present within tephra deposits
 968 identified in the GIK23415-9 marine core. n = total number of analyses from deposits. Veid.-969 970 Bárd. = Veidivötn-Bárdarbunga. 971
Table S12a: Original secondary standard analyses of the BCR2g standard made throughout
 972 973 analytical periods during which glass sample analyses presented in this work were analysed. 974 975 **Table S12b:** Original secondary standard analyses of the Lipari standard made throughout analytical periods during which glass sample analyses presented in this work were analysed. 976 977
Table S13: Similarity coefficient comparisons between the glass-based geochemical
 978 signatures of isochronous horizons and significant glass-based geochemical populations in 979

- the marine tephra framework for the North Atlantic between 25-60 ka BP. Method of
- Borchardt et al. (1972) utilised. Red text shows SC values between 0.97 and 0.999 grey text
- shows SC values less than 0.95.

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Table 1: Summary of isochronous horizons and significant geochemical populations forming the marine tephra framework for the North Atlantic between 25-60 ka BP. The designation of climatic events is based on pre-existing stratigraphic frameworks for the cores. The stratigraphic ordering of horizons between cores is approximate. FMAZ II, FMAZ IV and NAAZ II have been identified in multiple cores. H = Heinrich Event; DO = Dansgaard-Oeschger Event. bas = basaltic; rhy = rhyolitic. Vat. = Vatnafjöll; Veid.-Bárd. = Veidivötn-Bárdarbunga. Deposit types based on the classification scheme outlined in Abbott et al. (submitted). References are as follows: 1: this study; 2: Rasmussen et al. (2003); 3: Wastegård et al. (2006); 4: Davies et al. (2008); 5: Griggs et al. (2014); 6: Abbott et al. (2016); 7: Wastegård and Rasmussen (2014); 8: Kvamme et al. (1989); 9: Austin et al. (2004): 10: Brendryen et al. (2010).

Tephra horizon/deposit (pop.)	Climatic event	Composition	Volcanic source	Deposit type	Ref(s)
Isochronous horizons				Ĭ.	
GIK23415-9 173-174 cm (THOL-1)	Post H2	Tholeiitic bas	Kverkfjöll	1	1
FMAZ II	Post DO-3	Transitional alkali bas	Hekla/Vatnafjöll	2A	2, 3, 4, 5
MD04-2822 1836-1837 cm	Pre DO-4	Transitional alkali bas	Katla or Hekla/Vatnafjöll	1	1
GIK23415-9 225-226 cm (TAB-1)	H3	Transitional alkali bas	Katla or Hekla/Vat.	1	1
MD04-2829CQ 800-801 cm (THOL-1)	Pre DO-4	Tholeiitic bas	Grímsvötn	1	1
MD04-2829CQ 800-801 cm (THOL-2)	Pre DO-4	Tholeiitic bas	Kverkfjöll	1	1
MD99-2251 1680-1681 cm (TAB-1)	H3	Transitional alkali bas	Katla	2A	1
MD04-2822 2004-2005 cm	DO-8	Transitional alkali bas	Katla	1	1
MD04-2829CQ 930-931 cm	DO-8	Tholeiitic bas	Grímsvötn	1	1
MD04-2829CQ 934-935 cm	Pre DO-8	Tholeiitic bas	Grímsvötn	1	1
MD04-2820CQ 487-488 cm	Pre DO-8 (H4)	Tholeiitic bas	Grímsvötn	2A	6
MD04-2820CQ 497-498 cm	Pre DO-9	Transitional alkali rhy	Katla	2A	6
MD04-2822 2017-2018 cm	Pre DO-9	Tholeiitic bas	Grímsvötn	1	1
MD04-2820CQ 524-525 cm	Pre DO-11	Tholeiitic bas	Grímsvötn or Kverkfjöll	2A	6
FMAZ IV	Pre DO-12	Tholeiitic bas	Grímsvötn	2A	5,7
NAAZ II (II-RHY-1)	End of DO-15	Transitional alkali rhy	Tindfjallajökull	3	1, 3, 8, 9, 10
Significant geochemical populations	r.				
GIK23415-9 202-203 cm (TAB-1)	Pre H2	Transitional alkali bas	Katla	2B	1
GIK23415-9 202-203 cm (THOL-1)	Pre H2	Tholeiitic bas	Kverkfjöll	2B	1
GIK23415-9 202-203 cm (THOL-2)	Pre H2	Tholeiitic bas	Grímsvötn	2B	1
GIK23415-9 202-203 cm (THOL-3)	Pre H2	Tholeiitic bas	VeidBárd.	2B	1
MD99-2251 1713-1714 cm (TAB-1)	Pre H3	Transitional alkali bas	Katla	2B	1
MD99-2251 1713-1714 cm (THOL-1)	Pre H3	Tholeiitic bas	Grímsvötn	2B	1
MD99-2251 1772-1773 cm (TAB-1)	Post H4	Transitional alkali bas	Katla	2B	1
MD99-2251 1772-1773 cm (TAB-2)	Post H4	Transitional alkali bas	Katla (?)	2B	1
GIK23415-9 302-306 cm (THOL-1)	H4	Tholeiitic bas	Grímsvötn	4	1
GIK23415-9 302-306 cm (THOL-2)	H4	Tholeiitic bas	Grímsvötn (?)	4	1
MD99-2251 1812-1813 cm (THOL-1)	H4	Tholeiitic bas	Grímsvötn	2B	1
MD99-2251 1812-1813 cm (THOL-2)	H4	Tholeiitic bas	VeidBárd.	2B	1
MD99-2251 1812-1813 cm (THOL-3)	H4	Tholeiitic bas	VeidBárd.	2B	1
MD99-2251 1812-1813 cm (TAB-1)	H4	Transitional alkali bas	Katla	2B	1
MD99-2251 1904-1905 cm (ALK-1)	Post H5	Alkali bas	Vestmannaeyjar	2B	1
GIK23415-9 375-376 cm (THOL-1)	Pre H5	Tholeiitic bas	Grímsvötn	2B	1
NAAZ II (II-THOL-2)	End of DO-15	Tholeiitic bas	Grímsvötn	2A, 2B	8, 1
NAAZ II (II-TAB-1)	End of DO-15	Transitional alkali bas	Katla	2A, 2B	8, 1
NAAZ II (II-INT-1)	End of DO-15	Trachyandesite- Trachydacite	Unknown	2A, 2B	1





Uncorrelated isochronous horizons Significant geochemical populations









Deposit type

Type 1

Type 1 Type 1





 $(10\%) O_2 M + O_{26} N$

Figure 6



(b) MD99-2251 isochronous horizon and significant geochemical populations









(b) GIK23415-9 isochronous horizons and significant geochemical populations



