High variability of particulate organic carbon export along the North Atlantic GEOTRACES section GA01 as deduced from

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3 ²³⁴Th fluxes

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17 Abstract. In this study we report particulate organic carbon (POC) export fluxes for different biogeochemical

18 basins of the North Atlantic as part of the GEOTRACES GA01 expedition (GEOVIDE, May-June 2014). Surface

19 POC export fluxes were deduced by combining export fluxes of total ²³⁴Th with the POC to ²³⁴Th ratio of sinking

20 particles at the depth of export. Particles were collected in two size classes (> 53 μ m and 1-53 μ m) using *in-situ*

21 pumps and the large size fraction was considered as representative of sinking material. Surface POC export fluxes

22 revealed latitudinal variations between provinces ranging from 1.4 mmol $m^{-2} d^{-1}$ in the Irminger basin where the

23 bloom was close to its maximum, to 12 mmol m⁻² d⁻¹ near the Iberian Margin where the bloom had already

24 declined. In addition to the state of progress of the bloom, variations of the POC export fluxes were also related to

25 the phytoplankton size and community structure. In line with previous studies, the presence of coccolithophorids

26 and diatoms appeared to enhance the POC export flux, while dominance of pico-phytoplankton cells, such as

27 cyanobacteria, resulted in lower fluxes. The POC export to primary production (PP) ratio strongly varied regionally

and was generally low (≤ 14 %), except at two stations located near the Iberian margin (35%) and within the

29 Labrador basin (38%), which were characterized by unusual low *in-situ* PP. We thus conclude that the North

30 Atlantic during the GEOVIDE cruise was not as efficient in exporting carbon from the surface, as reported earlier

31 by others. Finally, we also estimated the POC export at 100 m below the surface export depth to investigate the

32 POC transfer efficiencies. This parameter was also highly variable amongst regions, with the highest transfer

33 efficiency at sites where coccolithophorids dominated.

34 1. Introduction

35 Through the sinking of particulate biogenic material, the biological carbon pump (BCP) plays a major role on the

36 sequestration of carbon-rich particles in the ocean interior. The North Atlantic harbors one of the most productive

37 spring phytoplankton bloom of the world's ocean (Esaias et al., 1986; Longhurst, 2010), generating an important

38 pulse of biogenic sinking particles (Buesseler et al., 1992; Honjo and Manganini, 1993; Le Moigne et al., 2013a),

- 39 which accounts up to 18% of the global BCP (Sanders et al., 2014). Yet, a substantial range of carbon export
- 40 efficiencies (1-47%) has been reported by earlier studies at different locations of the North Atlantic (Buesseler et
- 41 al., 1992; Buesseler and Boyd, 2009; Ceballos-romero et al., 2016; Herndl and Reinthaler, 2013; Lampitt et al.,
- 42 2008; Moran et al., 2003; Mouw et al., 2016; Thomalla et al., 2008), directly questioning about how carbon export
- 43 efficiency varies at a trans-Atlantic scale and what are the controlling factors.
- 44 The international GEOTRACES program aims to measure trace elements and isotopes along full-depth ocean
- 45 sections through each of the major ocean basins in order to provide maximum scientific rewards on a global scale 46
- 47 R/V Pourquoi Pas?), was a French contribution to this global survey. The studied area crossed five basins

(GEOTRACES, 2006). The GEOVIDE GA01 section in the high-latitude North Atlantic (15 May - 30 June 2014;

- 48 differentiated by their distinct biogeochemical and hydrodynamic characteristics: the Iberian basin, the west 49 European basin, the Icelandic basin, the Irminger basin and the Labrador basin (Fig.1).
- 50 The low nutrient availabilities (surface nitrate and silicate concentrations $< 1 \mu$ mol L⁻¹; nutrient analyses according
- 51 to Aminot and Kérouel, 2007) in the Iberian basin limits the biomass development giving the opportunity to pico-
- 52 phytoplankton, such as cyanobacteria, to grow (~ 35% of the total Chl-a at Station 13; Tonnard et al., in prep.;
- 53 pigment analyses according to Ras et al., 2008), a situation which is typical for the North Atlantic subtropical gyre
- 54 (Moore et al., 2008; Zehr and Ward, 2002). The Iberian basin can also be influenced by a local upwelling, close
- 55 to the Iberian margin (Costa Goela et al., 2016; Zúñiga et al., 2016; http://marine.copernicus.eu/) and potentially
- 56 fueling the area with nutrient-rich, but upwelling was not active during GEOVIDE (Shelley et al., 2016).
- 57 In the subpolar region, in the Irminger and Labrador basins, phytoplankton growth is strongly light-limited
- 58 seasonally (Riley, 1957) and the key parameter for alleviating these limitations is the progressive shoaling of the
- 59 mixed layer. There, micro-phytoplankton, such as diatoms, dominate the phytoplankton bloom (\geq 50% of the total
- 60 Chl-a; Tonnard et al., in prep.). Both basins are influenced by strong hydrodynamic features, such as the Irminger
- 61 gyre, the Eastern Greenland Current (EGC), the Western Greenland Current (WGC), the Labrador Current (LC;
- 62 Zunino et al., 2017) and the subduction of the Labrador Seawater (LSW) which was particularly intense (1700 m-
- 63 deep convection) during the winter 2013-2014 (Kieke and Yashayaev, 2015).
- 64 Between the subtropical and subpolar regions, the west European and Icelandic basins represent a transition zone
- 65 where nutrients and/or light can limit primary production (Henson et al., 2009). During GEOVIDE, the silicic acid
- 66 stock was low ($\leq 1 \mu$ mol L⁻¹) leading to the growth of nano-phytoplankton, such as haptophytes including
- 67 coccolithophorids (between 45 and 80% of the total Chl-a; Tonnard et al., in prep.). This region is influenced by
- 68 the Eastern Reykjanes Ridge Current (ERRC) and by the North Atlantic Current (NAC) with the southernmost
- 69 sub-branch evolving in a cyclonic eddy and the sub-arctic front (SAF). SAF separates cold and fresh waters from
- 70 the subpolar region and the warm and salty waters from the subtropical region (Zunino et al., 2017).
- 71 The North Atlantic is thus a heterogeneous basin in terms of nutrient status, phytoplankton communities and 72 hydrodynamic features.
- 73 This is of a crucial importance as ecosystem structure is thought to play an important role on the BCP. Guidi et al.
- 74 (2009) suggested that phytoplankton composition explained 68% of the variance in POC flux at 400 m. High
- 75 export efficiencies are reported in productive regions where diatoms dominate, but the exported material is
- 76 relatively labile and prone to remineralisation leading to low transfer efficiency and low deep export flux (Guidi
- 77 et al., 2009). Conversely, in oligotrophic regions, where diatoms are largely absent, primary production is low and
- 78 mostly regenerated. Consequently, export efficiencies are low but the eventual exported material is likely less

prone to dissolution - remineralisation, resulting in high transfer efficiencies (Henson et al., 2012; Lam et al., 2011;

80 Lima et al., 2014; Marsay et al., 2015). Phytoplankton size structure has also been shown to be an important factor

- 81 in controlling the POC export fluxes. Guidi et al. (2015) highlighted that the exported POC was more refractory
- 82 and the remineralisation depth was deeper when the fraction of micro-phytoplankton decreased or the fraction of
- 83 pico-phytoplankton increased.
- 84 Due to the complex impact of these biogeochemical factors on the POC export and according to the distinct
- 85 features of each biogeochemical basin, the efficiency of the North Atlantic to transfer POC to the deep ocean
- deserves more study.
- 87 In this context, we investigated POC export fluxes derived from the Thorium-234 (²³⁴Th) approach along a transect
- 88 in the high-latitude North Atlantic, from the Iberian margin to the sub-arctic Irminger and Labrador Seas. ²³⁴Th, a
- highly particle reactive element with a short half-life (24.1 d), is widely used to explore particle export over short
- time events such as phytoplankton blooms (Bhat et al., 1969; Buesseler et al., 1992; Coale and Bruland, 1985;
- 91 Cochran and Masqué, 2003). A deficit of 234 Th with respect to its radioactive parent 238 U (conservative in seawater)
- 92 is usually observed in the upper water column where particles sink. In the subsurface waters any excess of 234 Th 93 relative to 238 U, is taken to reflect particle break-up and remineralisation by heterotrophic bacteria and/or
- 24 zooplankton (Buesseler et al., 2008; Maiti et al., 2010; Savoye et al., 2004). A ²³⁴Th flux can be converted into a
- 95 POC flux by using the POC: 234 Th ratio of sinking particles at the depth of export (Buesseler et al., 2006).
- 96 In this study, we discuss carbon export fluxes determined at the base of the deficit zone according to the
- 97 biogeochemical properties found in each basins, with special emphasis on the stage and intensity of the bloom as
- 98 well as on the phytoplankton community structure. Using estimates of primary production from shipboard
- 99 incubations and satellite-derived Chl-*a*, we explore surface export efficiencies at different time scales over the
- studied area. In addition and using deep carbon export, we investigate POC transfer efficiency in the upper
- 101 mesopelagic.

102 2. Methods

103 **2.1.** Total ²³⁴Th and ²³⁸U

104 Total ²³⁴Th activities were determined from 4 L unfiltered seawater samples collected with 12L Niskin bottles. 105 Usually, 17 or 18 depths were sampled between the surface and 1000-1500 m, except at Stations 26 and 77 where 106 only 9 and 15 depths were sampled, respectively (Table S1). Deep samples (between 1000 and 3500 m) were taken 107 for the calibration of the low level beta counting (Rutgers van der Loeff et al., 2006) based on the knowledge that ²³⁴Th and ²³⁸U are generally in secular equilibrium at such depths (in this study, the deep ocean average ²³⁴Th/²³⁸U 108 109 ratio = 1.00 ± 0.02 ; n=15). Seawater samples were processed following the method developed by Pike et al. (2005). 110 Samples were acidified at pH 2 and spiked with a ²³⁰Th yield monitor in order to estimate the ²³⁴Th recovery during 111 the sample processing. After 12 hours of equilibration, pH was increased to 8.5 and KMnO₄ and MnCl₂ (analytical 112 grade, Merck) were added to form a manganese oxide precipitate. After a further 12 hours of equilibration, samples 113 were filtered on quartz-microfiber discs (QMA, Sartorius, 1 µm nominal porosity, 25 mm diameter). On board, 114 filters were dried overnight, mounted on nylon holders, and covered with Mylar and aluminum foil. The activity 115 of ²³⁴Th on each sample was counted using low level beta counters (RISØ, Denmark). Beta activity counting was 116 continued until a relative standard deviation (RSD) $\leq 2\%$ was reached. At the home-laboratory, residual beta

- activity was measured for each sample after a delay of six ²³⁴Th half-lives (~ 6 months) and these residual counts 117
- 118 were subtracted from the gross counts obtained on-board. All samples were then processed for Th recovery using
- ²²⁹Th as a second yield tracer. To do so, filters were dismounted from the nylon holders and transferred to clean 119 30 mL teflon vials (Savillex). All samples were spiked with ²²⁹Th, dissolved in a mix of 8M HNO₃/1M H₂O₂
- 120 121 (suprapur grade, Merck), heated overnight and filtered through Acrodisc® syringe filters (Pall, Nylon membrane,
- 122 nominal porosity=0.2 µm, diameter=25 mm). Part of the filtrate was pre-concentrated by evaporation and the
- 123 residue diluted in 1.4 M HNO₃ (suprapur grade, Merck). ²³⁰Th and ²²⁹Th concentrations were measured by sector
- field inductively coupled plasma mass spectrometry (SF-ICP-MS, Element 2, Thermo Scientific) in low resolution 124
- 125 mode. Each sample was analyzed 3 times and the precision of the ²³⁰Th:²²⁹Th ratios averaged 1.2% (RSD), which
- is within the range indicated by Pike et al. (2005). The total ²³⁴Th recovery, involving all the steps described above, 126
- was 91 \pm 14% (n=200). Uncertainty of total ²³⁴Th activity, estimated from error propagation, was between 0.04 127
- 128 and $0.10 \text{ dpm } \text{L}^{-1}$.
- 129 The ²³⁸U activity was deduced from salinity using the Eq. 1, given by Owens et al. (2011):
- $^{238}U = 0.0786 \times S 0.315$ 130 (1)

131 where ²³⁸U is the ²³⁸U activity in dpm L⁻¹ and S is salinity.

132

2.2. Particulate ²³⁴Th and POC sampling and analysis

133 Suspended particles were collected using *in-situ* large-volume filtration (100-1600 L) systems (Challenger 134 Oceanics and McLane pumps; ISP hereafter for "in-situ pumps") through paired 142 mm-diameter filters: a 53 µm 135 mesh nylon screen (SEFAR-PETEX®; polyester) and a 1 µm pore size quartz-microfiber filter (QMA, Sartorius), 136 respectively. The small size fraction (1-53 μ m) is referred to hereafter as SSF and the large size fraction (> 53 μ m) 137 as LSF. Prior to the cruise, filters were cleaned as follows: PETEX screens were soaked in 0.6 M HCl (Normapur, 138 Merck), rinsed with Milli-Q water, dried at ambient temperature in a laminar flow hood, and stored in clean plastic 139 bags; QMA filters were pre-combusted at 450 °C for 4 h and stored in aluminum foils until use. ISP were deployed 140 between 15 and 800 m on a stainless steel cable and the pumping time was approximatively 2-3 h (Table S2).

- 141 After collection, filters were processed on board. The 142 mm PETEX screen was cut into quarters using a clean
- 142 scalpel and two quarters were processed in this study. Particles were rinsed-off from the PETEX screen using 0.45 143 µm filtered seawater under a laminar flow hood. For one quarter of the PETEX screen, the rinsed-off particles
- 144 were re-filtered on a silver filter (SterliTech, porosity=0.45 µm, diameter=25 mm) and for another quarter on a
- 145 GF/F filter (Whatman®, porosity=0.7 µm, diameter=25 mm). The QMA filters were sub-sampled with a perspex
- 146 punch of 25 mm diameter. Silver, GF/F and QMA filters were dried overnight and prepared for beta counting (see
- 147 section 2.1). After counting the residual beta activity (~ 6 months later), samples were prepared for POC,
- 148 particulate nitrogen (PN) analyses along with their δ^{13} C and δ^{15} N isotopic compositions (here we present only POC
- 149 data). Filters were dismounted from filter holders and fumed with HCl vapor overnight inside a glass desiccator to
- 150 remove the carbonate phase. Samples were dried, packed in precombusted (450 °C overnight) silver cups, and
- 151 analyzed with an elemental analyzer - isotope ratio mass spectrometer (EA-IRMS, Delta V Plus, Thermo
- 152 Scientific). Acetanilide standards were used for the calibration. The detection limits and C blanks were respectively
- 153 0.63 and 0.80μ mol for Ag filters (n=11) and were 0.49 and 1.52 μ mol for QMA filters (n=13).

154The POC concentrations and 234 Th activities compared well between silver and GF/F filter types pointing to the155rather homogenous distribution of the particles on the Petex screen (234 Th_{GFF} = 0.63 × 234 Th_{silver} + 0.01 with r²=0.88,156p-value<0.01 and n=58; and POC_{GFF}= 0.86 POC_{silver} + 0.08 with r²=0.90, p-value<0.01 and n=58; Fig. S1),</th>

157 although concentrations from GF/F filters were systematically lower than those from silver filters, most likely

because of the different pore size filter (0.7 μm for GF/F filter vs 0.45 μm for silver filter).

2.3. Export fluxes of ²³⁴Th

Thorium-234 activity in surface waters can be described using a simple mass balance equation (Savoye et al.,
 2006), which accounts for production from ²³⁸U decay, ²³⁴Th decay, sinking flux and transport as follow:

162
$$\frac{dA_{Th}}{dt} = \lambda A_{U} - \lambda A_{Th} - P + V$$
(2)

163 where A_{Th} is the activity of total ²³⁴Th in dpm L⁻¹; A_U is the salinity-derived activity of ²³⁸U in dpm L⁻¹, λ is the

164 234 Th decay constant (0.0288 d⁻¹); P is the net removal of 234 Th on sinking particles in dpm L⁻¹ d⁻¹; V is the sum of

165 the advective and diffusive fluxes in dpm $L^{-1} d^{-1}$.

Assuming steady state (constant total ²³⁴Th activity with time) and neglecting the physical term V (Buesseler et al., 1992), the net export flux of particulate ²³⁴Th can be determined using the following equation:

168
$$P = \lambda \int_0^z (A_U - A_{Th}) dz$$
(3)

169 where P is the integrated flux of ²³⁴Th from the surface to the depth z in dpm m⁻² d⁻¹. Eq. 3 has been solved for z 170 taken as the depth (Eq) at the base of the 234 Th deficit zone (Eq = depth where 234 Th activity is back to secular 171 equilibrium with ²³⁸U) as well as for z representing the base of the primary production zone (PPZ), i.e. the depth 172 where *in-situ* fluorescence was only 10% of its maximum value (Owens et al., 2014). The Eq depth matched 173 relatively well with the PPZ depth, and on average, difference between both was only 16 m, with the largest 174 difference (~ 60 m) at Stations 1, 32 and 51 (Fig. 2). Considering that there can be export (or remineralisation) 175 below or above the PPZ depth, only the export fluxes at the Eq depth will be discussed as they represent the fully-176 integrated depletion of ²³⁴Th in the upper waters and thus the maximal export. The validity of the assumptions 177 used for solving Eq. 3 is discussed in Section 4.1.

178 In Section 4.1.2, we attempt to calculate the ²³⁴Th fluxes at the Eq depth by using a non-steady state (NSS) model

179 (Savoye et al., 2006), which can be described as follows:

180
$$P = \lambda \left[\frac{A_{U} \left(1 - e^{-\lambda \Delta t} \right) + A_{Th1} e^{-\lambda \Delta t} - A_{Th2}}{1 - e^{-\lambda \Delta t}} \right]$$
(4)

181 where Δt is the time interval between two visits of a single station; A_{Th1} and A_{Th2} are the ²³⁴Th activities at the first 182 and second visits, respectively. Without time series data, the calculation should not be performed sensu stricto 183 (Buesseler et al., 1992; Savoye et al., 2006). Thus, we chose to set the initial conditions for each station, as done 184 by Rutgers van der Loeff et al. (2011) in the South Atlantic. Satellite-derived PP data were used to estimate the 185 starting date of the bloom (i.e., when there is a PP increase of 30% above the winter value) and ²³⁴Th was assumed 186 to be in equilibrium with ²³⁸U at this time point. The time interval (Δt) for the calculations stretched from the 187 bloom start until the sampling date. All physical terms were considered negligible.

- 188 To estimate the intensity of shallow remineralization, export flux was also calculated for the Eq+100 m depth
- horizon. In case of any ²³⁴Th excess relative to ²³⁸U (i.e., ²³⁴Th/²³⁸U ratio > 1) below the Eq depth due to remineralisation, export fluxes integrated until Eq+100 m will be less than when integrated until Eq. Following Black et al. (2017) the reduction of the ²³⁴Th flux, R100, is expressed as:
- 192 $R100 = P_{Eq} P_{Eq+100}$ (5)
- 193 where R100 is the flux reduction in dpm m⁻² d⁻¹ and P is the 234 Th export flux estimated at Eq or Eq+100.

194 2.4. Scavenging fluxes of ²³⁴Th

To estimate the transfer rate of ²³⁴Th from the dissolved to the particulate form, i.e., the scavenging flux of ²³⁴Th (Coale and Bruland, 1985), we deduced the dissolved ²³⁴Th activities by subtracting the particulate (SSF+LSF) from the total ²³⁴Th activities, keeping in mind, though, that the sampling method for the total and particulate phases differed. Because the sampling resolution was different, total ²³⁴Th data were averaged at the sampling depth of particulate ²³⁴Th.

200 The mass balance equation for dissolved ²³⁴Th can be written as follows:

201
$$\frac{dA_{Thd}}{dt} = \lambda A_{U} - \lambda A_{Thd} - J + V$$
(6)

where A_{Thd} is the activity of dissolved ²³⁴Th in dpm L⁻¹; A_U and λ are defined in Eq. 2; J is the net removal flux from the dissolved to the particulate form (scavenging flux) in dpm L⁻¹ d⁻¹; and V is the sum of the advective and diffusive fluxes in dpm L⁻¹ d⁻¹.

Using again the steady state assumption (dissolved ²³⁴Th activities remain constant over time) and ignoring the
 physical terms (V), Eq. 6 becomes:

207
$$J = \lambda \int_0^z (A_U - A_{Thd}) dz$$
(7)

where J in dpm m⁻² d⁻¹ is the net flux of scavenging integrated to the depth z. In our case, the calculation was performed at the Eq depth for comparison with the 234 Th export flux (P in Eq. 3).

The comparison between the export flux (P) and scavenging flux (J) in terms of P/J ratio (export ratio) offers a valuable metric for estimating the export efficiency of 234 Th. A low P/J ratio (< 0.5) indicates that the removal of dissolved 234 Th is controlled by sorption onto suspended particles rather than export. Conversely, a high P/J ratio (> 0.5) indicates that 234 Th is preferentially exported rather than adsorbed and is thus efficiently removed from the upper waters.

215 2.5. POC:²³⁴Th ratios and POC export fluxes

We estimated POC export fluxes by multiplying the ²³⁴Th export flux with the POC:²³⁴Th ratio, both determined
at the Eq depth. A power law fit was used to determine the POC:²³⁴Th ratios at Eq (Fig. 3). Errors of the POC:²³⁴Th
ratios extrapolated at the Eq depth are deduced from the power law fit, using a root sum squared method. This

error is much larger than analytical errors of both POC concentrations and particulate ²³⁴Th activities. POC fluxes

- were determined by using the POC.²³⁴Th ratios of the LSF (> 53 μ m) as well as the SSF (1-53 μ m) samples, and both estimations were compared (Table 2).
- 222 The POC fluxes were between 1.1 to 1.5 fold higher when using the SSF POC:²³⁴Th ratio, except at stations 1, 26
- and 64. However, when considering the uncertainties, POC fluxes based on SSF and LSF POC.²³⁴Th ratios were
- not significantly different, and as we did not have the possibility to compare the POC:²³⁴Th ratios with those from
- sediment traps, we cannot affirm that the small particles participated to the export. As large and rapidly sinking
- particles usually drive most of the export (Lampitt et al., 2001; Villa-Alfageme et al., 2016), most of the studies
- dedicated to POC export fluxes in the North Atlantic used the POC:²³⁴Th ratios from the LSF (see Le Moigne et
- al., 2013b; Puigcorbé et al., 2017). Therefore, only the POC fluxes determined with the POC:²³⁴Th ratios from the
- LSF will be discussed (Table 3).

230 2.6. In-situ primary production

231 In order to determine the *in-situ* daily PP, stable isotope incubations were conducted using seawater collected at 232 different euphotic zone depths selected using photosynthetically active radiation (PAR) profiles, as described in 233 more detail in Fonseca-Batista et al. (2018). At each station, seawater was sampled from 3 to 6 depths (from 54 to 234 0.2% of surface PAR) and incubated on deck with a $H^{13}CO_3^-$ enriched substrate. After 24 h, incubated samples 235 were filtered through microglass fiber filters (MGF, 0.7 µm porosity, Sartorius). At the home-laboratory, POC 236 concentrations and isotopic composition were analyzed by EA-IRMS and uptake rates were deduced following the 237 Hama et al. (1983) method. Daily PP was then estimated by integrating the uptake rates from the surface down to 238 0.2% of surface PAR, which was located between 48 and 116 m depending on the station. The 0.2% of surface 239 PAR depth was roughly corresponding to the Eq depth (median difference between both depths: 20 ± 13 m) 240 although, a 42 m difference was observed at Station 1. Note that at Station 51, PP was determined 24 h after the 241 sampling of the total ²³⁴Th, particulate ²³⁴Th and POC.

242

2 2.7. Satellite primary production

243 PP was also obtained from satellite data products with a 9 km spatial resolution and 8-day temporal resolution, 244 available from the Ocean Productivity website Oregon University at State 245 (http://www.science.oregonstate.edu/ocean.productivity/) and obtained from MODIS and SeaWiFS satellites. 246 Three different models can be used to obtain satellite-derived PP: the standard Vertically Generalized Production 247 Model (VGPM; (Behrenfeld and Falkowski, 1997), the Eppley-VGPM (Eppley, 1972) and the Carbon-Based 248 Production Model (CbPM; Behrenfeld et al., 2005; Westberry et al., 2008). Among the model outputs, VGPM 249 derived PP (VGPM-PP) are closest to the *in-situ* PP measurements during our study (Fig. 4). Therefore, VGPM-250 PP is used in later discussion.

VGPM-PP data were averaged over 5 × 5 pixel boxes corresponding to a surface area of 2025 km² (45 km×45 km)
centered on the different sampling stations and the VGPM-PP was averaged for the week (8 days), the month (32
days) and the whole productive period prior to the sampling date. The whole productive period is the period
between the bloom start (defined by a PP increase of 30% above the winter value) and the sampling date (Fig. 5).
Differences between the different VGPM-PP estimates were smaller than a factor of 1.5.

256 **3.** Results

257 3.1. The Iberian basin (Stations 1 and 13)

Stations 1 and 13 were sampled 10 to 12 weeks after the start of the bloom (Fig. 4). At these stations, PP increased very early in the year (early to mid-March) and collapsed rapidly (end of March to mid-April). Within the Iberian

260 basin, low *in-situ* PP were determined (Table 3), with one of the lowest values measured at Station 1 (33 mmol m⁻

261 2 d⁻¹) and a moderate PP at Station 13 (79 mmol m⁻² d⁻¹; Fonseca-Batista et al., 2018; this issue).

- In line with low *in-situ* PP, low POC concentrations and particulate ²³⁴Th activities were determined in the Iberian basin (Table S2). POC:²³⁴Th ratios were low in both size fractions at Station 13, while Station 1 had high ratios, reaching 31 µmol dpm⁻¹ in surface for the LSF (Fig. 3). Similarly, Station 13 had the lowest LSF POC:²³⁴Th ratio
 extrapolated at Eq whereas Station 1 had one of the highest ratios (Table 3).
- 266 The 234 Th/ 238 U ratios were in the median of the range observed along the transect and reached minima of 0.68 and
- 267 0.70 in the upper 40 m at Stations 1 and 13, respectively (Fig. 2). Interestingly, these two stations vary also in their
- total particulate ²³⁴Th (sum of the SFF and LSF) over total ²³⁴Th ratios with only 9% of the ²³⁴Th in the particulate
- 269 phase at Station 1 and a ratio of 28% at Station 13 (in the median of those observed elsewhere along the transect).
- 270 At both stations, the ²³⁴Th export fluxes at the Eq depth were slightly higher than the median value observed along
- 271 the transect (1135 dpm m⁻² d⁻¹, n=11), reaching 1264 and 1418 dpm m⁻² d⁻¹ at Stations 1 and 13, respectively (Table
- 272 1). Compared to Station 1, the 234 Th scavenging flux was ~ 2 fold higher at Station 13 (1509 and 2898 dpm m⁻² d⁻
- 273 ¹, respectively; Table 1). Consequently, the export ratio (P/J) was higher at Station 1, reaching 0.84, compared to
- Station 13 (P/J ratio=0.49; Fig. 6). This indicates a balanced situation between P and J fluxes at Station 13 and a
 more efficient export of ²³⁴Th by sinking particles at Station 1.
- 276 Below Eq, significant excesses of ²³⁴Th relative to ²³⁸U (i.e., ²³⁴Th/²³⁸U ratio > 1.1) were observed at both stations, 277 indicating particle degradation (Fig. 2). However, significant shallow remineralisation was only observed at 278 Station 13 for which the R100 value was above uncertainty, reaching 410 \pm 218 dpm m⁻² d⁻¹ (Table 1). This 279 represents a flux reduction of 30% relative to the surface export flux.
- 280 Similarly, POC export fluxes varied between both stations with the highest (albeit with a strong associated error;
- 12 mmol m⁻² d⁻¹ at Station 1) and one of the lowest (2.2 mmol m⁻² d⁻¹ at Station 13) fluxes along the transect
 observed within this basin.

3.2. The west European basin (Stations 21 and 26)

- During 2014, the west European basin was the most productive with the highest PP peak observed at Station 21 (403 mmol m⁻² d⁻¹), 13 days before the sampling. At Station 26, the sampling took place during a secondary PP increase (Fig. 4 and 5). Altogether, sampling coincided with the bloom development in this basin, with *in-situ* PP reaching 135 and 174 mmol m⁻² d⁻¹ at Stations 21 and 26, respectively (Table 3).
- Along with high PP, relatively high surface POC concentrations and particulate 234 Th activities were measured averaging 3.7 µmol L⁻¹ and 0.2 dpm L⁻¹ for the LSF and, 5.4 µmol L⁻¹ and 0.5 dpm L⁻¹ for the SSF (Table S2). For
- both size fractions, POC:²³⁴Th ratios were high in the upper water column, reaching a maximum of 30 µmol dpm⁻
- ¹ for the LSF in surface waters at Station 21 (Fig. 3). At the Eq depth, the POC:²³⁴Th ratios for the LSF were in
- the median of those determined along the transect (4.4 μ mol dpm⁻¹, n=11) with nevertheless a lower ratio at Station
- **293** 21 (2.6 μmol dpm⁻¹; Table 3).

- The lowest ²³⁴Th/²³⁸U ratios were observed in the surface waters of the west European basin reaching minima of 294
- 295 0.57 and 0.77 at Stations 21 and 26, respectively (Fig. 2). Moreover, these low ratios were observed deeper in the
- 296 water column compared to the other basins. The integration of the ²³⁴Th deficit from the surface to the Eq depth
- led thus to high ²³⁴Th export fluxes at both stations. The ²³⁴Th export flux at Station 21 was one of the highest 297
- observed along the transect, reaching 1873 dpm m⁻² d⁻¹ (Table 1). The ²³⁴Th scavenging fluxes were also among 298
- 299 the highest observed along the transect, reaching 3917 and 2839 dpm $m^{-2} d^{-1}$ at Stations 21 and 26, respectively
- 300 (Table 1). The resulting export ratio (P/J) was close to 0.5 for both stations, indicating a balanced situation between
- 301 export and scavenging fluxes.
- 302 Excess of ²³⁴Th relative to ²³⁸U below Eq, was observed at both stations, with ²³⁴Th/²³⁸U ratios reaching 1.14 at 303 300 m for Station 21 (Fig. 2; Table S1). Consequently, the R100 value at this station was significantly positive $(360 \pm 255 \text{ dpm m}^{-2} \text{ d}^{-1}; \text{ Table 1})$, representing a 20% ²³⁴Th flux reduction. 304
- 305 Relatively high POC export fluxes at Eq were observed in the west European basin, reaching respectively 4.8 and
- 306 7.9 mmol m⁻²d⁻¹ at Stations 21 and 26. For the same area, other studies reported similar POC export fluxes during
- 307 May (Thomalla et al., 2008), and July-August (Lampitt et al., 2008; Le Moigne et al., 2013). However, Buesseler
- 308 et al. (1992) report much higher POC fluxes (up to 41 mmol m⁻² d⁻¹) for April-May during the North Atlantic
- 309 Bloom Experiment, highlighting an important temporal variability of POC export flux in this basin (Fig. 7).
- 310

3.3. The Icelandic basin (Stations 32 and 38)

- 311 In general, the different fluxes in the Icelandic basin presented similar characteristics to those in the west European 312 basin.
- 313 The bloom period started in May, one month before the sampling and the bloom maximum occurred after the
- 314 cruise (Fig. 4). Nevertheless, the basin was highly productive at Station 32 with *in-situ* PP reaching 105 mmol m⁻
- 315 2 d⁻¹ and was relatively productive at Station 38 (68 mmol m⁻² d⁻¹; Table 3 and Fig. 4 and 5).
- 316 POC concentrations and particulate ²³⁴Th activities were relatively high, but unlike for the west European basin
- the highest concentrations and activities were found in the SSF, reaching 5.8 µmol L⁻¹ and 0.4 dpm L⁻¹, respectively 317
- at Station 32 (Table S2). For surface waters of both stations, POC:²³⁴Th ratios in the SSF exceeded those in the 318
- 319 LSF (Fig. 3) but ratios were similar between the two size fractions at Eq depth (difference less than a factor of
- 320 1.1). The ratios extrapolated to Eq for the LSF were 3.6 and 4.2 μ mol dpm⁻¹ at Stations 32 and 38, respectively
- 321 and were in the median of the range along the transect (Table 3).
- 322 As for the west European basin, ²³⁴Th/²³⁸U ratios were low with station 38 having the lowest value for the whole
- 323 transect (0.50 in the surface; Fig. 2). Low ratios were also observed deeper in the water column and the combination
- 324 yielded the highest 234 Th export fluxes at Eq, reaching 2282 ± 119 dpm m⁻² d⁻¹ at Station 32 (Table 1). While the
- ²³⁴Th scavenging flux was high at Station 32, reaching 3690 dpm m⁻² d⁻¹, it was much lower at Station 38 (1495 325
- 326 dpm $m^{-2} d^{-1}$; Table 1). The export ratios (P/J) slightly exceeded the median value along the transect, reaching 0.62
- 327 and 0.76 at Stations 32 and 38, respectively. Despite similarities with the west European basin, the Icelandic basin
- 328 appeared more efficient to export ²³⁴Th by sinking particles.
- 329 Below the Eq depth, there was no significant excess of ²³⁴Th relative to ²³⁸U, resulting in R100 values being close
- 330 or below uncertainty and indicating absence of significant shallow remineralisation.
- 331 One of the highest POC export fluxes along the transect was determined at Station 32, reaching 8.3 mmol m⁻² d⁻¹
- 332 while the POC flux at Station 38 was lower (4.8 mmol m⁻²d⁻¹). Such POC export fluxes are lower than most values

- reported in earlier studies, ranging from 6 to up to 52 mmol $m^{-2} d^{-1}$ although Martin et al. (2011) reported a very low value of 0.8 mmol $m^{-2} d^{-1}$ (Ceballos-romero et al., 2016; Giering et al., 2016; Martin et al., 2011; Sanders et
- **335** al., 2010; Fig. 7).

336 3.4. The Irminger basin (Stations 44 and 51)

The ship crossed the Irminger basin one month after the beginning of the bloom and sampling occurred just 1 week (Station 44) to 3 weeks (Station 51) after the peak of the bloom (Fig. 4). At sampling time, the *in-situ* PP was amongst the highest observed along the whole section, reaching respectively 137 and 166 mmol m⁻² d⁻¹ at Stations 44 and 51. Such high values, in line with the satellite data, suggest that the bloom was still ongoing when visiting these two stations (Table 3 and Fig. 4 and 5).

- 342POC concentrations and particulate 234 Th activities were overall highest at these two stations, reaching 17 µmol L⁻343 1 and 1.2 dpm L⁻¹ for the SSF and 4.0 µmol L⁻¹ and 0.5 dpm L⁻¹ for the LSF at Station 44, respectively (Table S2).344POC: 234 Th ratios were moderate for both size fractions, reaching 14 µmol dpm⁻¹ for the SSF at Station 44 and 12345µmol dpm⁻¹ at Station 51 in the surface waters (Fig. 3). At the Eq depth, the extrapolated POC: 234 Th ratios were346similar between both size fractions at Station 51 but were 1.7 fold higher in the SSF at Station 44. The POC: 234 Th347ratio at Eq in the LSF at Station 44 fitted the median of the ranges determined along the transect, while the ratio at
- 348 Station 51 was relatively lower (2.9 μ mol dpm⁻¹, Table 3).
- The ²³⁴Th/²³⁸U ratios in the surface waters were higher than at other stations, reaching minima of 0.79 and 0.78 at 349 350 Stations 44 and 51, respectively. These low ²³⁴Th deficits were also restricted to the upper layer, especially at 351 Station 44 where the Eq depth was 40 m (Fig. 2). The particulate ²³⁴Th (sum of the SFF and LSF) contribution to total ²³⁴Th ratios varied widely, from 27% at Station 51 (in the median of those observed elsewhere along the 352 353 transect) to 94% at Station 44. The extremely high fraction of particulate ²³⁴Th at Station 44 reflects an important 354 particle concentration in surface waters. On the other hand, this high particulate fraction in the upper layer did not 355 induce a high export flux, since Station 44 had the lowest 234 Th export flux (321 ± 66 dpm m⁻² d⁻¹; Table 1) of all 356 stations. As a result, scavenging fluxes were much higher in this basin, reaching respectively 1802 and 2189 dpm 357 m⁻² d⁻¹ at Stations 44 and 51. This leads to very low P/J ratios in the Irminger basin (as low as 0.2 at Station 44),
- suggesting that export of ²³⁴Th is particularly inefficient in this basin, in agreement with the low export flux and
 the high particulate fraction in the upper layer.
- Below the Eq depth, there was no significant excess of ²³⁴Th relative to ²³⁸U, indicating no evidence for significant
 shallow remineralisation, with R100 values being either negative or below uncertainty.
- 362 The Irminger basin was characterized by low POC export fluxes (1.4 and 2.7 mmol $m^{-2} d^{-1}$ at Stations 44 and 51,
- 363 respectively). In the literature, a relatively large range of POC export fluxes has been reported for this basin.
- **364** Puigcorbé et al. (2017) observed POC export fluxes ranging from 1.5 to 43 mmol $m^{-2} d^{-1}$. Ceballos-Romero et al.
- 365 (2016) also determined much higher POC fluxes compared to those observed in the present study, with differences
- 366 reaching factors of 27 and 19 for the month before and after our sampling, respectively (Fig. 7).
- 367

3.5. The Labrador basin (Stations 64, 69 and 77)

368 Stations of the Labrador basin were sampled approximatively one month after the beginning of the bloom. Station

369 64 was sampled just after a second peak of the bloom while Stations 69 and 77 were sampled one week after this

370 peak (Fig. 4). At sampling time, the *in-situ* PP was low in the Labrador basin, ranging from 27 to 80 mmol $m^{-2} d^{-1}$

- ¹ at Stations 69 and 77, respectively (Table 3). In agreement with the satellite data shown in Fig. 4, this indicates
 that the decline of the bloom was ongoing in the Labrador basin.
- **373** POC concentrations and particulate ²³⁴Th activities were moderate to low, except at Station 77 where values were
- 374 higher in the surface, reaching 11 μ mol L⁻¹ and 0.45 dpm L⁻¹ for the SSF, and, 3.0 μ mol L⁻¹ and 0.20 dpm L⁻¹ for
- the LSF, respectively. Moderate POC:²³⁴Th ratios were observed in both size fractions, except in the upper layer
- at station 77 where SSF POC:²³⁴Th ratios were high (Fig. 3). At the Eq depth, POC:²³⁴Th ratios were similar in the
- **377** two size fractions and reached 9.2, 14 and 8.8 μ mol dpm⁻¹ at Stations 64, 69 and 77, respectively. Interestingly,
- 378 these ratios are higher than the median ratio determined along the transect.
- The surface 234 Th/ 238 U ratios were in the median of those observed along the transect (0.74 ± 0.06, n=8) with minima of 0.78, 0.66 and 0.73 at Stations 64, 69 and 77, respectively. These 234 Th deficits were nevertheless observed in a relatively shallow layer (Eq depths between 40 and 80 m in this basin; Fig. 2). Stations 64 and 69 were also characterized by a low particulate 234 Th activity (combined LSF and SSF) accounting for 10 and 15%
- 383 of the total ²³⁴Th activity in agreement with relatively low POC concentrations observed at these stations. The
- $\frac{234}{10}$ Th export flux at Station 64 was slightly greater than those of Stations 69 and 77 but, in general, the $\frac{234}{10}$ Th export fluxes of the Labrador basin were moderate, averaging 758 dpm m⁻² d⁻¹ (Table 1). $\frac{234}{10}$ Th scavenging fluxes were
- 386 also generally low in the Labrador basin, but with again, a slightly lower scavenging flux at Station 64 (Table 1).
- 387 A higher export ratio was thus estimated at Station 64 (P/J ratio = 0.75), suggesting a more efficient export close
- to the Greenland margin compared to Stations 69 and 77 (Fig. 6).
- Below Eq, there was a significant excess of ²³⁴Th relative to ²³⁸U at Stations 69 and 77, reaching respectively 1.08
- and 1.11. Evidence for shallow remineralisation was also clear from the R100 values exceeding uncertainties
- **391** (Station 69, R100=401 \pm 159 dpm m⁻² d⁻¹ and Station 77, R100=252 \pm 165 dpm m⁻² d⁻¹, Table 1). The flux
- reductions due to remineralisation below Eq were 50 and 40% of the fluxes at Eq, respectively.
- **393** High POC exports were observed within the Labrador basin and in particular at Station 69 where POC export flux
- reached 10 mmol m⁻² d⁻¹. As for the Irminger basin, Puigcorbé et al. (2017) determined a low POC export (0.7
- $mmol m^{-2} d^{-1}$ in May, one month before our sampling period, while Moran et al. (2003) observed higher fluxes
- reaching 47 mmol $m^{-2} d^{-1}$ in July, one month after our sampling period (Fig. 7).

397 4. Discussion

In the following section, we first discuss the potential impact of the physics and the non-steady state conditions on the ²³⁴Th export flux estimations. Then, temporal and regional variations of the carbon export fluxes are discussed with respect to the intensity and stage of the bloom, the phytoplankton size structure and the phytoplankton community. Finally, we examine carbon export and transfer efficiencies along the transect.

402

4.1. Validity of the export estimations

403

4.1.1. ²³⁴Th export fluxes under the potential influence of physical conditions

404 The GEOVIDE section sampled a diversity of dynamic regimes (Zunino et al., 2017) including continental margins

405 affected by strong zonal surface currents (LC, WGC and EGC; Mercier et al., 2015; Reverdin et al., 2003), local

- 406 and seasonal upwelling (close to the Iberian Margin), as well as deep convection zone in the Labrador Sea. In such
- 407 conditions, Eq. 3, which assumes negligible lateral and vertical advective and diffusive fluxes, may not always be

408 appropriate (Savoye et al., 2006). Whenever possible, we explore quantitatively or qualitatively, the potential409 errors arising from neglecting physical transport in our calculation.

- 410 Lateral processes associated with high velocity currents and intense mesoscale activity are known to affect the
- 411 ²³⁴Th distribution (Benitez-Nelson et al., 2000; Resplandy et al., 2012; Roca-Marti et al., 2016b; Savoye et al.,
- 412 2006). In our case, this may concern several stations located at or close to margins such as Stations 51 and 64,
- 414 Station 77 under the influence of the LC on the Newfoundland Margin and Station 1 under influence of the Portugal

which were respectively subject to the powerful East and West Greenland Currents at the Greenland Margin,

- 415 Current at the Iberian Margin (Fig. 1). However, the impact of lateral advection cannot be quantified from our
- 416 dataset, as the possible horizontal gradients of 234 Th cannot be resolved at sufficient resolution. As an alternative,
- 417 we compare stations close to each other such as Stations 44 and 51, both located in the Irminger Basin where
- 418 surface currents are strong. The Irminger basin in spring is a really patchy and dynamic area (Ceballos-romero et
- 419 al., 2016; Le Moigne et al., 2012; Puigcorbé et al., 2017) but the relatively high variability of the ²³⁴Th fluxes
- 420 found at these two stations (321 and 922 dpm $m^{-2} d^{-1}$, respectively) may also indicate a potential influence of lateral
- 421 advection. The higher export flux at Station 51 could reflect an input of ²³⁴Th depleted waters originating from the
- 422 Arctic and/or the Greenland shelf. However, Arctic (Cai et al., 2010; Roca-Marti et al., 2016a) and Greenland shelf
- 423 waters (Station 53, see Table S1) reveal very limited depletions of ²³⁴Th relative to ²³⁸U. Thus, it is reasonable to
- 424 consider that the ²³⁴Th deficit at Station 51 was essentially driven by vertical rather than horizontal processes.
- 425 Hydrodynamic processes could also impact open ocean sites, such as stations within the west European and 426 Icelandic basins (Stations 26 and 32) which are subjected to mesoscale activity. An inverse modeling study carried 427 out for the Porcupine Abyssal Plain located in the same region, suggests that the vertical transport of ²³⁴Th 428 associated with small-scale structures could represent up to 20% of the estimated vertical export flux (Resplandy 429 et al., 2012). This error is larger than our analytical uncertainty and should be kept in mind when considering the 430 export flux data in this area.
- In upwelling systems, the contribution of vertical advection on the ²³⁴Th distribution has been shown to be important (Buesseler, 1998; Buesseler et al., 1995). Near the Portuguese coast, the intensity of the upwelling is seasonally dependent (Costa Goela et al., 2016; Zúñiga et al., 2016) and was rather inactive at the time of the GEOVIDE cruise (<u>http://marine.copernicus.eu/</u>). Therefore, the input of ²³⁴Th-rich deep waters to the surface is likely to be limited, as already observed in the northern Iberian margin in early summer (Hall et al., 2000).
- 436 Downwelling systems, such as the intense convection that occurred in the Labrador basin during the winter prior
- 437 to our sampling (Kieke and Yashayaev, 2015), are also likely to impact the ²³⁴Th distribution. However, a strong
- 438 vertical advection would homogenize the ²³⁴Th activities in the water column, which is not the case during our
- 439 study (Fig. 2). Moreover, this convection ended more than two months before our sampling, a time lag that largely
- 440 exceeds the ²³⁴Th half-life thereby erasing any potential impact on the ²³⁴Th signal. Therefore, the influence of
- 441 vertical advection on ²³⁴Th export fluxes was neglected.

- 442 Finally, the contribution of the vertical diffusion was estimated using the vertical gradients of total ²³⁴Th activity
- 443 in upper waters and a Kz value ranging between 10^{-4} and 10^{-5} m² s⁻¹, as observed in the upper 1000 m between
- 444 Portugal and Greenland along the OVIDE transect (Ferron et al., 2014). The highest vertical diffusive flux was
- 445 determined at Station 69 and reached 181 dpm m⁻² d⁻¹, which is in the range of the ²³⁴Th flux uncertainties.
- 446 Therefore, the impact of the vertical diffusion has not been considered further.

- 447 In conclusion, hydrodynamic processes are likely to have at most a limited impact on the measured ²³⁴Th export
- fluxes.
- 449

4.1.2. Accounting for non-steady state conditions

As the cruise sampling scheme did not allow to collect samples through a time series, it was necessary to assume steady state conditions (i.e., no variation of ²³⁴Th activity with time). However, as documented in previous studies in the west European and Icelandic basins (Buesseler et al., 1992; Martin et al., 2011), this assumption can be questioned as large variations of ²³⁴Th activity were observed at a time scale of one to three weeks along with the onset of the seasonal biological productivity. As a consequence, the SS model was shown to poorly describe the magnitude of the ²³⁴Th export flux, leading to differences with the NSS model up to a factor of 3 (Buesseler et al., 1992; Martin et al., 2011).

457 During the weeks preceding GEOVIDE, large changes in satellite-derived PP were observed (Fig. 4). In order to
458 evaluate the potential error introduced by the SS approach, we attempted to apply a NSS model (see section 2.3;
459 Eq. 4).

460 The west European and Icelandic basins had the highest NSS ²³⁴Th fluxes (3540 dpm m⁻² d⁻¹ at Station 32) while 461 the Irminger basin had the lowest (516 dpm m⁻² d⁻¹ at Station 44; Table 1). The NSS ²³⁴Th fluxes were either larger 462 or similar to those obtained using the SS model. This results from the fact that the NSS approach used here assumes 463 the observed ²³⁴Th activity changes to only reflect a linear decrease from an initial ²³⁴Th activity in secular 464 equilibrium with 238 U, over the time elapsed since the onset of the bloom (Δt , see section 2.3). For stations sampled 465 shortly after the start of the bloom such as in the Irminger, Icelandic and Labrador basins (Δt ranges from 23 to 43 466 days), the fluxes predicted by the NSS model are from 1.4 to 2.1 fold higher than to the SS fluxes. In the west 467 European and Iberian basins, this difference is reduced (NSS fluxes are from 1.1 to 1.3 fold higher) due to a larger 468 Δt , ranging from 48 to 78 days.

469 As a conclusion, the SS export fluxes may have underestimated 234 Th export fluxes at some stations by a maximum

- 470 factor of 2 such as in the Icelandic basin. Yet, we need to keep in mind that this NSS approach has limitations by
- 471 assuming the equilibrium between ²³⁴Th and ²³⁸U at the bloom start and by considering only an increasing deficit
- 472 of ²³⁴Th activity over a given time period (Δt).

473 4.2. Influence of the intensity and stage of the bloom on POC exports

474 The GEOVIDE cruise was carried out in late spring (May-June), a period during which the productivity and the 475 carbon export can be important (Sanders et al., 2014). The ²³⁴Th proxy integrates the activity deficits over a 476 timescale of several weeks preceding the sampling and it appears thus essential to compare the sampling time in 477 light of the bloom development.

- 478 Apart from Stations 1 and 13, which were sampled after the bloom, the different basins were sampled during the
- 479 spring bloom, but at different stages. One of the lowest POC export flux was determined at Station 13 in the Iberian
- 480 basin, where the intensity of the bloom remained rather low during the whole productive period prior to the
- 481 sampling date (seasonal VGPM-PP=81 mmol $m^{-2} d^{-1}$, Fig. 5) due to oligotrophic conditions (depleted nutrients;
- 482 Fonseca-Batista et al., 2018). In contrast, the highest POC export flux was determined at Station 1, also in the
- 483 Iberian basin. Station 1 was sampled after the bloom period and satellite-data showed this station was relatively
- 484 productive in the early spring (185 mmol $m^{-2} d^{-1}$ in March, Fig. 4). This greater POC export observed when the

- 485 bloom had already declined may be caused by an ecosystem change, as already described in the Southern Ocean
- with the emergence of silicified diatoms because of nutrient stress (e.g., Baines et al., 2010; Claquin et al., 2002).
- 487 High POC export fluxes were also observed for the west European and Icelandic basins sampled during the bloom.
- 488 PP appeared maximal just before the sampling in the west European basin (Fig. 4 and 5) and could have promoted
- these high POC exports. Within the Icelandic basin, both stations were sampled during the productive period,
- 490 although the peak of the bloom was not yet reached (Fig. 4), suggesting that the export maximum might have
- 491 occurred later in the season. Both basins have previously been characterized by the presence of fast-sinking
- 492 particles during the bloom (data from cruises in Spring 2012 and Summer 2009; Villa-Alfageme et al., 2016)
- 493 promoting the high POC export fluxes.
- 494 The Irminger basin was sampled close to the bloom maximum, but unlike the west European and Icelandic basins
- the POC export flux was low there, probably reflecting accumulation of biomass preceding export. Indeed, this
- 496 area had the highest *in-situ* PP, a high proportion of particulate ²³⁴Th in surface waters (reaching 94% of the total
- 497 ²³⁴Th activity at Station 44) and a very low P/J ratio, indicating that ²³⁴Th was retained in the upper waters rather
- than being exported (Fig. 6; Table 1).
- The Labrador Sea basin was sampled just shortly after the peak of PP and was characterized by low *in-situ* PP, low nutrient concentrations, indicating the beginning of the decline of the bloom. The combination of the important PP a few weeks before our sampling (Fig. 4 and 5) and the decline of the bloom likely triggered the high POC export fluxes, as observed elsewhere (Martin et al., 2011; Roca-Marti et al., 2016b; Stange et al., 2016).
- 503 Overall, the magnitude of the POC export appears to depend on the degree of progress of the bloom. Indeed, the
- 504 negative relationship found between the POC export fluxes and the *in-situ* PP relative to the maximal VGPM-PP
- 505 along the season, representing the bloom stage, highlights that highest export occurs in post bloom periods (Fig.
- 506 8), as also evidenced from deep sediment trap studies (Lampitt et al., 2010), and is driven by large and rapidly
- sinking aggregates (Lampitt et al., 2001; Turner and Millward, 2002).

508 4.3. Influence of the phytoplankton size and community structure on POC exports

- 509 In the North Atlantic, the phytoplankton composition varies significantly, depending on the stage of the bloom and 510 on the evolution of environmental parameters such as micro- and macro-nutrient concentrations or stratification
- 511 depth (Moore et al., 2005). Spatial variations in phytoplankton size structure are known to exert a control on the
- 512 magnitude of the POC export flux (Boyd and Newton, 1999) and high POC exports are usually related to a greater
- 513 size of the sinking phytoplankton cells (Alldredge and Silver, 1988; Guidi et al., 2009).
- 514 Within the Iberian basin, the highest abundance of pico-phytoplankton was observed at Station 13 (Tonnard et al.,
- 515 in prep.). These conditions are typical of the subtropical and oligotrophic waters (Dortch and Packard, 1989).
- 516 Villa-Alfageme et al. (2016) highlighted that small cells are usually slow-sinking particles that can be easily
- 517 remineralised in the upper layers. A small sinking velocity (<100 m d⁻¹) allows time for bacteria and zooplankton
- 518 to degrade such particles, thus reducing the export flux. For the same area, Owens et al. (2014) also report a low
- 519 flux later in October, confirming a lower carbon export in general in this oligotrophic area. However, Station 1
- 520 was characterized by a greater POC export that could be related to the mixed proportion of micro-, nano- and pico-
- 521 phytoplankton and thus to the greater proportion of larger cells such as diatoms or haptophytes, increasing the
- 522 particle sinking velocity. The greater POC export there may also be related to the proximity to the margin, where
- 523 particle dynamics are intense and lithogenic particles are numerous (Gourain et al., 2018).

- 524 At higher latitudes, particle sinking velocity has been reported to be high (>100 m d^{-1} ; Villa-Alfageme et al., 2016),
- 525 as cells generally are of a larger size. Micro-phytoplankton, with dominance of diatoms, represented an important
- 526 fraction of the phytoplankton community in the west European, Irminger and Labrador basins and the dense
- 527 frustules of diatoms have been reported to act as ballast for the sinking organic matter (Klaas and Archer, 2002).
- 528 Fast-sinking particles could have promoted the relatively high POC export fluxes in those basins. However, in the
- 529 Icelandic basin, the dominance of nano-phytoplankton coincided with relatively high POC export. Both stations
- 530 in the Icelandic basin were dominated by haptophytes, including coccolithophorids (Tonnard et al., in prep.).
- 531 Despite their smaller size, the dense calcium carbonate shells of the latter could promote the export of POC
- 532 (Francois et al., 2002; Lam et al., 2011).
- 533 Our results suggest that high POC export fluxes can be mediated through either micro- or nano-phytopkankton 534 species, suggesting that sinking velocity is influenced by other parameters than the size, likely their composition 535 and density (Fig. 8).
- 536

4.4. Export and transfer efficiencies of POC

537 In order to characterize the strength of the biological carbon pump, we used two parameters: the export efficiency 538 (ThE), which is the ratio of the POC export flux at Eq over the PP (Buesseler, 1998) and the transfer efficiency 539 (T100) which is the ratio of the POC export flux at 100 m below Eq over the POC export flux at Eq (Fig. 9). Note 540 that the POC export flux at Eq+100 (Table 3) was calculated by multiplying the ²³⁴Th flux at Eq+100 by the POC to ²³⁴Th ratio of large particles for the same depth. The POC:²³⁴Th ratio at Eq+100 was deduced from a power law 541 542 fit (Fig. 3).

543 Based on in-situ PP values (Table 3), ThE ranged from 1 (Station 44) to 38% (Station 69) with a median value of 544 7% along the transect. The highest export efficiencies were determined at Stations 1 and 69 with values reaching 545 35 and 38%, respectively. Other stations were characterized by ThE \leq 14% with highest values (7 - 14%) at 546 Stations 32, 38, 64 and 77. Export efficiencies around 10% are common in the open ocean (Buesseler, 1998). A 547 lower export efficiency can be related to important microbial and zooplankton grazing activities or to biomass 548 accumulation in surface waters (Planchon et al., 2013, 2015). A high ThE can result from many factors such as the 549 presence of large and/or dense and fast sinking particles, low surface remineralisation, active zooplankton 550 migration or nutrient stress (Ceballos-romero et al., 2016; Le Moigne et al., 2016; Planchon et al., 2013). 551 Interestingly, stations with the highest ThE were also characterized by the lowest PP (Stations 1 and 69) while 552 stations with the lowest ThE were characterized by the highest PP (Stations 44 and 51). This inverse relationship 553 between PP and ThE was significant for all stations of the GEOVIDE cruise (regression slope: -0.20; r²=0.58; 554 p<0.01; n=11; Fig. S2) and has been explained in the Southern Ocean by the temporal decoupling between PP and 555 export due to biomass accumulation in surface waters (Henson et al., 2015; Planchon et al., 2013) as well as by 556 other processes such as zooplankton grazing and bacterial activity (Maiti et al., 2013; Le Moigne et al., 2016; 557 Roca-Marti et al., 2016a). Such particle recycling has also been observed in the North Atlantic (Collins et al., 558 2015; Giering et al., 2014; Marsay et al., 2015) limiting POC export to the deep ocean. A recent study in the 559 Icelandic and Irminger basins highlights the impact of the bloom dynamics on the particle export efficiency 560 resulting in strong seasonal variability of the ThE (Ceballos-Romero et al., 2016). Our estimates are generally in 561 the lower range of export efficiencies reported by others for the North Atlantic with values ranging from 1 to 42% 562 in the western European basin (Buesseler et al., 1992; Lampitt et al., 2008; Thomalla et al., 2008), from 5 to 8%

in the Icelandic basin (Ceballos-romero et al., 2016), from 4 to 16% in the Irminger basin (Ceballos-romero et al., 2016) and from 4 to > 100% in the Labrador basin (Moran et al., 2003). This wide range confirms that export
efficiencies are highly variable in the North Atlantic in late spring. The overall low export efficiency of the North
Atlantic is characteristic of highly productive areas of the world ocean.

567 However, it should be kept in mind that the ThE calculation is based on two parameters that are integrating 568 processes over different time scales: 24 h for *in-situ* PP and several weeks for export. Strong variability of PP 569 during this longer period would highly impact the ThE ratio. Therefore, ThE ratios were also estimated using the 570 VGPM-derived 8-day, 32-day and seasonal PP (Table 3). As seen in Section 2.7, there are no significant 571 differences between the VGPM-PP estimates regardless of the integrations times, and thus no significant 572 differences between the corresponding ThE values. Stations 1 and 69 are exceptions with ThE values decreasing 573 from 35 to 12% and from 38 to 8%, respectively, due to unusually low *in-situ* PP during our study which led to 574 over-estimated ThE.

575 Carbon transfer efficiencies (T100) ranged from 30 (Station 69) to 78% (Station 32). Generally, the fluxes at these 576 greater depths were characterized by greater error bars (see Fig. 9) due to the increasing uncertainty of the ²³⁴Th 577 fluxes with increasing depth. The highest T100 were observed within the Icelandic basin with values reaching 78 578 and 74% at Stations 32 and 38 respectively. On the contrary, the lowest T100 values were observed at Stations 1, 579 13, 21 and 69 (between 30 and 49%) highlighting greater carbon remineralisation between Eq and Eq+100 m at 580 these latter stations, as well as confirming important regional variability of the transfer efficiency as reported also 581 by others (Lam et al., 2011; Lutz et al., 2002). The low T100 (and high R100) values observed in the eastern part 582 of the transect (Stations 1, 13 and, to a lesser extent Station 21) likely reflect an important bacterial activity in 583 these warmer waters (>13°C in the upper 100 m; Iversen and Ploug, 2013; Marsay et al., 2015; Rivkin and 584 Legendre, 2001), efficiently degrading the probably slow-sinking particles. Such recycling is characteristic for 585 regeneration-based microbial food webs in oligotrophic regimes (Karl, 1999; Thomalla et al., 2006). In the 586 Icelandic basin, the high T100 may be related to the large abundance of coccolithophorids (Tonnard et al., in prep.) 587 known to enhance the POC transfer due to their ballasting effect (Francois et al., 2002; Lam et al., 2011). Indeed, 588 Bach et al. (2016) found that a bloom of coccolithophorids can increase the transfer efficiency through the 589 mesopelagic layer by 14-24%. Finally, the Labrador and Irminger basins exhibit relatively similar T100 (between 590 50 and 69%), except at Station 69 where the lowest T100 was observed. This is also in agreement with the highest 591 R100 and carbon remineralisation flux determined with the Baxs proxy (Lemaitre et al., 2018). The central 592 Labrador basin, in proximity of Station 69, was characterized by strong subduction of the LSW during the winter 593 preceding the GEOVIDE cruise. This downwelling could have promoted an important organic matter export 594 leading to important prokaryotic heterotrophic activity in mesopelagic waters. This enhanced remineralisation was 595 still observed during GEOVIDE as traced by a large mesopelagic Baxs content (Lemaitre et al., 2018).

596 5. Conclusion

597 Overall, POC export varied by a factor of ~ 9 along the transect highlighting an important spatial variation. POC
598 flux results obtained from other studies in the North Atlantic range from similar to up to 27 times larger values,
599 with rapid changes over a month duration, confirming the large temporal variability of the POC export fluxes.

- The magnitude of the POC export seems to be associated with the state of the bloom. Accumulation of biomass in
 surface waters during the bloom may induce a limitation of the POC export flux while during the post bloom
 period increasing numbers of rapidly sinking particles increases POC export.
- 603 The magnitude of the fluxes seems also related to the phytoplankton size and community structure. One of the 604 lowest POC export fluxes was found at the stations where pico-phytoplankton dominated the community. In 605 contrast, the areas composed by micro- and nano-phytoplankton were characterized by high POC export fluxes.
- 606 These areas were dominated by diatoms or coccolithophorids, known to strongly ballast the POC export fluxes.
- 607 This suggests that the size as well as the composition and density of the particles likely play an important role on
- the particulate sinking velocities and thus on the magnitude of the POC export fluxes.
- 609 For most stations, the fraction of primary production that is exported from the surface zone (export efficiency) was
- 610 $\leq 14\%$, which is in agreement with the global ocean export efficiency (~10%; Buesseler, 1998). Export efficiency
- 611 was also inversely related to primary production, indicating that the North Atlantic during our study behaved like
- 612 most of the highly productive areas of the world's ocean, with a low export efficiency. Finally, the fraction of POC
- 613 that is not remineralised in the mesopelagic zone (transfer efficiency) fits within the range of measured transfer
- 614 efficiencies reported elsewhere (e.g., Black et al., 2017; Buesseler and Boyd, 2009). The highest transfer
- 615 efficiencies were determined at the stations where coccolithophorids dominated.

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925 Table 1: Summary of the ²³⁴Th export and scavenging fluxes using steady state (SS) and non-steady state (NSS) models. The ²³⁴Th export fluxes using the SS model are calculated at the depths corresponding to the bottom of the primary production zone (PPZ), the equilibrium (Eq) depth and 100 m below Eq (Eq+100); the latter being used to estimate a remineralisation flux of ²³⁴Th (R100). Negative R100 values indicate an increase of the export flux between Eq and Eq+100. Note that the depth was fixed to 100 m at Station 26 because of the lower sampling vertical resolution. Consequently, the export flux at Eq+100 and the R100 were not determined at Station 26.

Basin	Station		Export Th export (SS) depth dpm m ⁻² d ⁻¹		Th export (NSS) dpm m ⁻² d ⁻¹			Th scavenging (SS dpm m ⁻² d ⁻¹				
		007	m				apm	m²	a⁻¹	apm	m²-	a⁼'
		PPZ	155	1327	±	137						
	1	Eq	90	1264	±	104	1442	±	80	1509	±	189
		Eq+100	190	1348	±	199						
		R100		-84	±	224						
Iberian												
		PPZ	82	1247	±	99						
	10	Eq	110	1418	±	111	1588	±	86	2898	±	285
	13	Eq+100	210	1008	±	187						
		R100		410	±	218						
		PPZ	82	1723	±	82						
		Eq	110	1873	±	97	2352	±	70	3917	±	212
	21	Eq+100	210	1513	±	235		_			_	
West		R100	210	360	±	255						
European		IX100		000	÷	200						
Luiopean		PPZ	95	1432	±	117						
	26				_	117	1069		00	2020		220
	26	Fixed	100	1486	±	117	1968	±	98	2839	±	220
		PPZ	75	4 4 5 5		00						
			75	1455	±	92	0540		440	0000		400
	32	Eq	130	2282	±	119	3540	±	113	3690	±	199
		Eq+100	230	2200	±	227						
		R100		81	±	256						
Icelandic												
		PPZ	70	1136	±	80						
	38	Eq	80	1134	±	95	2345	±	115	1495	±	160
	50	Eq+100	180	949	±	151						
		R100		185	±	178						
		PPZ	37	321	±	66						
		Eq	40	321	±	66	516	±	90	1802	±	71
	44	Eq+100	140	454	±	114						
		R100		-132	±	132						
Irminger												
Julia		PPZ	37	495	±	67						
		Eq	100	922	±	103	1625	±	108	2189	±	260
	51	Eq+100	200	873	±	103	1020	÷	100	2100	÷	200
		R100	200	49	±	154						
		PPZ	83	853	 	129						
		Eq	80	855		95	1423	<u>т</u>	122	1110	<i>.</i> т.	192
	64				±		1423	±	122	1142	±	192
		Eq+100	180	733	±	200						
		R100		123	±	221						
			05									
		PPZ	35	684	±	57						
Labrador	69	Eq	40	758	±	57	1068	±	53	1257	±	112
_00.000	00	Eq+100	140	357	±	148						
		R100		401	±	159						
		PPZ	55	693	±	77						
	77	Eq	60	696	±	77	1169	±	75	1529	±	148
	77	Eq+100	160	444	±	146						
			100	444	÷	140						

Basin	Station	LSF POC flux			SSF POC flux			
	#	mmol m ⁻² d ⁻¹			mmol m ⁻² d ⁻¹			
Iberian	1	12	±	22	6.9	±	2	
	13	2.2	±	0.3	3.3	±	0.6	
west	21	4.8	±	0.8	6.3	±	1.4	
European	26	7.9	±	5.0	6.1	±	3.7	
Icelandic	32	8.3	±	0.5	8.8	±	0.5	
	38	4.8	±	0.4	5.2	±	0.7	
Irminger	44	1.4	±	0.5	2.4	±	0.5	
	51	2.7	±	0.3	3.8	±	0.5	
	64	7.8	±	1.5	5.5	±	4.9	
Labrador	69	10	±	1	13	±	1	
	77	6.1	±	1.5	7.5	±	0.9	

Table 3: POC (particulate organic carbon) to ²³⁴Th ratios (in µmol dpm⁻¹) in the LSF, POC export fluxes (in mmol m⁻² d⁻¹), *in-situ* primary production (PP; Fonseca-Batista et al., 2018 and this study) and satellite-derived PP from the Vertically Generalized Production Model (VGPM-PP) integrated over 8 days, 32 days and over the whole

season (in mmol m⁻² d⁻¹) and the POC fluxes at Eq+100 m (in mmol m⁻² d⁻¹). Because of the lower vertical sampling resolution at Station 26, no POC export flux was

936 937 938 939 determined at Eq+100. *The sampling to determine the *in-situ* PP at Station 51 occurred 24 h after the sampling of the particulate ²³⁴Th and POC.

Station	POC: ²³⁴ Th at Eq	POC flux at Eq	in-situ PP	8-days VGPM-PP	32-days VGPM-PP	seasonal VGPM-PP	POC flux at Eq+100	
#	µmol dpm⁻¹	mmol m ⁻² d ⁻¹						
1	9 ± 17	12 ± 22	33 ± 2	76 ± 3	80 ± 11	96 ± 62	5.3 ± 23.2	
13	1.6 ± 0.2	2.2 ± 0.3	79 ± 3	64 ± 7	72 ± 18	81 ± 63	0.7 ± 0.2	
21	2.6 ± 0.4	4.8 ± 0.8	135 ± 2	161 ± 21	260 ± 97	201 ± 119	2.3 ± 0.4	
26	5.3 ± 3.3	7.9 ± 5.0	174 ± 19	77 ± 14	74 ± 19	112 ± 59		
32	3.6 ± 0.1	8.3 ± 0.5	105 ± 11	105 ± 7	95 ± 13	87 ± 13	6.5 ± 0.7	
38	4.2 ± 0.1	4.8 ± 0.4	68 ± 7	82 ± 5	94 ± 34	109 ± 32	3.5 ± 0.6	
44	4.4 ± 1.3	1.4 ± 0.5	137 ± 2	89 ± 3	110 ± 65	101 ± 66	0.8 ± 0.4	
51	2.9 ± 0.01	2.7 ± 0.3	*166 ± 32	95 ± 7	125 ± 118	125 ± 118	1.7 ± 0.2	
64	9.2 ± 1.1	7.8 ± 1.5	54 ± 18	59 ± 18	109 ± 115	103 ± 122	4.9 ± 1.5	
69	14 ± 0.04	10 ± 1	27 ± 5	108 ± 8	134 ± 80	134 ± 80	3.1 ± 1.3	
77	8.8 ± 1.9	6.1 ± 1.5	80 ± 21	108 ± 8	134 ± 80	134 ± 80	3.1 ± 1.3	

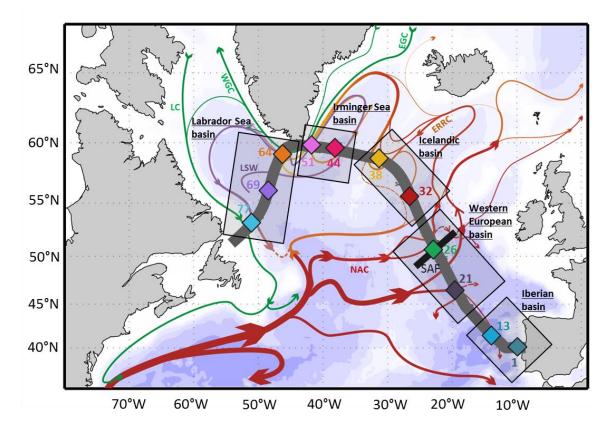
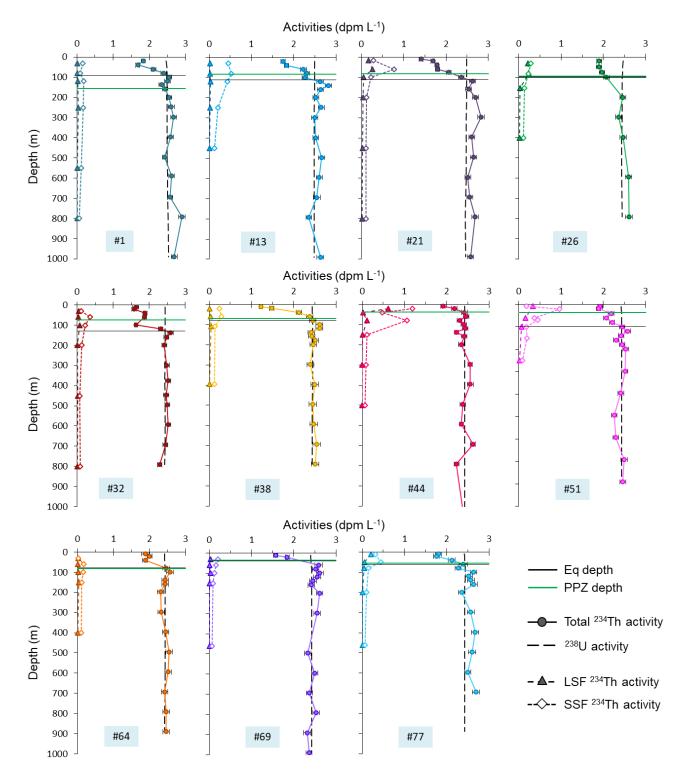
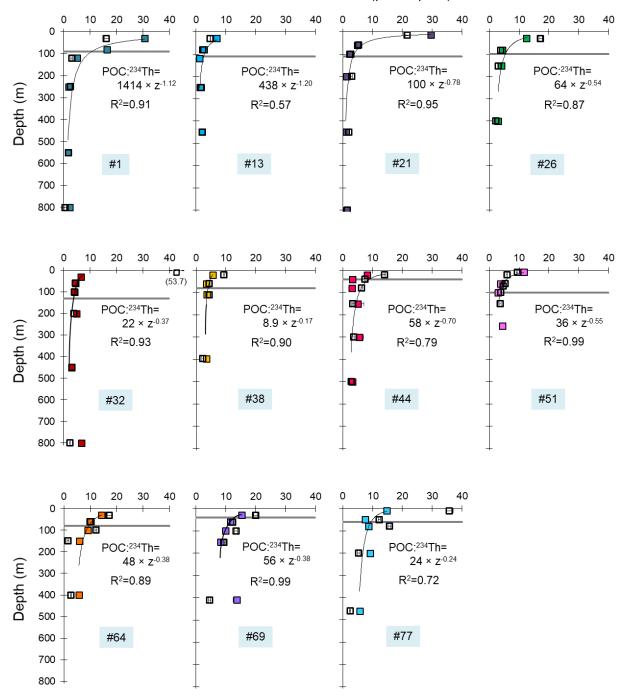


Figure 1: Simplified schematic of the surface circulation in the North Atlantic (adapted from Daniault et al., 2016)
superimposed with the GEOVIDE cruise track (thick grey line) and stations (colored diamonds). Main surface currents are
indicated: East Greenland Current (EGC), West Greenland Current (WGC), Labrador Current (LC), Eastern Reykjanes
Ridge Current (ERRC), North Atlantic Current (NAC). The Sub-Arctic Front (SAF) and the Labrador Seawater (LSW)
when in surface (i.e. within the Labrador basin) are also represented. The color codes for sampled stations are also used in
the following figures.



949Figure 2:Profiles of the total 234 Th (closed circles), total 238 U (black dotted vertical line) and particulate 234 Th activities for950the small size fraction (SSF; 1-53 µm; open diamonds) and for the large size fraction (LSF; >53 µm; closed triangles). All951activities are expressed in dpm L⁻¹. The horizontal black line is the Eq depth (depth where 234 Th returns to equilibrium with952 238 U), and the horizontal green line is the depth of the PPZ (primary production zone). Error bars are plotted but may be953smaller than the size of the symbols. Note that the Eq depth at Station 26 is set at 100 m because of the lower sampling954vertical resolution.



POC:²³⁴Th ratios (µmol dpm⁻¹)



Figure 3: Profiles of the POC:²³⁴Th ratios (μmol dpm⁻¹) in the SSF (open symbols) and LSF (closed symbols). The Eq depth,
 where ²³⁴Th is back to equilibrium with ²³⁸U, is indicated with the grey horizontal line. The thin black line represents the
 power law fit (POC:²³⁴Th=a×Z^{-b}) of the LSF. The median percentage errors on POC:²³⁴Th ratios are respectively
 representing 5 and 6% of the value for the SSF and the LSF. Error bars are plotted but may be smaller than the size of the
 symbols.

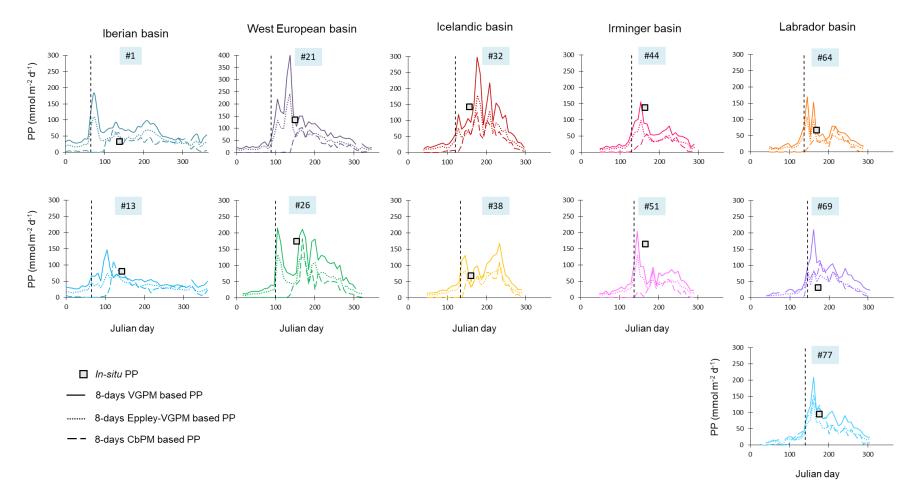


Figure 4: In-situ (squares) and satellite VGPM-derived (continuous lines), VGPM-Eppley-derived (dotted lines) and CbPM-derived (dashed lines) primary production (PP;
 in mmol m⁻² d⁻¹) data at the time of our sampling and along the year 2014. The start of the bloom, defined by a PP increase of 30% above the winter value, is indicated with the black vertical dashed line.

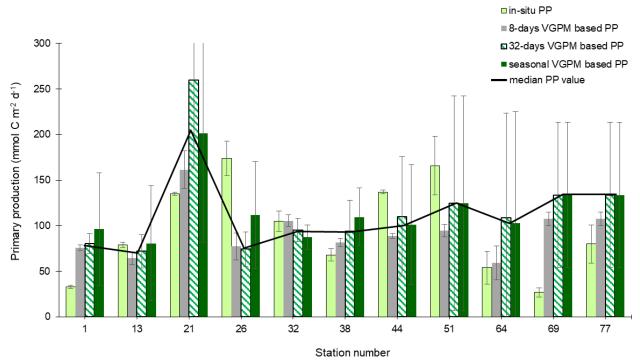
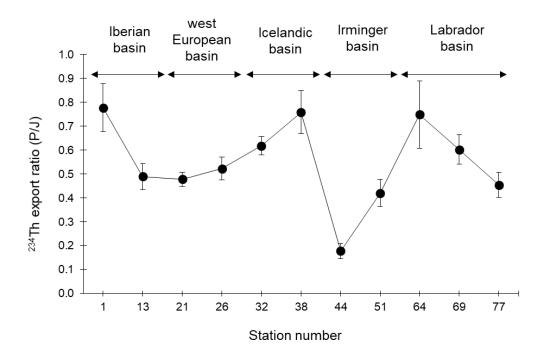


Figure 5: Comparison of *in-situ* and satellite VGPM (8-days, 32-days and seasonal averages) primary productivities (mmol m⁻² d⁻¹) along the GEOVIDE transect. The median of the four values is also indicated (black line).



969 <u>Figure 6:</u> Variability of the ²³⁴Th export ratio (i.e., the ratio of the ²³⁴Th export flux over the ²³⁴Th scavenged flux; P/J ratio)
 970 along the GEOVIDE section.

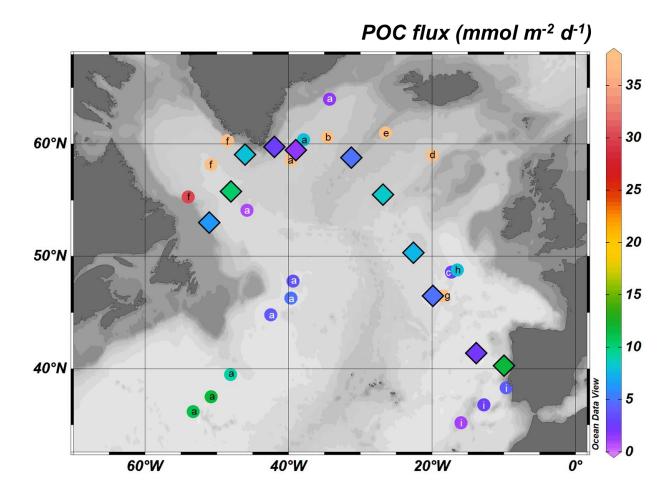
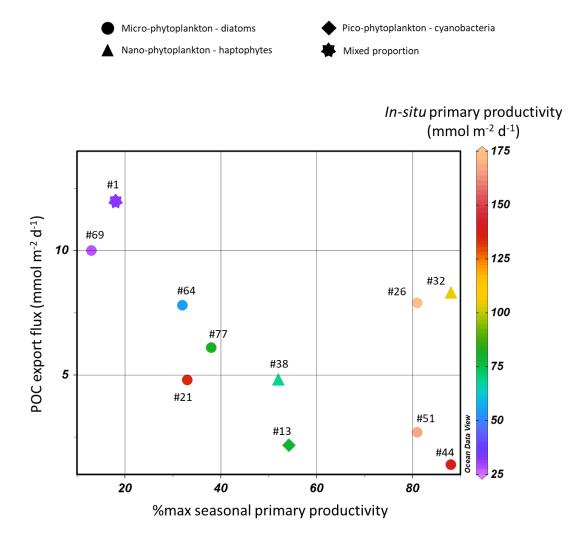
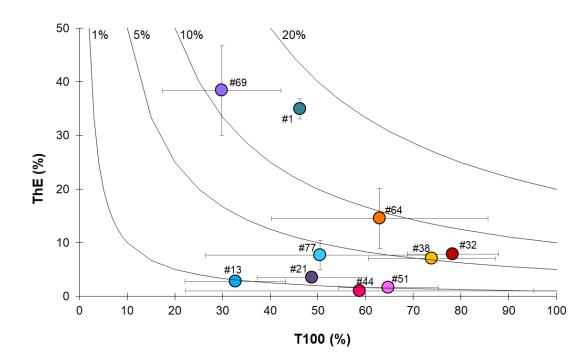




Figure 7: Comparison of the POC export fluxes from this study (diamonds with black borders) with other ²³⁴Th-derived estimates of POC exports in the North Atlantic (a: Puigcorbé et al., 2017; b: Ceballos-Romero et al., 2016; c: Thomalla et al., 2008; d: Sanders et al., 2010; e: Martin et al., 2011; f: Moran et al., 2003; g: Buesseler et al., 1992; h: Le Moigne et al., 2013; i: Owens et al., 2015).



987	Figure 8: Percentage of the <i>in-situ</i> primary productivity (PP) relative to the maximal VGPM-PP along the season (%max
988	seasonal primary productivity) in function of the POC export fluxes at the Eq depth. The %max seasonal primary
989	productivity illustrates the stage of the bloom (i.e., a %max seasonal primary productivity equalling 100%
990	corresponds to a sampling time at the bloom peak). This relationship is significant when not taking into account the
991	stations sampled between two PP peaks (Stations 26, 32 and 38, see Fig. 4): R2=0.77 and p-value<0.01. The <i>in-situ</i>
992	PP measured at sampling time is indicated with the colours in order to indicate the bloom intensity. The dominating
993	phytoplankton community is also indicated, with circles indicating micro-phytoplankton dominance (with a
994	majority of diatoms), triangles nano-phytoplankton dominance (with a majority of haptophytes) and diamonds
995	pico-phytoplankton dominance (with a majority of cyanobacteria). Note that Station 1 is represented by a star
996	because of the mixed proportion of micro-, nano- and pico-phytoplankton.



1001
1002Figure 9: Export efficiency (ThE = Export at Eq / *in-situ* PP) versus transfer efficiency (T100 = Export flux at Eq+100 /
Export flux at Eq). The black lines represent the modelled 1, 5, 10 and 20% of PP exported to depths > Eq+100 m.