1	Unusual subpolar North Atlantic phytoplankton bloom in 2010: volcanic
2	fertilisation or North Atlantic Oscillation?
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15	Volcanic eruption and North Atlantic Oscillation considered as drivers
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### 26 Abstract

27 In summer and autumn 2010, a highly anomalous phytoplankton bloom, with 28 chlorophyll concentration more than double that of previous years, was observed in 29 the Irminger Basin, southwest of Iceland. Two unusual events occurred during 2010 30 which had the potential to promote the unusual bloom. Firstly, in spring 2010, the 31 Eyjafjallajökull volcano in Iceland erupted, depositing large quantities of tephra into 32 the subpolar North Atlantic. Secondly, during the winter of 2009/2010 the North 33 Atlantic Oscillation (NAO) became extremely negative, developing into the second 34 strongest negative NAO on record. Hydrographic conditions were highly anomalous 35 in the region, with an influx of freshwater spreading through the basin, and unusual 36 nutrient and mixed layer depth conditions. Here we use a combination of satellite, 37 modelled and *in situ* data to investigate whether the input of iron from the volcanic 38 eruption or change in hydrographic conditions due to the extreme negative NAO were 39 responsible for the anomalous phytoplankton bloom. We conclude that changes in 40 physical forcing driven by the NAO, and not the volcanic eruption, stimulated the 41 unusual bloom.

### 43 **1. Introduction**

44 In April/May 2010, the Eyjafjallajökull volcano in Iceland erupted, disrupting 45 air travel throughout Europe and producing an estimated 270 million cubic metres of 46 airborne tephra, of which roughly half fell in Iceland, with much of the remainder 47 deposited in the surface waters of the North Atlantic [Gudmundsson et al., 2012; 48 Karlsdottir et al., 2012]. Because volcanic ash contains iron, an important 49 micronutrient for phytoplankton growth, eruptions are postulated to have potentially 50 large effects on oceanic primary production (PP; Duggen et al., 2010). Marine 51 sediment records contain evidence of volcanic ash deposition events throughout 52 Earth's history, and results from field investigations suggest that these events may 53 have been responsible for large scale climatic changes [Robock, 2000]. The eruption 54 of Mt. Pinatubo in 1991, for example, coincided with a drawdown of CO<sub>2</sub> in the 55 Northern Hemisphere, hypothesized to be driven by increased PP stimulated by 56 natural iron fertilisation following ash deposition [Sarmiento, 1993]. Satellite-derived 57 ocean colour data has recently been used to investigate localised increases in PP 58 following volcanic eruptions in Mikakejima, Japan [Uematsu et al., 2004], Montserrat 59 [Duggen et al., 2007], Kasatochi, Alaska [Langmann et al., 2010] and Anatahan, 60 Mariana Islands [Lin et al., 2011].

Another unusual event with the potential to alter phytoplankton bloom dynamics in the subpolar North Atlantic also occurred in 2010. The NAO index has been mostly in a positive state since the early 1970s, with occasional negative years (Supplementary Figure S1). However, the winter of 2009/2010 had exceptional NAO conditions, being the second strongest negative NAO on record. Negative NAO conditions are typified by reduced westerly winds and a southward displacement of the North Atlantic storm track. The altered heat fluxes result in a tri-polar structure in

sea surface temperature, being warmer in subpolar and subtropical regions and cooler at mid-latitudes (e.g. *Visbeck at al.*, 2003). A rapid wind-driven response in surface currents follows interannual variations in the NAO. In negative NAO periods, the North Atlantic Current shifts eastwards and heat and freshwater transports into the northern North Atlantic increase with the expansion of the subtropical gyre [*Bersch*, 2002].

74 The role of the NAO in driving interannual variability in phytoplankton 75 populations has been explored in both the Continuous Plankton Recorder data [*Reid et* 76 al., 1998; Edwards et al., 2001] and in biogeochemical modelling studies [Henson et 77 al., 2009; Patara et al., 2011]. The sustained positive NAO period in the 1980's and 78 1990's resulted in reduced phytoplankton populations in both the eastern and western 79 North Atlantic [Edwards et al., 2001]. Correlations at the decadal scale between the 80 NAO and phytoplankton abundance are fairly weak [Barton et al., 2003], however 81 interannual variability in mixing driven by NAO-induced changes in heat and 82 freshwater fluxes are well correlated with phytoplankton bloom timing and magnitude 83 [Henson et al., 2009]. Negative NAO periods typically exhibit a decrease in 84 phytoplankton abundance in the subtropics and an earlier, enhanced bloom in 85 subpolar regions [Patara et al., 2011].

We observed a widespread, anomalously strong phytoplankton bloom in satellite ocean colour data throughout the Irminger Basin in the subpolar North Atlantic during spring and summer 2010. The Irminger Basin lies between Greenland and Iceland and is a site of occasional deep convection [*Bacon et al.*, 2003]. Phytoplankton dynamics in the basin are typical of the classic North Atlantic spring bloom (*sensu Sverdrup*, 1953), with deep mixed layers in winter ensuring that phytoplankton growth is light limited [*Henson et al.*, 2006a]. The spring bloom is

93 initiated by a shoaling mixed layer associated with a period of net heat flux into the 94 ocean [Waniek and Holliday, 2006; Henson et al., 2006a]. Typically, diatoms 95 dominate the initial stages of the bloom, becoming outcompeted by smaller flagellates 96 as silica is depleted [Henson et al., 2006b; Heath et al., 2008]. However, unlike most 97 regions of the subpolar North Atlantic, nitrate is not completely depleted during the growing season, with residual concentrations of ~ 2-4  $\mu$ mol l<sup>-1</sup> at the end of summer 98 99 [Henson et al., 2003]. This observation has led to the suggestion that phytoplankton 100 growth is iron limited in summer in the Irminger Basin [Sanders et al., 2005]. To 101 directly test this hypothesis, two cruises to the region in spring and summer 2010 were 102 planned as part of the Irminger Basin Iron Study (IBIS) programme. The eruption of 103 the Eyjafjallajökull volcano occurred coincidentally just prior to the first cruise 104 allowing the investigation of the influence of volcanic ash deposition on the 105 biogeochemistry of the region [Achterberg et al., 2013].

In addition to the two cruises in 2010, the Irminger Basin was also visited in spring and summer 2002, and so here we are able to compare the unusual conditions in 2010 to the more typical conditions observed in 2002. We use a combination of satellite data, *in situ* data and model output to investigate both unusual forcing events – the volcanic eruption and the extreme negative NAO – as possible mechanisms to explain the anomalous bloom.

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# 113 **2. Data and Methods**

MODIS Aqua Level-3 satellite-derived chlorophyll concentration (chl) data (R2012.0) at 8-day, 9 km resolution for July 2002-December 2010 were obtained from NASA (http://oceancolor.gsfc.nasa.gov/). Satellite-derived primary production PP estimates were made using the standard Vertically Generalised Productivity Model

118 [Behrenfeld and Falkowski, 1997]. Volcanic ash carried in the atmosphere can interfere with satellite ocean colour retrievals, resulting in a false increase in chl 119 120 [Claustre et al., 2002]. To investigate whether airborne ash may have interfered with 121 the satellite chl retrieval, the time series of MODIS Level-3 aerosol optical thickness 122 (AOT) data in the Irminger Basin was examined. This showed only two 8-day periods during which AOT was greater than the range found in previous years (1<sup>st</sup>-8<sup>th</sup> 123 May and 18<sup>th</sup>-25<sup>th</sup> June; Supplementary Figure S2). The first period occurs during the 124 125 active eruption phase of the volcano, but the elevated AOT does not coincide with an 126 anomalous increase in chlorophyll (Figure 1b). The second period of unusually high 127 AOT occurs more than one month after the eruption ended, and again does not 128 correspond with anomalously high chl. MODIS Level-3 processing by NASA has a 129 high level of quality control, with 27 flags including those for failed atmospheric 130 correction and failed chlorophyll retrieval. In this case, we conclude that the NASA 131 processing successfully excluded the majority of airborne ash-affected pixels from the 132 Level-3 chl product. In situ chl measurements from 2002 and 2010 in the central 133 Irminger Basin (Figure 1c) agree well with the satellite values, giving further 134 confidence that volcanic ash is not unduly impacting the satellite-derived chl data.

Monthly wind data for 1998-2010 were obtained from the NCEP/NCAR Reanalysis project [*Kalnay et al.*, 1996]. Monthly optimally interpolated fields of salinity and temperature for 1998-2010 were obtained from the Hadley Centre (EN3 dataset derived from a variety of data sources, including Argo floats and the World Ocean Database). Full details of the quality control and data processing can be found in *Ingleby and Huddleston* [2007]. Density was calculated from temperature and salinity fields using the International Equation of State for Seawater (IES80). Mixed layer depth was estimated from individual Argo float profiles collected in the central
Irminger Basin from 2001-2011 using the *Holte and Talley* [2009] density algorithm.

144 Two cruises to the Irminger Basin took place aboard the RRS Discovery during 145 (26 April-9 May 2010; D350) and after (4 July-11 August 2010; D354) the 146 Eyjafjallajökull eruption as part of the IBIS programme [Achterberg et al., 2013]. 147 Here we compare nutrient and chl concentrations measured on the 2010 cruises to 148 those measured during the UK Marine Productivity programme to the same region in 149 2002 [Holliday et al., 2006]. The 2002 cruises took place during similar periods as in 150 2010 (18 April-27 May 2002; D262 and 25 July-28 August 2002; D264). On all 151 cruises, macronutrients were determined using a Skalar San Plus autoanalyser 152 [Kirkwood, 1996] and samples for chl were filtered through 25 mm Whatman GF/F 153 filters, extracted in 90% acetone and determined fluorometrically. Surface chl 154 samples were taken from the ship's continuous thermosalinograph outflow from an 155 intake of 5 m depth. Nutrient data are reported as integrated euphotic zone values, 156 where euphotic depth was estimated from satellite chl using the method of Morel 157 [1988]. The temperature and salinity characteristics of the Irminger Basin in 2010 are 158 compared to data collected south of Greenland in 2008 (21 August-25 September 159 2008; D332; Bacon, 2010). A map showing all the cruise stations used in this study is 160 presented in Supplementary Figure S3. The 'central Irminger Basin' is defined here 161 as the region bounded by the 1500 m depth contour and north of  $58^{\circ}N$ , east of  $42^{\circ}W$ . 162 Results reported for the central Irminger Basin represent a spatial average of all 163 measurements made in this region

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165 **3. Results** 

## 166 **3.1 Chlorophyll concentration**

167 Satellite-derived chlorophyll images reveal the extent of the anomalous bloom in the Irminger Basin in summer 2010. In early-mid July, the central Irminger Basin 168 169 experienced unusually high chl concentrations, as revealed in the anomaly map in Figure 1a. The chl concentration in individual pixels was as much as 5 mg m<sup>-3</sup> greater 170 171 than the mean of previous years. Examination of the time series of satellite chl data 172 averaged over the central Irminger Basin (Figure 1b), shows that the unusual chl 173 signal in 2010 was a function of the anomalous timing of the bloom peak, in addition 174 to the extended period of high chl concentration, which continued throughout the 175 autumn. The exceptionally high chl concentration in summer 2010 is confirmed by 176 comparison of *in situ* chl samples with the conditions observed in 2002 (Figure 1c). Although spring chl was similar in both years (0.8-0.9 mg  $m^{-3}$ ), chl was significantly 177 higher in summer 2010 (2.2 mg m<sup>-3</sup>) than in 2002 (0.9 mg m<sup>-3</sup>). In 2003-2009 and 178 179 2011, the evolution of chl through the year follows the typical North Atlantic bloom 180 pattern with a rapid increase in chl in late April/early May, followed shortly after by maximum chl values of ~ 1 mg m<sup>-3</sup> in late May/early June. A reduction in chlorophyll 181 182 during summer is followed by a smaller autumn bloom in late August. In 2010, the 183 bloom progresses in an altogether different manner. The bloom starts as normal in 184 early May, but instead of rapidly reaching a peak before dying back in summer, the 185 chl concentration continues to rise, finally reaching its peak in mid-July, with elevated 186 concentration persisting through autumn 2010. Chlorophyll concentrations then 187 returned to normal levels in 2011. Clearly, the normal seasonal cycle of 188 phytoplankton growth was perturbed in the Irminger Basin in 2010, but was this due 189 to changes in physical forcing or a volcanic fertilisation effect?

190 **3.2 Hydrographic conditions** 

191 The NAO in winter 2009/2010 was in an extreme negative phase 192 (Supplementary Figure S1). During negative phases of the NAO, surface pressure 193 increases in the Icelandic low and decreases in the Azores high, resulting in weaker 194 westerly winds and a southward shift of the storm track [Visbeck et al., 2003]. In the 195 case of 2010, the wind during December 2009-June 2010 is either northward (in 196 January and March) or eastward (in February and May), i.e. in the opposite direction 197 entirely, compared to mean conditions. Wind speed is also exceptionally high in May 198 2010 (Figure 2).

199 The hydrographic conditions in the Irminger Basin in 2010 were also 200 anomalous. Transects of density anomaly along 33 °W (Figure 3) show a fresh 201 anomaly intruding into the Irminger Basin from ~ April onwards, inundating the 202 entire basin by August and persisting until December. The anomaly extends over 203 only the top 50-80 m of the water column, i.e. it likely originates from anomalous 204 surface, or near-surface, conditions. Theta-S plots from 2010 provide insight into the 205 likely source of the anomalous water mass in the central Irminger Basin (Figure 4). 206 The theta-S characteristics of the upper ocean in the central Irminger Basin in 2010 207 are very similar to data collected along a transect south of Cape Farewell in early 208 September 2008 [Bacon et al., 2010]. Although the 2010 surface waters are warmer 209 than in 2008, they occupy a theta-S space that is absent in the 2008 data; that is, a 210 mixing region between the typical central Irminger Basin surface layer (salinity of ~ 211 34.87-35.05, as in 2008) and the central gyre fresh pool (salinity of ~ 34.65-34.75). A 212 plot of isohalines in the region also demonstrates the anomalous extent of freshwater 213 in the Irminger Basin in 2010 (Figure 5). In March, the location of the isohaline 214 (salinity of 35.02) in 2010, relative to previous years, is unusually far east at 52-54 215 °N. By June 2010 the isohaline is further north and east than is typical at 58-60 °N, indicating a significant change in surface water properties, and by September 2010 the
isohaline is north and east of its usual position, indicating anomalously fresh water
has inundated a large portion of the Irminger Basin. The unusual hydrographic
conditions are also reflected in the mixed layer depth, which remained shallower than
normal throughout the autumn (Figure 6).

221 The freshwater intrusion appears to have also introduced anomalous 222 macronutrient conditions, particularly in the southern Irminger Basin. Comparison of 223 nutrient concentrations during the 2010 cruises to similarly timed spring and summer 224 cruises in 2002 suggest that nitrate concentration in spring was similar in the two 225 years, but the summer nitrate was lower in summer 2010 than 2002 (Figure 7). The 226 silica concentration was slightly lower in spring 2010 than in 2002, but similar during 227 the summer cruise (Figure 7). The ratio of N:Si consumed between the spring and 228 summer cruises was 1:1 in 2002, the expected value for a diatom dominated bloom 229 [Brzezinski, 1985], but was 3:1 in 2010.

230 **3.3 V** 

## 3.3 Volcanic dust deposition

231 In addition to the unusual hydrographic conditions observed in 2010, the Eviafjallajökull volcano in Iceland was actively erupting between 15<sup>th</sup> April and 23<sup>rd</sup> 232 233 May. The ash ejected from the volcano was approximately 7.6 % iron oxide by 234 weight [Achterberg et al., 2013] and so could potentially have added substantial 235 quantities of iron to the surface ocean. Estimates of the iron deposition over the 236 region were made by Achterberg et al. [2013] using an atmospheric Lagrangian 237 particle dispersion model for the spatial distribution of ash deposition [Stohl et al., 238 2011] and estimates of ash sinking rates and modelled iron dissolution. A map of iron 239 deposition from the volcanic eruption (Figure 8a) shows that a maximum of  $\sim 0.08$ nmol  $l^{-1}$  of iron were likely deposited in the very northern part of the Irminger Basin, 240

and ~  $0.005 \text{ nmol } l^{-1}$  in the central basin, over the course of the eruption. The time series of iron deposition in this region (Figure 8b) shows that the vast majority was deposited on just one day:  $14^{\text{th}}$  May 2010.

Chlorophyll concentrations and hydrographic conditions returned to normal in 245 2011, and the NAO index, although still negative, was within the range of previous 246 winter conditions (Figure S1). This implies that the anomalous conditions of 2010 247 resulted from a one-off perturbation to the system, rather than a prolonged regime 248 shift, and that winter mixing in 2010/11 'reset' the system for the following year's 249 spring bloom.

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### 251 **4. Discussion**

Satellite-derived chl from the Irminger Basin typically depicts a short, intense bloom in spring, followed by a summer minimum [*Henson et al.*, 2006a]. However, 254 2010 exhibited highly anomalous conditions, with an extended period of high chl 255 persisting from early May through October, with a peak in mid-July. Total annually 256 integrated primary production in the Irminger Basin, estimated from satellite-derived 257 data was ~ 50% greater in 2010 than the mean of 2003-2009 and 2011, implying 258 potentially significant impacts on  $CO_2$  drawdown and carbon export.

The atmospheric conditions in winter 2009/2010 reflect the 'Warm Arctic-Cold Continents' climate pattern, which has occurred only 3 times in the last 160 years [*Overland et al.*, 2010]. The particularly cold winter in northern Europe and the eastern USA in 2009/2010 was in contrast to exceptionally warm Arctic temperatures, e.g. in Baffin Bay, northeastern Canada, where winter air temperature was ~ 8 °C warmer than usual [*Overland et al.*, 2010]. These conditions resulted in increased ice melt over much of the Arctic [*Stroeve et al.*, 2011]. As indicated by the unusual

266 progression of isohalines (Figure 5) and the theta-S characteristics of waters in the 267 southern central Irminger Basin in 2010 (Figure 4), this upper ocean freshwater signal 268 is likely to have originated from south of Cape Farewell where a pool of relatively 269 fresh water typically resides [Reverdin et al., 2002]. The region of the freshwater 270 pool is strongly influenced by the Labrador Current that flows south-eastward, 271 transporting very fresh water from Davis Strait to Newfoundland. Data from 272 Lagrangian drifters has demonstrated that the Labrador and North Atlantic currents 273 intensified under the negative NAO conditions of 1996, although the recirculation in 274 the Irminger Basin weakened [Flatau et al., 2003]. In addition, an increased 275 northward transport of heat and freshwater into the Irminger Basin along the western 276 side of the Mid-Atlantic Ridge was observed [Bersch, 2002]. In 2010, the extreme 277 negative NAO may have resulted in intensification of the Labrador Current and hence 278 freshwater influx to the central subpolar gyre. Anomalous wind conditions in 2010 279 may have then driven the fresh surface waters northeastward, overcapping the central 280 Irminger Basin.

The freshwater intrusion may also have altered nutrient conditions in the 281 region, as Labrador Current water has an excess of both phosphate and silicate 282 283 relative to nitrate [Harrison and Li, 2007] and transports Arctic species of plankton 284 south to the Newfoundland shelf [Head and Pepin, 2010]. The unusual nutrient 285 drawdown ratios shown in Figure 7 arise partly from the lower euphotic zone silica 286 concentrations in spring 2010, relative to nitrate and phosphate, which seems counter 287 to a Labrador Current origin for the freshwater anomaly. Historical macronutrient 288 data extracted from the World Ocean Database for 1980-2004 suggests that silica 289 concentrations in spring 2010 were indeed lower than the mean, suggesting that the 290 diatom bloom had already begun prior to the spring cruise. Silica concentrations in

291 the central Irminger Basin were, in general, around average in summer 2010 292 (Supplementary Figure S4), except along the southern transect (Figure 7f). The 293 anomalously high silica concentrations in the southern Irminger Basin may be a 294 remnant signature of the enhanced nutrient concentrations introduced to the basin with 295 the freshwater anomaly. The anomalous nutrient supply conditions are also reflected 296 in the unusual spring to summer N:Si drawdown ratio, which was 3:1 in 2010 rather 297 than the typical 1:1, suggesting that either the seasonal consumption by phytoplankton 298 of silica relative to nitrate was reduced in 2010 relative to 2002, or that an additional 299 input of silica occurred between May and August, perhaps associated with the 300 freshwater anomaly. This latter hypothesis is consistent with our observation that the 301 spring cruise occurred before the freshwater had fully inundated the basin (Figures 3 302 and 5), but by the time of the summer cruise, the freshwater anomaly had arrived at  $\sim$ 303 60 °N coinciding with higher than normal silicate concentrations in the southern 304 central Irminger Basin (Figures 5 and 7).

305 A change in the N:Si drawdown ratio may also imply a change in phytoplankton community structure in 2010, from the typical diatom dominated 306 307 population expected in spring in the Irminger Basin [Sanders et al., 2005] to a 308 phytoplankton population that consumed nitrate in greater quantities than silica. This 309 observation would be consistent with the canonical North Atlantic seasonal 310 succession, where diatoms are succeeded by other phytoplankton functional types as 311 silica becomes depleted. However, silica concentrations were greater than 2 mmol m<sup>-</sup> <sup>3</sup> in most of the Irminger Basin during the summer cruise, i.e. above the level where 312 313 diatoms may be out-competed by other species (Brown et al., 2003). Nevertheless, 314 the N:Si drawdown ratio was not 1:1 as expected for diatoms, suggesting that the 315 summer phytoplankton constituted a mixed community. A full taxonomic analysis of the phytoplankton community structure for the 2010 cruises is ongoing, but initial observations confirm the presence of an unusually large proportion of *Coccolithus pelagicus* (A. Poulton, pers. comm.), a species abundant in the Labrador Sea [*Ziveri et al.*, 2004]. *C. pelagicus* may thus have been advected into the Irminger Basin with the freshwater anomaly and contributed to the unusual nutrient drawdown ratios in 2010.

321 Either the influence of the warmer than usual air temperatures in the region, 322 and/or the freshwater intrusion resulted in an unusually shallow mixed layer depth in 323 autumn 2010. This is likely to have extended the phytoplankton growing season (as 324 seen in the satellite-derived chl data; Figure 1b), which is typically light limited in late 325 autumn and winter [Henson et al., 2006a]. Even if the light conditions were suitable 326 for phytoplankton growth for an extended period of time in 2010, this is unlikely to be 327 the sole reason for the anomalous bloom, as sufficient nutrients would also be 328 required. Although an additional source of nutrients may have been introduced to the 329 region by the unusual hydrographic conditions, it seems unlikely, if not impossible, 330 that this alone could have fuelled the exceptional increase in PP (satellite-derived 331 estimates suggest annually integrated PP was ~ 50% greater in 2010 than the mean). 332 Any additional nitrate introduced to the region would likely need to be supplemented 333 by increased rates of nutrient recycling to sustain these high rates of PP.

In situ measurements of ammonium concentration within the mixed layer revealed a 500-fold increase between the spring and summer cruises, confirming that intense recycling was occurring during the summer bloom (E. Achterberg, unpublished data). We have no other ammonium data from this region, and so cannot establish whether the concentrations were unusually high compared to other more typical years. However, atypically for a high latitude, open ocean region, very high ammonium concentrations (up to 2  $\mu$ mol l<sup>-1</sup>) were observed immediately below the

mixed layer in summer. The freshwater cap present in the Irminger Basin in 2010 may have allowed retention of the ammonium within the euphotic zone, potentially providing a significant quantity of nitrogen to support the high productivity observed throughout the summer. Our results suggest that the relatively warm spring/summer of 2010, combined with the shallow mixed layer, may have favoured increased rates of nitrogen recycling relative to other years.

347 The anomalous bloom may also have been influenced by reduced grazing 348 pressure. The abundance and behaviour of mesozooplankton in the central Irminger 349 Basin in summer 2010 was comparable to other years, based on measurements of 350 individual body size, gut clearance rates and weight-specific ingestion (S. Giering, 351 unpublished data) However, zooplankton were only grazing ~ 0.5-5 % of PP per day, 352 i.e. top down control of the bloom was very weak. Zooplankton seem to have been 353 unable to increase their grazing rate to respond to the anomalous bloom in 2010, so 354 that, in contrast to normal conditions, PP remained high throughout summer and into 355 autumn. Analysis of a decadal time series of copepods from the Continuous Plankton 356 Recorder survey, suggested that the abundance and population dynamics of Calanus 357 finmarchicus are insensitive to climate variability in this region [Heath et al., 2008]. 358 Our results suggest that the mesozooplankton population was incapable of altering its 359 behaviour in response to variability in conditions, so that grazing rates did not 360 increase despite anomalously high phytoplankton abundance. Although the bloom 361 may not have been under top-down control, maintaining the high phytoplankton 362 biomass through summer and autumn would still have required sufficient additional 363 nutrients.

364 An alternative explanation for the anomalous bloom evolution in 2010 lies 365 with the eruption of the Eyjafjallajökull volcano. Ash deposited onto the sea surface

366 during an eruption will leach iron and other metals into the upper ocean. However, 367 for any additional deposited iron to have a fertilisation effect, phytoplankton growth 368 must be iron limited. Some evidence for iron limitation in this region comes from 369 iron addition experiments conducted in summer 2007 in the Iceland Basin and in 370 summer 2010 in the Irminger Basin, which both demonstrated an increase in chl 371 compared to controls [Nielsdottir et al., 2009; Ryan-Keogh et al., 2013]. Indirect 372 evidence suggesting potential iron limitation in the Irminger Basin also came from the 373 Marine Productivity cruises in 2002. Typically, one would expect that after silica 374 becomes depleted, diatoms would become out-competed by other phytoplankton 375 functional types, which would consume the remaining nitrate. However, Sanders et 376 al. (2005) established that, although silica is depleted in surface waters in summer, 377 non-limiting concentrations of nitrate remain at the end of the growing season, 378 suggesting that iron limitation of phytoplankton growth may inhibit complete nitrate 379 drawdown in the Iceland, and possibly Irminger, Basins.

Results from an atmospheric transport model, combined with estimates of iron content and dissolution rates (see *Achterberg et al.*, 2013 for details), suggest that ash from the eruption may have contributed an additional ~ 0.005 nmol  $1^{-1}$  of dissolved iron to the central Irminger Basin, the majority of it in one day,  $14^{th}$  May (Figure 8). Elevated chl concentration (~ 0.3 mg m<sup>-3</sup> higher than the mean) is evident in the Irminger Basin in anomaly maps for the 9<sup>th</sup>-16<sup>th</sup> May period (Supplementary Figure S5), but has receded by the following week, when chl is lower than the mean.

The relatively transient nature of any stimulation of phytoplankton growth by ash-borne iron was confirmed by shipboard experiments conducted with ash from the Eyjafjallajökull eruption. Bioassays conducted during the summer 2010 cruise found that addition of 9 mg  $1^{-1}$  of ash  $\notin$  4.6 -16 nM DFe) to Iceland Basin populations

391 induced an increase in phytoplankton biomass, although the response was weaker than 392 to addition of 2 nM FeCl<sub>3</sub>, i.e. much of the dissolved iron released from the ash in 393 seawater was not bioavailable [Achterberg et al., 2013]. The ash also sank rapidly, 394 with settling experiments suggesting that the particles fell through the upper mixed 395 layer (~ 30 m deep in spring) in ~ 90 minutes, during which time all the salts leached 396 from the particles [Achterberg et al., 2013]. This is even faster than the previously 397 reported residence time of atmospheric iron inputs in the surface ocean of 2-7 weeks 398 [Sarthou et al., 2003]. These results are consistent with the observation during the 399 summer 2010 (i.e. post-eruption) cruise of a lack of elevated surface iron 400 concentrations in the region offshore of Evjafjallajökull [Achterberg et al., 2013]. 401 Taken together these observations imply that any fertilisation effect due to ash 402 deposition was short-lived.

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## 404 **5. Conclusion**

A highly anomalous chlorophyll bloom occurred in the central Irminger Basin in 2010, persisting throughout the summer and autumn. We present here several observations of unusual conditions in 2010 that may have influenced the development of the anomalous phytoplankton bloom:

The North Atlantic Oscillation was in an extreme negative phase in winter
2009/10; the second most negative on record since winter 1968/69 (Supplementary
Figure S1).

Wind direction was reversed in spring 2010 compared to normal conditions,
coming from the south or east (Figure 2).

A surface freshwater anomaly was observed extending throughout the central
Irminger Basin in summer 2010 to a depth of ~ 30 m (Figure 3).

The freshwater anomaly had characteristics similar to that of the central gyre
fresh pool, situated south of Cape Farewell (Figure 4), and migrated northeastwards
into the Irminger Basin during summer 2010 (Figure 5).

419 • The mixed layer shoaled earlier than usual and stayed relatively shallow
420 through autumn (Figure 6).

The central Irminger Basin phytoplankton community composition had an
unusually large proportion of *Coccolithicus pelagicus*, usually abundant in the
Labrador Sea.

Nutrient utilisation was anomalous, with N:Si drawdown ratios of 3:1, rather
than the usual 1:1 (Figure 7).

426 • Zooplankton grazing rates did not increase in tandem with PP during the
427 summer.

• The Eyjafjallajökull volcano erupted in April/May 2010 depositing ash in the northern Irminger Basin, which may have added a total of ~ 0.005-0.08 nmol l<sup>-1</sup> of iron to the region (Figure 8).

Although we can provide no definitive answer to what caused the anomalous bloom,
the balance of evidence points to the unusual hydrographic conditions of 2010, with
the volcanic eruption playing a relatively minor role.

Ongoing analysis of the cruise data collected as part of the IBIS programme may reveal some additional clues to the causes, and importantly, consequences of the anomalous bloom. A key question is whether the bloom resulted in an increase in organic carbon export, and therefore may have had a significant impact on the biogeochemistry of the region. Introduction of iron to the surface ocean via deposition of volcanic ash has been postulated as a driver of reductions in atmospheric CO<sub>2</sub> [*Watson et al.*, 1997], due to the stimulation of additional carbon export. In the

441 case of the Eyjafjallajökull eruption and the Irminger Basin, the volcanic eruption 442 seems to have had a fairly minor effect. A volcanic eruption would need to produce 443 substantially more ash, over a longer period, with direct deposition over a high 444 nutrient-low chlorophyll area in order to affect atmospheric CO<sub>2</sub> concentrations.

445

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**Figure 1:** Anomalous chlorophyll conditions in the Irminger Basin in 2010. a) Chlorophyll anomaly (mg m<sup>-3</sup>) for  $12^{\text{th}}$ - $19^{\text{th}}$  July 2010, relative to the mean of the same period in 2003-2009 and 2011. The region denoted in this study as the 'central Irminger Basin' is outlined in black. The position of the transect plotted in Figure 3 is marked with a vertical black line. b) Satellite-derived time series of chlorophyll concentration in the central Irminger Basin in 2010 (thick black line), the mean (thin grey line) and standard deviation (grey shading) of years 2003-2009 and 2011. Black horizontal line marks the period of the volcanic eruption. c) *In situ* measured chlorophyll concentration (collected from the underway system at ~ 5 m depth) in the central Irminger Basin collected on cruises in spring and summer 2002 (grey bars) and 2010 (white bars). Error bars denote the spatial variability of data within the basin (standard deviation).



**Figure 2:** Left column: Mean (1998-2009) wind speed over the North Atlantic in selected months (at 10 m above sea level). Right column: Wind speed in selected months of 2010. Note that the wind direction in February 2010 is reversed with respect to the mean, and that northward wind speeds in May 2010 are anomalously strong.



**Figure 3:** Density anomaly (kg m<sup>-3</sup>) from the Hadley Centre EN3 dataset for selected months in 2010, relative to the mean of 1998-2009, along a transect at 33 °W. Note the presence of an anomalously light, fresh water mass entering the Irminger Basin during May, capping the entire region by August and persisting until October.



**Figure 4:** Theta-S plot showing data from early September 2008 along a transect south of Cape Farewell (blue lines) and in the southern Irminger Basin along 59 °N (green lines). Also plotted are data from August 2010 in the central (black dots) and southern (55-56 °N) Irminger Basin (red dots).



**Figure 5:** Position of isohalines (salinity = 35.02) in March, June and September for years 2002-2011. The position of the isohaline in 2010 is marked with a thick red line. Note that in 2010 the appearance of the freshwater anomaly perturbs the isohaline to the northeast, suggesting the water mass originated to the southwest. Dashed grey arrow in last panel is a sketch of the hypothesised path of the freshwater intrusion.



**Figure 6:** Monthly mixed layer depth in the central Irminger Basin in 2010 (thick black line), the mean (thin grey line) and standard deviation (grey shading) of years 2001-2009 and 2011. Note that the mixed layer in 2010 is shallower than the mean in winter and throughout autumn. Also plotted are chlorophyll concentration in 2010 (black dashed line) and the mean of 2003-2009 and 2011 (grey dashed line) taken from Figure 1b.



**Figure 7**: Comparison between integrated euphotic depth macronutrients in the central Irminger Basin in a) 2002 and b) 2010 from cruises in spring (gray bars) and summer (white bars). Error bars denote the spatial variability of data within the basin (standard deviation). c) Nitrate and d) silicate concentrations (mmol m<sup>-3</sup>) collected underway at ~ 5 m depth on cruises in spring 2002 (small dots) and 2010 (diamonds), and similarly in summer for e) nitrate and f) silicate.



**Figure 8**: a) Modelled dissolved iron deposition (nmol  $l^{-1}$ ) from the eruption of the Eyjafjallajökull volcano (April 15-May 23, 2010). For details of the model and estimation of iron deposition see Stohl et al. (2011) and Achterberg et al. (2013). b) Daily modelled iron deposition rates (nmol  $l^{-1}$  day<sup>-1</sup>) in the central Irminger Basin during the volcanic eruption.