Review on "Inputs and processes affecting the distribution of particulate iron in the North Atlantic along the GEOVIDE (GEOTRACES GA01) section" by Gourain et al.

I stopped reviewing the manuscript at line 366. Due to the state of the English language, sentences are difficult, some almost impossible to understand. This needs to be fixed before the next round of review should start. I am suggesting that a native English speaking person has a look and improves the English. Sorry for that, but I need to recommend major revision. However, I am still willing to review the next version, but insist on good English grammar. Sincerely,

Christian Schlosser

Dear Dr Schlosser,

This new version of the manuscript has been proof-read by a native English speaker. Following her advices and suggestions, we modified the structure of many sentences.

You will also find our detailed answers to the specific points your raised below.

We truly hope that you will now find this manuscript suitable for publication.

Kind regards,

Arthur Gourain, on behalf of all coauthors.

Some minor points:

Line 35: Change to "...(0.21 mol mol-1)..." Done.

Line 38ff: Include the second decimal digit for 0.7 and remove the ratio for the continental crust (you just told it 2 sentences earlier.

Done.

Line 42: Maybe "suspending" would be better than "delivering". We modified it.

Line 51: Here "deliver" would be better than "bring" Done.

Line 65ff: What do you mean with dissolution? Dissolution of inorganic PFe? Is not that also part of the regeneration?

Yes, we mean dissolution of inorganic PFe such as iron contained in basaltic grains, and this is now specified in line 67.

From our point of view, the regeneration is the recycling of biogenic PFe.

Line 101: Replace "They" by "Bottles" Done.

Line 123: Rewrite sentence "before to pass". Done. Line 189: Remove "really" Done. Line 190ff: What do you mean with "These authigenic particles lead to an enrichment of Mn in particle compositions." The elemental composition of the particles is driven by the various origins of these particles (lithogenic, biogenic, ...). For example, if the particulate bulk is dominated by lithogenic particles, its composition will have a strong imprint of lithogenic elements, e.g. Fe, Al, Ti. Under certain conditions, manganese oxides can be generated, with the consequence of depleting dissolved manganese concentrations, and increasing the ambient particulate Mn concentrations. As this statement was indeed confusing, we reorganized the order of sentences in this section (lines 192-200). Line 195: Replace "of direct and recent" by "for" Done Line 226: "modal", do you mean "mode"? Yes indeed, we changed it. Line 242: Change "Total particulate concentrations spanned a large range of concentrations from below detection to 304 nmol L-1 for PFe, 1544 nmol L-1 for PAI, 3.5 nmol L-1 for PMn and 402 nmol L-1 for PP." Done. Line 275ff: include "depth" after you introduce the depth of a sample, for example "at 25 m depth" in line286. Please apply throughout the paragraph/manuscript. Done. Line 299: Remove "candidate" Done. Line 300: Replace "ice shelves" by "glaciers" Done. Line 350: Remove "candidate" Done.

Figure 5: Please include in the legend what the different factors are (eg. lithogenic). Makes it easier to follow Done.

# Inputs and processes affecting the distribution of particulate iron in the North Atlantic along the GEOVIDE (GEOTRACES GA01) section

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- Abstract
- The aim of the GEOVIDE cruise (May-June 2014, R/V Pourquoi Pas?) aimed was to provide a better
- 27 understanding on of trace metal biogeochemical cycles in the North Atlantic Ocean. As marine particles play a
- 28 key role in the global biogeochemical cycle of trace elements in the ocean, we discuss the distribution of
- $29 \qquad \text{particulate iron (PFe), in } \\ \frac{\text{light-relation to the distribution of of}}{\text{particulate aluminium (PAI), manganese (PMn) and}} \\$
- phosphorus (PP) distributions. Overall, 32 full vertical profiles were collected for trace metal analyses,
   representing more than 500 samples. This resolution provides a solid basis for assessing concentration
- distributions, elemental ratios, size-fractionation, andor adsorptive scavenging processes in key areas of the
- 33 thermohaline circulation. Total particulate iron (PFe) concentrations ranged from as low as 9 pmol L<sup>-1</sup> in surface
- 34 <u>waters of the Labrador Sea waters to 304 nmol L-1 near the Iberian margin, while median PFe concentrations of</u>
- 35 1.15 nmol L<sup>-1</sup> were measured over the sub-euphotic ocean interior.
- Within the Iberian Abyssal Plain, the ratio of PFe over to particulate aluminium (PAI) wais identical to the
- 37 continental crust molar ratio (0.21), indicating the important influence of crustal particles in the water column.

Overall, the lithogenic component explained more than 87% of PFe variance along the section. Within the Irminger and Labrador basinss, the formation of biogenic particles led to an increase of the PFe/PAl ratio (up to 0.7 mol mol-¹) compared to the continental crust ratio (0.21 mol mol-¹). Continental mMargins provide important high quantities of particulate trace elements (up to 10 nmol L-¹ of PFe) to the open ocean, and, in the case of the IberianF-or example, margin, horizontal advection of PFe was visible more than 250km away from the Iberian margin. Additionally, several several benthic nepheloid layers spreading overwere observed more than 200 m above the seafloor were encountered along the transect, especially in the Icelandic, Irminger and Labrador basins, delivering particles with high PFe content of ,-up to 89 nmol L-¹.

### 1. Introduction

Particles play a key role in the ocean where they drive the residence time of most elements (Jeandel and Oelkers, 2015), and strongly influence the global biogeochemistry of macro- and micro-nutrients including iron (Milne et al., 2017). In the surface ocean, biological activity produces biogenic suspended matter through planktonic organisms, while atmospheric deposition (Baker et al., 2013; Jickells et al., 2005), riverine discharge (Aguilar-Islas et al., 2013; Berger et al., 2008; Ussher et al., 2004) or ice-melting (Hawkings et al., 2014; Lannuzel et al., 2011, 2014) bring mostly lithogenic derived particles to surface waters. These particulate inputs are highly variabley, both spatially and seasonally, around—in the world's oceans. At depth, benthic and shelf sediment resuspension (e.g. Aguilar-Islas et al., 2013; Cullen et al., 2009; Elrod et al., 2004; Fitzwater et al., 2000; Hwang et al., 2010; Lam et al., 2015; Lam and Bishop, 2008; McCave and Hall, 2002), and hydrothermal activity (Elderfield and Schultz, 1996; Lam et al., 2012; Tagliabue et al., 2010, 2017; Trefry et al., 1985), provides important amounts of particles to the water column. Moreover, authigenic particles can be produced *in-situ* by aggregation of colloids (Bergquist et al., 2007) or oxidation processes (Bishop and Fleisher, 1987; Collier and Edmond, 1984). Thus, oceanic particles result from a complex combination of these different sources and processes (Lam et al., 2015).

Particles represent the main part of the In the upper water column, the total iron pool in the upper water column is dominated by marine particles (Radic et al., 2011) which, and strongly interact with the dissolved pool (e.g. Ellwood et al., 2014). Indeed, dissolved iron can be scavenged onto particles (Gerringa et al., 2015; Rijkenberg et al., 2014), incorporated into biogenic particles (Berger et al., 2008) or produced by remineralisation of particles (Dehairs et al., 2008; Sarthou et al., 2008). Interestingly, the concept of "reversible scavenging" (i.e. release at depth of dissolved iron previously scavenged onto particles) has been advocated recently (Dutay et al., 2015; Jeandel and Oelkers, 2015; Labatut et al., 2014), while other studies reveal distinct dissolution processes (e.g. Oelkers et al., 2012; Cheize et al., 2018). Slow dissolution of particulate iron at margins has also been evoked as a continuous fertilizer of primary production and should be considered as a source of dissolved iron (e.g. Jeandel et al., 2011; Jeandel and Oelkers, 2015; Lam and Bishop, 2008). Within or below the mixed layer, the rates of regeneration processes can also impact the bioavailable pool of iron, among other trace metals (e.g. Ellwood et al., 2014; Nuester et al., 2014). However, the rates of these processes are not yet fully constrained. The study of particulate iron is thus essential to better constrain its marine biogeochemical cycle. Interest has gorwn in tThis subject\_received a growing interest over the last 10 years, in particular (e.g. Bishop and Biscaye, 1982; Collier and Edmond, 1984; Frew et al., 2006; Lam et al., 2012; Milne et al., 2017; Planquette et al., 2011, 2013; Sherrell et al., 1998) and, to our knowledge, only two studies have been performed at on an ocean-wide scale and published

so far: the GA03 GEOTRACES North Atlantic Zonal Transect (Lam et al., 2015; Ohnemus and Lam, 2015) and the GP16 GEOTRACES Pacific Transect (Lam et al., 2017; Lee et al., 2017).

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In this Within this global context, this paper presents the particulate iron distribution in of the North Atlantic Ocean, along the GEOTRACES GA01 section (GEOVIDE), and discusses the various sources and processes affecting its particulate iron distribution, using particulate aluminium, phosphorus or manganese distributions to support our conclusions.

# 2. Methods

### 2.1. Study area

Particulate samples were collected at 32 stations during the GEOVIDE (GEOTRACES GA01 section) cruise between May and June 2014 aboard the R/V *Pourquoi Pas?* in the North Atlantic Ocean (Sarthou et al., 2018). The sampling spanned several biogeochemical provinces (Figure 1), starting over that first comprised the Iberian margin (IM, Stations 2, 14 and 14), and proceding to the Iberian Abyssal Plain (IAP, Stations 11 to 17), the Western European Basin (WEB, Station 19 to Station 29) and the Icelandic Basin (IcB, Stations 32 to 36). Then, samples were collected above the Reykjanes Ridge (RR, Station 38), in the Irminger Basin (IrB, Stations 40 to 60), close to the Greenland shelf (GS, Stations 53, 56 and 61), the Labrador Basin (LB, Stations 63 to 77) and finally close the Newfoundland shelf (NS, Station 78) (Figure 1). The North Atlantic is characterized by a complex circulation (briefly described in section 23.1 and in detail by Zunino et al. (2017) and García-Ibáñez et al. (2015), and is one of the most productive regions of the global ocean (Martin et al., 1993; Sanders et al., 2014), with a complex phytoplankton community structure composed of diverse taxa (Tonnard et al., in prep.).

# 2.2. Sampling

Samples were collected using the French GEOTRACES clean rosette, equipped with twenty-two 12\_L GO-FLO bottles (two bottles were leaking and were never-not\_deployed during the cruise). GO-FLO bottles (General Oceanics) were initially cleaned in the home laboratory (LEMAR) following the GEOTRACES procedures (Cutter and Bruland, 2012). The rosette was deployed on a 14\_mm Kevlar cable with a dedicated, custom-designed clean winch. Immediately after recovery, the GO-FLO bottles were individually covered at each end with plastic bags to minimize contamination. They were then transferred into a clean container (class-100) for sampling, and the filters processed under a laminar flow unit. On each cast, nutrient and/or salinity samples were taken to check potential leakage of the GO-FLO bottles.

Filters were cleaned following the GEOTRACES protocols (http://www.geotraces.org/images/Cookbook.pdf) and kept in acid-cleaned 1 L LDPE bottles (Nalgene) filled with ultrapure water (Milli-Q, resistivity of 18.2 M $\Omega$  cm) until use. All filters were 25 mm diameter in order to optimize the signal over the filter blank except at the surface depth where 47 mm diameter filters were used. The filters were mounted on acid-cleaned polysulfone filter holders (Nalgene<sup>TM</sup>) were used. Prior to filtration, the GO-FLO bottles were shaken three times, as recommended in the GEOTRACES cookbook to avoid settling of particles in the lower part of the bottle. GO-FLO bottles were pressurized to <8 psi with 0.2  $\mu$ m filtered dinitrogen gas (N2, Air Liquide). Seawater was then filtered directly through paired filters (Pall Gelman Supor<sup>TM</sup> 0.45  $\mu$ m polyetersulfone, and Millipore mixed ester

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cellulose MF 5 µm) mounted in Swinnex polypropylene filter holders (Millipore), following Planquette and Sherrell (2012) inside the clean container. Filtration was operated until the bottle was empty or until the filter clogged; the volume filtered ranged from 2 Liters for surface samples to 11 L within the water column. After filtration, filter holders were disconnected from the GO-FLO bottles and a gentle vacuum was applied using a syringe in order to remove any residual water under a laminar flow hood. Filters were then removed from the filter holders with plastic tweezers that (which were rinsed with Milli-Q between samples). Most of the remaining seawater was removed via 'sippinged' by capillary action, when placing the non-sampled side of the filter onto a clean 47 mm supor filter. EThen, each filter pair was then placed in an acid-cleaned polystyrene PetriSlides (Millipore), double bagged, and finally stored at -20°C until analysis at LEMAR. Between casts, filter holders were thoroughly rinsed with Milli-Q, placed in an acid bath (5% Trace metal grade HCl) for 24 hours, then rinsed with Milli-Q.

At each station, process blanks were collected as follows:  $2_L$  of a deep (1000 m) and a shallow (40 m) seawater samples were first filtered through a 0.2  $\mu$ m pore size capsule filter (Pall Gelman Acropak 200) mounted on to the outlet of the GO-FLO bottle before to-passing through the particle sampling filter, which was attached directly to the swinnex filter holder.

### 2.3. Analytical methods

<u>IBack in</u> the home laboratory, sample handling was performed inside a clean room (Class 100). All solutions were prepared using ultrapure water (Milli-Q) and all plasticware had been acid-cleaned before use. Frozen filters, collected within the mixed layer-depth or within nepheloid layers, were first cut in half using a ceramic blade: one filter half was dedicated to total digestion (see below), while the other half was archived at -20°C for SEM analyses or acid leaching of "labile" metals (Berger et al., 2008; to be published separately).

or acid leaching of "labile" metals (Berger et al., 2008; to be published separately). Filters were digested following the method described in Planquette and Sherrell (2012). Filters were placed on the inner wall of acid-clean 15 mL PFA vials (Savillex TM), and 2 mL of a solution containing 2.9 mol L-1 hydrofluoric acid (HF, suprapur grade, Merck) and 8 mol  $L^{-1}$  nitric acid (HNO<sub>3</sub>, Ultrapur grade, Merck) was added to each vial. Vials were then closed and refluxed at 130°C on a hot plate for 4 hours, after which the and filters were removed. After cooling, the digest solution was evaporated at 110°C until-to near dryness. Then, 400 μL of concentrated HNO<sub>3</sub> (Ultrapur grade, Merck) was added, and the solution was re-evaporated at 110°C. Finally, the obtained residue was dissolved with 3\_mL of a-0.8 mol L-1 HNO3 (Ultrapure grade, Merck). This archived solution was transferred to an acid cleaned 15 mL polypropylene centrifuge tube (Corning®) and stored at 4°C until analyses. All analyses were performed on a sector field inductively coupled plasma mass spectrometer (SF-ICP-MS Element 2, Thermo-Fisher Scientific). Samples were diluted by a factor of 7 on the day of analysis in acid-washed 13 mm (outer diameter) rounded bottom, polypropylene centrifuge tubes (VWR) with 0.8 mol L-1 HNO<sub>3</sub> (Ultrapur grade, Merck) spiked with 1µg L-1 of 14ndium (115In) solution in order to monitor the instrument drift. Samples were introduced with a PFA-ST nebulizer connected to a quartz cyclonic spray chamber (Elemental Scientific Incorporated, Omaha, NE) via a modified SC-Fast introduction system consisting of an SC-2 autosampler, a sixport valve and a vacuum-rinsing pump. The autosampler was contained under a HEPA filtered unit (Elemental Scientific). Two 6-points, matrix-matched multi-element standard curves with concentrations bracketing the range of the samples were run at the beginning, the middle and the end of each analytical run. Analytical replicates were made every 10 samples, while accuracy was determined by performing digestions of the certified reference

material BCR-414 (plankton, Community Bureau of Reference, Commission of the European Communities), PACS-3 and MESS-4 (marine sediments, National Research Council Canada), following the same protocol as used for the samples. Recoveries were typically within 10% of the certified values (and within the error of the data, taken from replicate measurements, Table 1). Once all data were normalized to an <sup>115</sup>In internal standard and quantified using an external standard curve, the dilution factor of the total digestion was accounted for. Obtained The elemental concentrations were obtained per filter (pmol/filter) and were then corrected by the process blank-correcteds described above. Finally, pmol/filter values were divided by the volume of water filtered through the stacked filters.

Total concentrations (sum of small size fraction (0.45-5  $\mu$ m) and large (>5  $\mu$ m) size fraction) of particulate trace elements are reported in Table S1 (supplementary data).

2.4. Ancillary data:

 Potential temperature (0), salinity (S), and transmissometry data were retrieved from the CTD sensors (CTD SBE911 equipped with a SBE43).

# 2.54. Positive matrix factorisation

Positive Matrix Factorisation (PMF) was run to characterise the main factors influencing the particulate trace elements variances along the GEOVIDE section. In addition to PFe, PAI, PMn, and PP, nine additional elements were included in the PMF: Y, Ba, Pb, Th, Ti, V, Co, Cu and Zn. The analysis PMF whas been conducted on samples where all the 13-elements previously cited were above their detection limits; after selection, 445 of the 549 existing data points were used. Analyses were performed using the PMF software, EPA PMF 5.0, developed by the USA Environmental Protection Agency (EPA). Three to six factor mModels have been tested withwere run on the data, several factors number (from 3 to 6) T, after full error estimation of each model, we decide to use the configuration that provideding the lowest errors estimations and in consequence the (i.e. was the most reliable) was the:

In consequence, models were set up with four factor model. To ensure stability this model wass and were run 100 times to observe the stability of the obtained results. After displacement, error estimations and bootstraps error estimations, the model was recognised as stable.

# 2.65. Derived and ancillary parameters

To investigate The PFe/PAl ratio can be used to estimate the proportion of lithogenic particlesiron within the bulk particulate fractiiron, we used. A comparison with the Upper Continental Crust (UCC) Fe/Al molar ratio (0.21) of Taylor and McLennan (1995) s. 0.21, was used to calculate the lithogenic components of particles (%PFe<sub>litho</sub>) following Eq. (1):

%PFe litho =  $100 * \left(\frac{PAI}{PFe}\right)$  sample  $* \left(\frac{PFe}{PAI}\right)$  UCC ratio \_\_\_\_\_(1)

Then the non-lithogenic PFe is simply obtained using Eq. (2):

196 %PFe non\_litho = 100 - %PFe litho \_\_\_\_(2)

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Note that while tBoth the lithogenic and non-lithogenic fraction of PFe are estimated using the UCC ratio. Spatial and temporal variation of the lithogenic components ratio may falsely influence the estimated fraction value. The %PFe<sub>litho</sub> and %PFe<sub>non-litho</sub> proxies are interesting tools to evaluate the importance of lithogenic and non-lithogenic (either biogenic or authigenic), but they have to be used carefully, as the spatial and temporal variation of the lithogenic component ratios may falsely influence the estimated fraction value.

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In addition to PAI, PMn can be used as a tracer of inputs from shelf resuspension (Lam and Bishop, 2008). Indeed, Mn is really sensitive to bacteria-mediated oxidation mediated by bacteria (Tebo et al., 1984; Tebo and Emerson, 1985) and forms manganese oxides (MnO<sub>2</sub>). These authigenic particles lead to an enrichment of Mn in particles compositions. In order to track the influence of shelf resuspension, a percentage of sedimentary inputs "%bulk sediment inputs" can be estimated using the PMn/PAl ratio from the GEOVIDE samples and the PMn/PAl UCC

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%bulk sediment PMn 
$$= 100 * {PAI \choose PMn} sample * {PMn \choose PAI} UCC ratio ___(3)$$

This proxy iscan be a good indicator of direct and recent sediment resuspension. We assume that particles newly

value (0.0034; Taylor and McLennan, 1995) according to the following equation:

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resuspended in the water column will have the same PMn/PAl ratio thans the UCC ratio, leading to a "%bulk sediment Mn" of 100%. This value will decrease by authigenic formation of Mn oxides. This proxy assumes homogeneity of the sediment PMn/PAl ratio throughout the GEOVIDE section. However, which this is maybe

215 not be completely the case at every station. In consequence, this proxy isshould only a toolbe used to identify new 216

benthic resuspension at specific locations; and inter-comparison between several locations is not possible may not be appropriate. When a sample presents a "%bulk sediment Mn" greater than 100%, we have assigned a maximum

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value of 100% to simplify the following discussion. As the Mn cycle can also be affected influenced by bioticlogie uptake (e.g. Peers and Price, 2004; Sunda and Huntsman, 1983), this proxy is only used at depths where biologic

220 activity wais negligible (i.e. below 150m depth).

Potential temperature ( $\theta^{\circ}\theta$ ), salinity (S), and transmissometry data were retrieved from the CTD sensors (CTD

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SBE911 equipped with a SBE43). 223

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3. Results

3.1. Hydrography and biological-setting

Here, we briefly describe the hydrography encountered during the GEOVIDE section (Figure 2), as a thorough description is available in García-Ibáñez et al. (2015). In the beginning At the start of the section-, Tithe warm and salty Mediterranean Water (MW, S=36.50, 0°=11.7°C) was sampled between 600 and 1700 m in the Iberian Abyssal Plain (IAP). MW resulted from the mixing between the Mediterranean Overflow Water plume coming from the Mediterranean Sea and local waters. Surface water above the Iberian Shelf was characterised by low

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salinity (S=34.95) at station 2 and 4 compared to surrounding water masses. Close to the floor of the Iberian Abyssal Basin, the North East Atlantic Deep Water (NEADW, S=34.89,  $\theta^{\circ}$ =2.0°C) spread southnorth ward. The North Atlantic Central Water (NACW, S>35.60,  $\theta^{\circ}$ >12.3°C) was the warmest water mass of the transect and was observed in the subsurface layer of the Western European Basin and Iberian Abyssal Plain. An old Labrador Sea Water (LSW, S=34.87,  $\theta^{\circ}$ =3.0°C) flowed inside the Western European and Icelandic Basins, between 1000 and 2500m depth.

-In the Icelandic Basin, below the old LSW, the Iceland-Scotland Overflow Water (ISOW, S=34.98,  $\theta^{\circ}$ =2.6°C)

In the Icelandic Basin, below the old LSW, the Iceland-Scotland Overflow Water (ISOW, S=34.98,  $\theta^{\circ}$ =2.6°C) spread along the Reykjanes Ridge slope. This cold water, originating from the Arctic, led to the formation of NEADW after mixing with surrounding waters. North Atlantic hydrography was impacted by the northward flowing of the North Atlantic Current (NAC), which carried up-warm and salty waters from the subtropical area. ByDue to air-sea interactions and mixing with surrounding water, the NACW is cooled down-and freshened in the subpolar gyre and is transformed in Subpolar Mode Water (SPMW). When NAC crossed the Mid Atlantic ridge through the Charlie Gibbs Fracture Zone (CGFZ), it created the Subpolar Mode Water (SPMW). The recirculation-formation of SPMW inside the Icelandic and Irminger Basins leads to the formation of regional modal waters: the Iceland Subpolar Mode Water (IcSPMW, S=35.2,  $\theta^{\circ}$ =8.0°C) and the Irminger Subpolar Mode Water (IrSPMW, S=35.01,  $\theta^{\circ}$ =5.0°C), respectively. IcSPMW was a relatively warm water mass with potential temperature up to 7°C (García-Ibáñez et al., 2015). Another branch of the NAC mixed with Labrador Current waters to form the relatively fresh SubArctic Intermediate Water (SAIW, S=<34.8, 4.5°C< $\theta^{\circ}$ <6°C).

The Irminger Basin is a complex area with a multitude of water masses. In the middle of the basin, an old LSW, formed one year before (Straneo et al., 2003), spread between 500 and 1200 m depth. Close to the bottom, the Denmark Strait Overflow Water (DSOW, S=34.91) flowed across the basin. Greenland coastal waters were characterised by low salinity values, down to S=33. The strong East Greenland Current (EGC) flowed southward along the Greenland shelf in the Irminger Basin. When reaching At the southern tip of Greenland, this current entersed the Labrador Basin along the west coast of Greenland and followed the outskirts outline of the basin until the Newfoundland shelf. In the Labrador Basin, the deep convection of SPMW at 2000 m was involved in the formation of the LSW (S=34.9,  $\theta$ °C=3.0°C) (García-Ibáñez et al., 2015; Yashayaev and Loder, 2009). Above the Newfoundland Shelf, surface waters were affected by discharge from rivers and ice-melting and characterised by extremely low salinity for open ocean waters, below 32 in the first 15 meters.

# 3.2. Section overview

Total particulate iron (PFe), aluminium (PAI), manganese (PMn) and phosphorus (PP) concentrations spanned a large range of concentrations from below detection to 304, 1544, -3.5 and 402 nmol L<sup>-1</sup> respectively. The ranges of concentrations are comparable to other studies recently published (Table 2).

PFe, PAI, and PMn were predominantly found (>90%) in particles larger than 5  $\mu$ m, except in surface waters, where 9  $\pm$  8.6 % of PFe, 38.8  $\pm$  8.6 % of PP, 10.9  $\pm$  15.4 % of PAI and 32.8  $\pm$  16.6 % of PMn 38.8  $\pm$  8.6 % of PP were hosted by smaller particles (0.45-5  $\mu$ m). The ranges of concentrations are comparable to other studies recently published (Table 2). Data are shown in Figure 3.

3.3. Open Ocean stations: stations: from the Iberian Abyssal Plain (stations 11 to 17), Western European Basin (stations 19 to 29), Icelandic Basin (stations 32 to 36), Reykjanes Ridge (station 38), Irminger Basin (stations 40 to 60; except Stations 53 and 56) and to the Labrador Basin (stations 6311 to 77, except stations 53, 56and 61)

This concerns all stations between from station 11 to 77, with the exception of stations 53, 56 and 61 that which were sampled close to the Greenland and Newfoundland coast (Figure 1)s, respectively.

Particulate iron concentration vertical-profiles presented showed identical patterns at all of the open ocean stations sampled in each oceanic basin encountered along the section. Median PFe wasere low at 0.25 nmol  $L^{-1}$  within the first 100 m and steadily increased with depth. However, at two stations, elevated concentrations were determined in the upper 100 m, up to 4.4 nmol  $L^{-1}$  at station 77 at 40 m and 7 nmol  $L^{-1}$  at station 63 between 70 and 100 m depth. PFe concentrations gradually increased with depth, with a median PFe of 1.74 nmol  $L^{-1}$  below 1000m. Close to the seafloor of some stations (26, 29, 32, 34, 49, 60, and 71), high concentrations of PFe were observed, up to 88 nmol  $L^{-1}$  (station 71 at 3736 m). These high PFe values were associated with low beam transmissometry values  $\leq$  inferior or equal to 977-%. (Figure 9b and supplementary table S2).

Particulate aluminium (PAI) and manganese (PMn) profiles were similar to PFe profiles, with low concentrations measured in the first 100 m (1.88 nmol L<sup>-1</sup> and 55 pmol L<sup>-1</sup>, respectively) and—which increased towards the seafloor. Close to the seafloor, high concentrations were determined at the same stations cited above for PFe, with a maximum of 264 nmol L<sup>-1</sup> and 3.5 nmol L<sup>-1</sup> for PAI and PMn respectively at station 71 (supplementary Table S1). Highest particulate phosphorus (PP) concentrations were in the first-uppermost 50 m, with a median value of 66 nmol L<sup>-1</sup>. Deeper in the water column, below 200 m, PP concentrations decreased to values below 10 nmol L<sup>-1</sup>. Inter-basins differences were observed within the surface samples, with median PP concentration being higher in the Irminger Basin (127 nmol L<sup>-1</sup>) than in the Iberian Abyssal Plain (28 nmol L<sup>-1</sup>) (Figure 3).

Finally, above the Reykjanes Ridge, PP, PMn, PAI and PFe concentrations were in the same range than-as the surrounding open ocean stations. However, close to the seafloor, high concentrations were measured, with PFe, PAI, and PMn reaching 16.2 nmol L<sup>-1</sup>, 28.8 nmol L<sup>-1</sup>, and 0.51 nmol L<sup>-1</sup> at 1354 m, respectively (Figure 3 and supplementary material Table S1).

3.4. Margins and Shelves: Iberian Margin (stations 1 to 4), Greenland coast (stations 53, 56 and 61) and Newfoundland Shelf (station78)

The Iberian margin was characterised by low beam transmissometry values at station 2 (88% at 140 m, Figure 49b) suggesting significant particle concentrations. Particulate iron concentrations varied between from 0.02 nmol L<sup>-1</sup> to 304 nmol L<sup>-1</sup>. Within the first 50\_m, PFe concentrations decreased towards the shelf break where PFe dropped down from 2.53 nmol L<sup>-1</sup> (station 2) to 0.8 nmol L<sup>-1</sup> (Station 1). At tall three stations. PFe concentrations increased with depth at all three stations and reached a maximum at the boclose to the seafloor. As an For example, 300 nmol L<sup>-1</sup> of PFe was measured determined at 138.5 m at three of station 2 (138.5 m) with more than 300 nmol L<sup>-1</sup> of PFe. Lithogenic tracers, such as PAI or PMn, presented similar profiles to PFe with concentrations ranging between from 0.11 and 1544 nmol L<sup>-1</sup>, and from below detection limit to 2.51 nmol L<sup>-1</sup> respectively (Figure 3, supplementary material Table S1). Total particulate phosphorus (PP) concentrations were relatively

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low in the surface ranging from undetectable values below detection to 38 nmol  $L^{-1}$ ; concentrations decreased with depth and were less than 0.7 nmol  $L^{-1}$  below 1000 m depth.

In the vicinity of the Greenland shelf, PFe concentrations had a high median value of 10.8 nmol  $L^{-1}$  and were associated with high median PAI and PMn concentrations of 32.3 nmol  $L^{-1}$  and 0.44 nmol  $L^{-1}$ , respectively. Concentrations of PP were high at the surface with a value of 197 nmol  $L^{-1}$  at 25 m of station 61. Then, PP concentrations decreased strongly, to less than 30 nmol  $L^{-1}$ , below 100 meters depth. Furthermore, beam transmissometry values in surface waters at these three stations, were the lowest of the entire section, with values below 85-%.

Close to the Newfoundland margin, surface waters displayed a small load of particulate trace metals as PFe, PAl, and PMn were below 0.8 nmol L<sup>-1</sup>, 2 nmol L<sup>-1</sup>, and 0.15 nmol L<sup>-1</sup>,\_respectively. Then, close to the bottom of station 78, at 371 m, beam transmissometrytry values dropped to 94% and were associated with extremely high concentrations of PFe=168 nmol L<sup>-1</sup>, PAl=559 nmol L<sup>-1</sup>, and PMn=2 nmol L<sup>-1</sup>. Total PP concentrations in the first 50 m ranged from 35 to 97 nmol L<sup>-1</sup>. Below 50 m-the surface, PP remained relatively high with values up to 16 nmol L<sup>-1</sup> throughout the water column. (Figure 3 and supplementary material-Table S1).

4. Discussion

Our goal in this work was to investigate mechanisms that drive the distribution of PFe in the North Atlantic, in particular the different routes of supply and removal. Possible candidate sources of PFe include lateral advection offshore from margins, atmospheric inputs, continental run-off, melting ice shelves and icebergs, resuspended sediments, hydrothermal inputs and biological uptake. Removal processes include remineralization, and dissolution processes and sediment burial.

In the following sections, we examine each of these sources and processes, explore the evidence for their relative importance, and use compositional data to estimate the particle types and host phases for iron and associated elements.

<u>4.1. Analysis of the principal factors controlling variance: near-ubiquitous influence of crustal particles in the water column</u>

PThe positive matrix factorisation analysis (Figure 5) has been realisedwas undertaken on the entire dataset, in consequence, the factors described below are highly influenced by the major variations of particulate element concentrations (usually at the interfaces, i.e. margin, seafloor, surface layer). The positive matrix factorisation, shown in Figure 5, indicates the overall variances explained by each of the 45 factors. The first factor is characterised by lithogenic elements, representing 86.8% of the variance of PFe, 75.8% of PAI and 90.5% of PTi. The second factor is correlated with both Mn and Pb and explains no less than 76.5% and 77.0% of their respective variances. Ohnemus and Lam (2015) observed this co-relation between manganese and lead particles and explained it by the co-transport on Mn-oxides (Boyle et al., 2005). The formation of barite is causing explains the third factor, and constraineding 87.7% of the Ba variance in the studied regions. Biogenic barite accumulation within the mesopelagic layer is related to bacterial activity and organic remineralisation of biogenic material (Lemaitre et al., 2018a). A biogenic component is the fourth factor and explained most of particulate phosphorus PP variance, 83.7%. The micronutrient trace metals, copper, cobalt and zinc, had more than a quarter of their variances influenced by this factor. Note that the biogenic contribution to

PFeparticulate iron and other particulate trace elements will be discussed in another paper (Planquette et al., in pren)

These results indicate that along the GA01 section, PFe distributions were predominantly controlled by lithogenic material and to a smaller extent by remineralisation processes (as seen by a Factor 3 contribution of 4.1%). This does not rule out some biogenic influences on PFe distribution, especially in <a href="the-surface">the surface</a>, but this contribution is <a href="weiled-obscured">weiled-obscured</a> by the high lithogenic contribution. The PMF analysis has been realised on the entire dataset, in <a href="consequence">consequence</a>, the factors described are highly influenced by the major variations of particulate element <a href="consequence">concentrations (usually at the interface, i.e. margin, seafloor, surface layer,...)</a>.

To further investigate the influence of crustal material on the distribution of PFe, it is instructive to examine the distribution of the molar ratio of PFe/PAl, and the resulting %PFe<sub>litho</sub> (see section 2.6 for definition of this parameter) along the section as a way to assess the lithogenic inputs (Lannuzel et al., 2014; Ohnemus and Lam, 2015; Planquette et al., 2009) (Figure 6) along the section.

The PFe/PAI ratio can be used to estimate the proportion of lithogenic particles within the bulk particulate material. A comparison with the Upper Continental Crust (UCC) ratio of Taylor and McLennan (1995), 0.21, was used to calculate the lithogenic components of particles (PFe<sub>htho</sub>) following Eq. (1):

%PFe litho = 
$$100 * {PAI \choose PEP} sample * {PFe \choose PAI} UCC ratio (1)$$

Then the non-lithogenic PFe is simply obtained using Eq. (2):

 $%PFe non\_litho = 100 - %PFe litho$  (2)

Both the lithogenic and non-lithogenic fraction of PFe are estimated using the UCC ratio. Spatial and temporal variation of the lithogenic components ratio may falsely influence the estimated fraction value. The PFe<sub>litho</sub> and PFe<sub>non-litho</sub> proxies are interesting tools to evaluate the importance of lithogenic and non-lithogenic (either biogenic or authigenic), but have to be used with consideration.

Overall, the estimated lithogenic contribution to PFe varies from 25% (station 60, 950 m) to 100% at stations located within the Western European Basin. Note that 100% of estimated lithogenic PFe does\_no\_t necessary mean that biogenic particles are absent; they may just be masked by the important loaddominance of lithogenic particles. Important inter-basins variations are observed along the section (Figure 6). The IAP and WEB basins are linked with high median values of the proxy % PFe<sub>litho</sub>, 90%., which is also reflected in the MW and NEADW PFe/PAI ratio which that displays a value PFe/PAI close to the crustal one (Figure 7), ou 8 dans la nouvelle version??). This could be linked to a the lateral advection of iron rich lithogenic particles sourced from the Iberian margin and to atmospheric particles-inputs (Shelley et al., 2017). This point is discussed—wit inh more detail in section 4.32.14. Then, between the stations 26 and 29, the %PFe<sub>litho</sub> proxy values dramatically decreased, and reached values underless than 55%; between the stations 26 and 29, to reach median %PFe<sub>litho</sub> value under 55% in the Iceland, Irminger and the Labrador basins. This feature is likely be-associated to—with the presence of the Sub-Arctic Front, located between 49.5 and 51°N latitude and 23.5 and 22°W longitude (Zunino et al., 2017). Indeed, this front which separates cold and fresh water of subpolar origin from warm and salty water of subtropical origin

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was clearly identifiable at station 26-by the steep gradient of the isotherms and isohalines between station 26 and 29; salinity dropping from 35.34 psu-to 35.01-psu-(Figure 2). North of of the Sub-Arctic Front, LSW and ISOW display high PFe/PAl ratios, ranging from 0.36 to 0.44 mol mol<sup>-1</sup>. These high ratios, compared to the crustal one, could be associated towith higher contentproportions of PFe from biogenic origin, especially in the case of the LSW.

# 4.23. Tracking the different inputs of particulate iron

4.23.1. Inputs at margins: Iberian, Greenland and Newfoundland

Inputs from continental shelves and margins have been demonstrated to support high productivity in shallow coastal areas. Inputs of iron from continental margin sediments supporting the high productivity found in shallow coastal regions have been demonstrated in the past (e.g. Cullen et al. (2009), Elrod et al. (2004), Jeandel et al. (2011), Ussher et al. (2007)) and sometimes, were shown to be advected at great distances from the coast (e.g. Lam and Bishopet al., 2008).

In the following section, we will investigate these possible candidate-sources in proximity of to the different margins encountered. Along the GEOVIDE section, sediments at margins were of various compositions (Dutkiewicz et al., 2015). Sediments originating from the Iberian margin were mainly constituted of silts and clays (Cacador et al., 1996; Duarte et al., 2014). East Greenland margin sediments were a mixture of sands and grey/green muds, while, sediments from the West Greenland margin were mainly composed of grey/green muds (Loring and Asmund, 1996). At the western end of the section, sediments from the Newfoundland margin were composed of gravelly and sandy muds (Mudie et al., 1984). The different sediment compositions of the three margins sampled during GEOVIDE have different mineralogy/composition, which are reflected in their different PFe/PAI ratios (Figure 8). While the Iberian Margin had a PFe/PAI close to UCC ratio, the highest ratio could be seen at the East Greenland (stations 53 and 56) and West Greenland (station 61) Margins, with median PFe/PAI reaching 0.45 mol mol<sup>-1</sup>. The Newfoundland margin displayed an intermediate behaviour, with Fe/AI ratios of 0.35 mol mol<sup>-1</sup>.

In addition to PAI, PMn can be used as a tracer of inputs from shelf resuspension (Lam and Bishop, 2008). Indeed, Mn is really sensitive to oxidation mediated by bacteria (Tebo et al., 1984; Tebo and Emerson, 1985) and forms manganese oxides (MnO<sub>2</sub>). These authigenic particles lead to an enrichment of Mn in particle compositions. In order to track the influence of shelf resuspension, a percentage of sedimentary inputs "%bulk sediment inputs" can be estimated using PMn/PAI ratio from GEOVIDE samples and the PMn/PAI UCC value (0.0034; Taylor and McLennan, 1995) according to the following equation:

%bulk sediment PMn =  $100 * \left(\frac{PAl}{PMn}\right)$  sample \*  $\left(\frac{PMn}{PAl}\right)$  UCC ratio\_\_\_\_(3)

This proxy is a good indicator of direct and recent sediment resuspension. We assume that particles newly resuspended in water column will have the same PMn/PAI ratio than the UCC ratio leading to a "bulk sediment Mn" of 100%. This value will decrease by authigenic formation of Mn oxides. This proxy assumes homogeneity of the sediment PMn/PAI ratio through the section which is maybe not completely the case at every station. In

consequence, this proxy is only a tool to identify new benthic resuspension at specific location and intercomparison between several locations is not possible. When a sample presents a "%bulk sediment Mn" greater than 100%, we assign a value of 100% to simplify the following discussion. As the Mn cycle can also be affected by biologic uptake (e.g. Peers and Price, 2004; Sunda and Huntsman, 1983), this proxy is only used at depths where biologic activity is negligible (i.e. below 150m depth). The Iberian margin The Iberian margin was an important source of lithogenic-derived iron-rich particles-in to the Atlantic Ocean during GEOVIDE; shelf resuspension impacts wereas perceptible up tontil 280 km away from the margin (Station 11) in the Iberian Abyssal Plain (Figure 8). On the shelf, at station 2, high sediment resuspension was observable by resulted in the low beam transmissometry value (87.6%) at the immediate vicinity of the seafloor (153 m). This sediment resuspension led to an extensive input of lithogenic particles within the water column associated with high concentrations of PFe (304 nmol L-1), PAI (1500 nmol L<sup>-1</sup>), and PMn (2.5 nmol L<sup>-1</sup>) (Figure 3, Table S1). Moreover, one hundred percent 100% of PFe iswas estimated to have a lithogenic origin (Figure 810) while 100% of the PMn was estimated to be the result of a recent sediment resuspension according to the %Fejitho and "%bulk sediment Mn" proxies (supplementary material, Table S2Figure 8), confirming the resuspended particle input. Coastal waters of the Iberian Shelf are impacted by the runoff for the Tagus River, which is characterised by high suspended matter discharges, ranging between 0.4 to 1 × 106 tons yr 1, and with a high anthropogenic signature (Jouanneau et al., 1998). During the GEOVIDE section, the freshwater input was observable at stations 1, 2 and 4 in the first 20 m; salinity was below 35.2 psu while surrounding waters masses had salinity up to 35.7 psu. Within the freshwater plume, particulate concentrations were important at station 2 with PFe of 1.83 nmol L<sup>-1</sup>-Further away from the coast, the particulate concentrations remained low at 20m depth, with PFe, PAI, and PMn concentrations of 0.77 nmol L<sup>-1</sup>, 3.5 nmol L<sup>-1</sup>, and 0.04 nmol L<sup>-1</sup>, respectively at station 1. The low expansion of the Tagus plume is likely due to the rapid settling of suspended matter. Indeed, our coastal station 2 was already located at around 50 km of the Iberian coast and according to Jouanneau et al. (1998), the surface particle load can be observable at a maximum 30km of the Tagus estuary. In additionBesides, ADCP data acquired during GEOVIDE (Zunino et al., 2017) and several other studies have\* reported an intense current spreading northward coming from the Straits of Gibraltar and Mediterranean Sea, leading to a-the strong resuspension of benthic sediments above the Iberian Shelf, e.g. Biscaye and Eittreim (1977), Eittreim et al. (1976), McCave and Hall (2002), Spinrad et al. (1983). The importance of the sediment resuspension was observable by low beam transmissometry value (87.6%) at the bottom of station 2. This important sediment resuspension led to an extensive input of lithogenic particles within the water column associated with high concentrations of PFe (304 nmol L<sup>-1</sup>), PAI (1500 nmol L<sup>-1</sup>), and PMn (2.5 nmol L<sup>-1</sup>) (Figure

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Mn" proxies (supplementary material, Table S2), confirming the resuspended particle input.

Moreover, one hundred percent of PFe is estimated to have a lithogenic origin (Figure 10) while 100% of the PMn

was estimated to be the result of a recent sediment resuspension according to the %Feinle and "%bulk sediment

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At distance from the shelf, within the Iberian Abyssal Plain, an important lateral advection of PFe from the margin\* was observable (Figure 108). These lateral inputs occurred at two depth ranges: between 400 and 1000 m as seen at stations 4 and 1, with PFe concentrations reaching 4 nmol L<sup>-1</sup>, and between 2500 m and the bottom (3575 m) of station 1, with PFe concentrations reaching 3.5 nmol L<sup>-1</sup>. While 100% of PFe had a lithogenic signature, the sedimentary source input estimation decreased, between 40% and 90% of the PMn (Figure 108). Transport of lithogenic particles was observable until station 11 (12.2°W) at 2500 m where PFe concentration was 7.74 nmol L<sup>-1</sup> and 60% of PMn had a sedimentary origin (Figure 49). It is nNoteworthy that, no particular increase in PFe, PMn or PAl was seen observed between 500 and 2000 m depth, where the MOW spreads. This, which is consistent with the that was observed DFe concentrations (Tonnard et al., this issue), by yet in contrast with to the dissolved aluminium (DAI) concentrations values (Menzel\_Barraqueta et al, subm. 2018, this issue) which were high in the MOW<sub>2</sub> and with the study of Ohnemus and Lam (2015) that reported a maximum PFe concentration at 695 m depth associated with the particle-rich Mediterranean Overflow Water (Eittreim et al., 1976) in the IAP. However, their station was located further south of our station 1. The shallower inputs observed at stations 1 and 4 could therefore be attributed to sediment resuspension from the Iberian margin and nepheloid layer at depth for station 1.

Surface coastal waters of the Iberian Shelf are impacted by the runoff for the Tagus River, which is characterised by high suspended matter discharges, ranging between 0.4 to 1 × 10<sup>6</sup> tons yr<sup>-1</sup>, and with a high anthropogenic [trace element] signature (Jouanneau et al., 1998). During the GEOVIDE section, the freshwater input was observable at stations 1, 2 and 4 in the first 20 m; salinity was below 35.2 psu while surrounding waters masses had salinity up to 35.7 psu. Within the freshwater plume, particulate concentrations were high at station 2 with PFe of 1.83 nmol L<sup>-1</sup>. Further away from the coast, the particulate concentrations remained low at 20m depth, with PFe, PAI, and PMn concentrations of 0.77 nmol L<sup>-1</sup>, 3.5 nmol L<sup>-1</sup>, and 0.04 nmol L<sup>-1</sup>, respectively at station 1. The low expansion of the Tagus plume is likely due to the rapid settling of suspended matter. Indeed, our coastal station 2 was already-located at aroundapproximately 50 km effrom the Iberian coast, whereas the surface particle load can only be observed at a maximum distance of 30 km from the Tagus estuaryand according to (Jouanneau et al., (1998), the surface particle load can be observable at a maximum 30km of the Tagus estuary. Overall, the Iberian margin appears to be an important source of lithogenic-derived iron rich particles in the Atlantic Ocean. Therefore, the Iberian margin appears to be an important source of lithogenic derived iron rich particles in the Atlantic Ocean; shelf resuspension impact was perceptible until 280 km away from the margin (Station 11) in the Iberian Abyssal Plain.

# South Greenland

Several studies already demonstrated the importance of icebergs and sea ice as source of dissolved and particulate iron (e.g. van der Merwe et al., 2011a, 2011b; Planquette et al., 2011; Raiswell et al., 2008). The Greenland shelf is highly affected by external fresh water inputs as ice melting or riverine runoff (Fragoso et al., 2016), that are important sources of iron to the Greenland Shelf (Bhatia et al., 2013; Hawkings et al., 2014; Statham et al., 2008). Along During GEOVIDE, the Greenland shelves were a source of particulate-rich meteoric water leading to a transfer of DFe to PFe by an-enhanced biological activity. Indeed,

Bothboth East and West Greenland shelves (stations 53 and 601) had high concentration of particles (beam transmissometry of 83%) and particulate trace elements, reaching 22.1 nmol L-1 and 18.7 nmol L-1 of PFe, respectively (station 53 at 100m and station 61 at 136 m). Several studies have already demonstrated the importance of icebergs and sea ice melting as sources of dissolved and particulate iron (e.g. van der Merwe et al., 2011a, 2011b; Planquette et al., 2011; Raiswell et al., 2008). The Greenland shelf is highly affected influenced by external fresh water inputs such as sea-ice-melting or riverine runoff (Fragoso et al., 2016), that which are important sources of iron to the Greenland Shelf (Bhatia et al., 2013; Hawkings et al., 2014; Statham et al., 2008). During the cruise, the relative freshwater observed (S\_<.33 psu) within the first 25 meters of stations 53 and 61 wasere associated with high PFe (19 nmol L-1), PAI (61 nmol L-1), PMn (0.6 nmol L-1) and a-low beam transmissometry ( $\leq$  85%) (Figure 94 and Table S1). The associated pParticles associated were enriched in iron compared to aluminium, as PFe/PAI ratio was 0.3 within the meteoric water plume. High biological production, in agreement with-The high PP concentrations (reaching 197 nmol L-1)-resulting from high biological production, induced by the supply of bioavailable dissolved iron from meteoric water (Raiswell et al., 2008; Statham et al., 2008;, Tonnard et al., submitted, this issue), led to a transfer of DFe to the particulate phase. This is in line with the factfinding that around 30% PFe had a non-lithogenic origin. In addition, only 40% PMn originated from resuspended sediments. Interestingly, these two proxies remained constant from the seafloor to the surface (Station 49, Figure 108), with around 25% of the PMn of sedimentary origin, which could be due to an important mixing happening occurring on the shelf. The lithogenic PFe could result from the release of PFe from Greenland bedrock captured during the ice sheet formation on land-. The spatial extent of the off-shelf lateral transport of particles was not important on the east Greenland coast. Indeed, no visible increase of particulate trace metal concentrations was visible at the first station off-shelf, station 60 (Figure 108), except at 1000 m depth, where a strong increase (up to 90%) of sedimentary PMn was seen. This is probably due to the East Greenland Coastal Current (EGCC) that was located at station 53 whice constrained these inputs while stations 56 and 60 were under the influence of another strong current, the East Greenland-Irminger current (EGIC) (Zunino et al., 2017). To the west of the Greenland margin, lateral transport of particles was slightly more important. Noticeable concentrations of particulate lithogenic elements were observable until station 64 located 125 km away from shoreline. These particles had a decreaseding PFe lithogenic contribution (50%) with a similar (25%) sedimentary PMn content than closer to the margin. The increasing nature of non-lithogenic PFe is linked to the bloom in surface waters (associated with a PFe/PAI ratio of 0.30 mol mol<sup>-1</sup>, a PP of 197 nmol L<sup>-1</sup> at station 61 and a Chl a concentrations of 6.21 mgChl-a concentration of 6.21 mg m<sup>-3</sup> at station 61), with the gravitational settling of biogenic PFe settling down along the transport of particles. Therefore, particles newly resuspended from Greenland sediments are an important source, representing around onea third of the pMn pool, combined with surface inputs such as riverine runoff and/or ice-melting that are delivering particles on the shelf, and also biological production. Unlike the Iberian shelf, the Greenland margin was not an important provider of particulate metals inside the Irminger and Labrador Basins, due to the circulation that constrained the extent of the margin plume.

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Therefore, particles newly resuspended from Greenland sediments are an important source, representing around

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particles on the shelf and biological production. Unlike the Iberian shelf, Greenland margin was not an important provider of particulate metals inside the Irminger and Labrador Basin, due to the circulation that constrained the extent of the margin plume.

544 The Newfoundland Shelf

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Previous studies <u>have</u> already described the influence of fresh water on the Newfoundland shelf from the Hudson Strait and/or Canadian Artic Archipelago (Fragoso et al., 2016; Yashayaev, 2007). Yashayaev (2007) also monitored strong resuspension of sediments associated with the spreading of <u>the</u> Labrador Current along the West Labrador margin.

Close to the Newfoundland coastline, at station 78, high fresh water discharge (≤ 32 psu) was observed in surface waters (Benetti et al., 2017). Interestingly, these freshwater signatures were not associated with elevated particulate trace metal concentrations. Distance of meteoric water sources implied a long travel time for the water to spread through the Labrador Basin to our sampling stations. Along the journey, particles present originally may have been removed from the water column by gravitational settling.

The proportion of lithogenic PFe was relatively high and constant in the entirethroughout the water column, with a median value of 70%. At station 78, 100% of the PMn had a sedimentary origin close to the seafloor (371 m). The spreading of the recent sediment resuspension was observable until 140 m depth where the contribution of sedimentary Mn was still 51% (Figure 108, Table S2). This could correspond to an intense nepheloid layer as previously reported by Biscaye and Eittreim (1977) (see also section 3.3.2). The high PFe concentration (184 nmol L<sup>-1</sup>, station 78, 371 m) associated with a high percentage of sedimentary PMn (95%) observed at the bottom of this station, was therefore the result of an important resuspension of shelf sediments. This was confirmed with low transmissometry values of 95%.

The important phytoplanktonic community present (maximum Chl-a= 4.91 mg m<sup>-3</sup>, Tonnard et al., in prep), is linked to a low PFe cocnetration of 0.79 nmol L<sup>-1</sup> at 10 m, but, with a high PFe/PAl ratio, up to 0.4, and PP concentration of 97 nmol L<sup>-1</sup>, confirming the biological influence. Either the biogenic particles settled quickly, and/or they were quickly remineralized. Concerning this latter process, intense remineralization at station 77 (7 mmol C m<sup>-2</sup> d<sup>-1</sup> compared to 4 mmol C m<sup>-2</sup> d<sup>-1</sup> in the Western European Basin) has been reported by Lemaitre et al. (2018a and 2018b), which could explain the low PFe values throughout the water column.

Along the GEOVIDE section, continental shelves provided an important load of particles within the surrounding water column. The three margins sampled during GEOVIDE behaved differently; the Iberian margin discharged high quantities of lithogenic particles far away from the coast while the Greenland and Newfoundland margins did not reveal important PFe concentrations. Spreading of particles is tightly linked to hydrodynamic conditions, which in the case of the Greenland margin, prevented long distance seeding of PFe. Moreover, each margin showed a specific PFe/PAI ratio (Figure 8) indicating different composition of the resuspended particles. Resuspended particles represent the composition of sediment at the margin if oxido reductive transformation of iron and aluminium are considered negligible under these circumstances. Differences between margins were due to the presence of non-crustal particles, either biogenic or authigenic. Biological production in surface waters and authigenic formation of iron hydroxide produce particles with a higher PFe/PAI content and their export through

the water column to the sediment increased the PFe/PAI ratio at depth. Regions where biological production is intense such as in the vicinity of Newfoundland presented higher PFe/PAI ratios of resuspended benthic particles. Along the GEOVIDE section, continental shelves provided an important load of particles withinto the surrounding water column. The three margins sampled during GEOVIDE behaved very differently; the Iberian margin discharged high quantities of lithogenic particles far away from the coast while the Greenland and Newfoundland margins did not reveal important PFe concentrations. Spreading of particles is tightly linked to hydrodynamic conditions, which in the case of the Greenland margin, prevented long distance seeding of PFe. Moreover, each margin showed a specific PFe/PAI ratio (Figure 98) indicating different composition of the resuspended particles. Resuspended particles represent the composition of sediment at the margin if oxido-reductive redox transformation of iron and aluminium are considered negligible under these circumstances. Differences between margins were due to the presence of non-crustal particles, either biogenic or authigenic. Biological production in surface waters and authigenic formation of iron hydroxide produced particles with a higher PFe/PAI content and their export through the water column to the sediment increased the PFe/PAI ratio at depth. Regions where biological production is intense such as in the vicinity of Newfoundland presented higher PFe/PAI ratios of resuspended benthic particles.

### 4.23.2 Benthic resuspended sediments

Along the GEOVIDE section, Benthic nepheloid layers (BNLs) BNLs are provideding high concentrations of particulate trace elements to in the deep open ocean, contributing highlysignificantly to the total trace elements budget of as iron, Along the GA01 section, BNLs were observable in each province, although they had different intensities with different strengths (Figures 3 and 120).

Benthic nepheloid layers (BNLs) are important layers where local resuspension of sedimentary particles (Bishop and Biseaye, 1982; Eittreim et al., 1976; Rutgers Van Der Loeff et al., 2002) occur due to strong hydrographic stresses (i.e. boundary currents, benthic storms and deep eddies) interacting with the ocean floor (Biseaye and Eittreim, 1977; Eittreim et al., 1976; Gardner et al., 2017, 2018). Along the GA01 section, BNLs were observable in each province with different strengths (Figures 3 and 12).

In BNLs located within the WEB, PFe concentrations reached up to 10 nmol L-1 (stations 26 and 29, Table S1). These concentrations were smaller than PFe concentrations encountered in BNL from the Icelandic (stations 32 and 34), Irminger (stations 42 and 44) and Labrador Basins (stations 68, 69 and 71), where benthic resuspension led to PFe concentrations higher than 40 nmol L-1, even reaching 89 nmol L-1 at the bottom of station 71 (3736 m). Moreover, in the Irminger and Labrador Basins, PFe/PAl molar ratios within BNLs were higher than the ones measured within the WEB at station 26 and 29. In the Irminger Basin, PFe/PAl reached 0.4 mol mol-1 (Figure 101), which could reveal a mixture of lithogenic and biogenic matter that had been previously exported. This feature was also observed in the Labrador Basin, with PFe/PAl ratio ranging between 0.34 and 0.44 mol mol-1. In contrast, BNLs sampled in the WEB have-clearly have a lithogenic imprint, with PFe/PAl molar ratios close to the crustal one. Resuspended sediments with a non-crustal contribution seem to holdhave a higher PFe contents than sediments with a-lithogenic characteristics. Nevertheless, interestingly all BNLs present during GEOVIDE were spreading identically, with impacts observable up to 200 meters above the oceanic seafloor (Figure 101), as reflected in beam transmissometry values, and PFe concentrations, whichthat returned to a-background levels at 200 m above the seafloor. The presence of these BNLs has also been reported by Le Roy et al. (submitted, this

issue) using radium-226 activity. Important differences of PFe intensities could also be due to different hydrographic components and topographic characteristics. BNLs are occurring due to strong hydrographic stresses (i.e. boundary currents, benthic storms and deep eddies) interacting with the ocean floor (Biscaye and Eittreim, 1977; Eittreim et al., 1976; Gardner et al., 2017, 2018). They are. As previously explained, two main triggers of BNLs are benthic storms and deep eddies; by definition, these processes are highly variable geographically and temporally, but we have no physical data equid-whichwould allow us to investigate further this hypothesis further.

this hypothesis <u>further</u>.

<u>Benthic nepheloid layers (BNLs) are important layers where local resuspension of sedimentary particles (Bishop and Biscaye, 1982; Eittreim et al., 1976; Rutgers Van Der Loeff et al., 2002) occur due to strong hydrographic</u>

stresses (i.e. boundary currents, benthic storms and deep eddies) interacting with the ocean floor (Biscaye and

Eittreim, 1977; Eittreim et al., 1976; Gardner et al., 2017, 2018). Along the GA01 section, BNLs were observable in each province with different strengths (Figures 3 and 12).

Along the GEOVIDE section, BNLs are providing high concentrations of particulate trace element in the deep open ocean, contributing highly to the total trace elements budget as iron.

### 4.23.3. Reykjanes Ridge inputs

Above the Reykjanes ridgeRidge, high PFe concentrations were determinedmeasured, reaching 16 nmol L<sup>-1</sup> just above the seafloor, while increased DFe concentrations were reported to the East-cast of the ridge (Tonnard et al., this issue). The exact sources of iron-rich particles cannot be well constrained, as they could come from active hydrothermal vents or resuspension of particulate matter from new crustal matter produced at the ridge. According to the oceanic circulation (Zunino et al., 2017; Garcia-Ibanez et al., 2017), hydrothermal particles could have been seen in the ISOW within the Icelandic Basin. Nevertheless, at the vicinity of the ridge, scanning electron microscope (SEM) analyses of our samples did reveal a number of biological debris and clays but not the presence of iron (oxy-)hydroxide particles (supplementary figure S1), which are known to be highly produced close to hydrothermal vents (Elderfield and Schultz, 1996). Their absence could thus indicate an absence of vents. However, data from other proxies, such as helium-3, are would be necessary to elaim-confirm with more accuracy the presence or absence of an hydrothermal source close to station 38.

# 4.32.4. Atmospheric inputs

Atmospheric deposition is an important input of trace elements in surface of the open ocean (e.g. (Jickells et al., 2005). Atmospheric inputs, both wet and dry, were reported to be low during the GEOVIDE cruise (Menzel-Bbarraqueta et al., 2018, this issue; Shelley et al., 2017; 2018). In fact, oceanic particles measurements in surface waters along the section did not reveal high PFe or PAl eoneentrations, concentrations. therefore, the surface composition of particles did not seem to be highly affected by atmospheric deposition at the time of the cruise. One pattern is also interesting to note: the surface waters of the Iberian Abyssal Plain and Western European Basin, between stations 11 and 23 presented a characteristic feature with really low PFe/PAl elemental ratios, of 0.11, smaller than the UCC ratio of 0.21 (Figure 6). Such low ratios have been reported in the same region by Barrett et al. (2012). One possible explanation is given by Buck et al. (2010) who described Fe-depleted aerosols in this area of the North Atlantic with PFe/PAl ratio below UCC ratio. However, Shelley et al. (2017) found a

higher PFe/PAl ratio around 0.25 is this area (their samples geoa5-6). This result, highlights some of the difficulties that in linking atmospheric inputs to water column data (Baker et al., 2016), and implies a probable fractionation after aerosol deposition. In addition, there is high spatial and temporal variability of atmospheric deposition (Mahowald et al., 2005) and a certain degree of uncertainty about the dissolution processes of atmospherically-transported particles (Bonnet and Guieu, 2004).

### 5. Conclusions

The investigation of the PFe compositions of suspended particulate matter along the GEOVIDE section in the North Atlantic reflects the pervasive influence of crustal particles, augmented by sedimentary inputs at margins, and within benthic nepheloid layers at depths. In consequence, variance of particulate iron along the section is mainly explained by lithogenic factors.

Resuspension of sedimentary particles from continental shelves are responsible of high particulate iron concentrations within the surrounding water column, and could be observed at long distances, in the case of the Iberian margin. Due to the hydrodynamics conditions, lithogenic particles are exported off shore up to 280 km away off the Iberian margin. Our results also demonstrate the impact of Aarctic meteoric water in the biogeochemical cycle of trace elements on the Greenland shelf, while in surface waters, the enhancement of productivity by new bioavailable iron is leading to a transfer of dissolved iron to the particulate phase.

Above the Reykjanes Ridge, resuspension of particles were responsible offor the PFe enrichment of the Iceland Scottish Overflow Water.

Our dataset allowsed the investigation of scavenging processes that were sometimes visible at depths greater than

1000 m, these effects being the most pronounced within the WEB.

Overall, PFeparticulate-iron distributions in the North Atlantic isare strongly affectinfluenced by sources at its boundaries (i.e. continental margins and seafloor). When combined with other datasets from the GEOTRACES program in a modelling study, for example, use of ttThis work, within the frame of the GEOTRACES program, data will will-facilitate a greaterallow a better understanding of particulate iron the eyele of cycling in the particulate ironNorth Atlantic, when combined to other datasets in a modelling exercise for example.

This investigation of the PFe compositions of suspended particulate matter along the GEOVIDE section in the North Atlantic indicates reflects the pervasive influence of crustal particles, augmented by sedimentary inputs at margins, and at depths, within benthic nepehloid layers. Rary particles are responsible of high concentration In particular, hallowed the exportof—while in surface, twill II faudrait une phrase d'ouverture plutôt

Indeed, along the GEOVIDE section, continental shelves provided an important load of particles within the surrounding water column, with PFe mostly residing in non-biogenic particulate form. The Iberian margin discharged high quantities of lithogenic particles originating from riverine inputs far away from the coast while the Greenland margin did not reveal a long distance seeding of PFe, due to hydrodynamic conditions. Both Greenland and Newfoundland margins PFe resuspended particles were under a strong biogenic influence that were exported at depth. This resulted in different remineralisation fluxes among the different provinces. Scavenging processes could also be visible at depths greater than 1000 m, these effects being the most pronounced

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700 Overflow Water. A similar feature occurs for the Labrador Sea Water, as it flows from the Irminger Basin to the 701 Western European Basin. 702 703 704 705 Acknowledgments 706 We are greatly indebted to the captain and crew of the N/O Pourquoi Pas? for their help during the GEOVIDE 707 mission and clean rosette deployment. We would like to give special thanks to Fabien Pérault and Emmanuel de 708 Saint Léger for their technical expertise, to Catherine Schmechtig for the GEOVIDE database management and 709 Greg Cutter for his guidance in setting up the new French clean sampling system. We also would like to thanks 710 Reiner Schlitzer for the Ocean Data View software (ODV). 711 This work was supported by the French National Research Agency (ANR-13-BS06-0014, ANR-12-PDOC-0025-712 01), the French National Centre for Scientific Research (CNRS-LEFE-CYBER), the LabexMER (ANR-10-713 LABX-19), and Ifremer. It was supported for the logistic by DT-INSU and GENAVIR. 714 715 References 716 717 Aguilar-Islas, A. M., Rember, R., Nishino, S., Kikuchi, T. and Itoh, M.: Partitioning and lateral transport of iron to the Canada Basin, Polar Sci., 7(2), 82-99, doi:10.1016/j.polar.2012.11.001, 2013. 718 719 Baker, A. R., Adams, C., Bell, T. G., Jickells, T. D. and Ganzeveld, L.: Estimation of atmospheric nutrient inputs 720 to the Atlantic Ocean from 50°N to 50°S based on large-scale field sampling: Iron and other dust-associated elements, Global Biogeochem. Cycles, 27(3), 755-767, doi:10.1002/gbc.20062, 2013. 721 722 Baker, A. R., Landing, W. M., Bucciarelli, E., Cheize, M., Fietz, S., Hayes, C. T., Kadko, D., Morton, P. L., 723 Rogan, N., Sarthou, G., Shelley, R. U., Shi, Z., Shiller, A. and van Hulten, M. M. P.: Trace element and isotope 724 deposition across the air-sea interface: progress and research needs, Philos. Trans. R. Soc. A Math. Phys. Eng. 725 Sci., 374(2081), 20160190, doi:10.1098/rsta.2016.0190, 2016. Barrett, P. M., Resing, J. A., Buck, N. J., Buck, C. S., Landing, W. M. and Measures, C. I.: The trace element 726 727 composition of suspended particulate matter in the upper 1000m of the eastern North Atlantic Ocean: A16N, Mar. 728 Chem., 142-144, 41-53, doi:10.1016/j.marchem.2012.07.006, 2012. 729 Benetti, M., Reverdin, G., Lique, C., Yashayaev, I., Holliday, N. P., Tynan, E., Torres-Valdes, S., Lherminier, P., 730 Tréguer, P., and Sarthou, G.: Composition of freshwater in the spring of 2014 on the southern Labrador shelf and 731 slope, Journal of Geophysical Research: Oceans, 122, 1102-1121, 10.1002/2016jc012244, 2017.

Finally, resuspended sediments above the Reykjanes Ridge increased the PFe composition of the Iceland Scottish

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within the Labrador Basin.

- 733 Berger, C. J. M., Lippiatt, S. M., Lawrence, M. G. and Bruland, K. W.: Application of a chemical leach technique
- 734 for estimating labile particulate aluminum, iron, and manganese in the Columbia River plume and coastal waters
- 735 off Oregon and Washington, J. Geophys. Res., 113, C00B01, doi:10.1029/2007JC004703, 2008.
- 736 Bergquist, B. A., Wu, J. and Boyle, E. A.: Variability in oceanic dissolved iron is dominated by the colloidal
- 737 fraction, Geochim. Cosmochim. Acta, 71(12), 2960–2974, doi:10.1016/j.gca.2007.03.013, 2007.
- 738 Bhatia, M. P., Kujawinski, E. B., Das, S. B., Breier, C. F., Henderson, P. B. and Charette, M. A.: Greenland
- 739 meltwater as a significant and potentially bioavailable source of iron to the ocean, Nat. Geosci., 6(4), 274-278,
- 740 doi:10.1038/ngeo1746, 2013.
- 741 Biscaye, P. E. and Eittreim, S. L.: Suspended Particulate Loads and Transports in the Nepheloid Layer of the
- 742 Abyssal Atlantic Ocean, Dev. Sedimentol., 23(C), 155–172, doi:10.1016/S0070-4571(08)70556-9, 1977.
- 743 Bishop, J. K. B. and Biscaye, P. E.: Chemical characterization of individual particles from the nepheloid layer in
- 744 the Atlantic Ocean, Earth Planet. Sci. Lett., 58(2), 265–275, doi:10.1016/0012-821X(82)90199-6, 1982.
- 745 Bishop, J. K. B. and Fleisher, M. Q.: Particulate manganese dynamics in Gulf Stream warm-core rings and
- surrounding waters of the N.W. Atlantic, Geochim. Cosmochim. Acta, 51(10), 2807-2825, doi:10.1016/0016-
- 747 7037(87)90160-8, 1987.
- Bonnet, S. and Guieu C.: Dissolution of atmospheric iron in seawater, Geophys. Res. Lett., 31(3), L03303,
- 749 doi:10.1029/2003GL018423, 2004.
- 750 Boyle, E. A., Bergquist, B. A., Kayser, R. A. and Mahowald, N.: Iron, manganese, and lead at Hawaii Ocean
- 751 Time-series station ALOHA: Temporal variability and an intermediate water hydrothermal plume, Geochim.
- 752 Cosmochim. Acta, 69(4), 933–952, doi:10.1016/j.gca.2004.07.034, 2005.
- 753 Buck, C. S., Landing, W. M., Resing, J. A. and Measures, C. I.: The solubility and deposition of aerosol Fe and
- other trace elements in the North Atlantic Ocean: Observations from the A16N CLIVAR/CO2repeat hydrography
- 755 section, Mar. Chem., 120(1–4), 57–70, doi:10.1016/j.marchem.2008.08.003, 2010.
- 756 Cacador, I., Vale, C. and Catarino, F.: The influence of plants on concentration and fractionation of Zn, Pb, and
- 757 Cu in salt marsh sediments (Tagus Estuary, Portugal), J. Aquat. Ecosyst. Heal., 5(3), 193-198,
- 758 doi:10.1007/BF00124106, 1996. Cheize, M., Planquette, H. F., Fitzsimmons, J. N., Pelleter, E., Sherrell, R. M.,
- 759 Lambert, C., Bucciarelli, E., Sarthou, G., Le Goff, M., Liorzou, C., Chéron, S., Viollier, E., and Gayet, N.:
- 760 <u>Contribution of resuspended sedimentary particles to dissolved iron and manganese in the ocean: An experimental</u>
- 761 <u>study, Chemical Geology. doi: 10.1016/j.chemgeo.2018.10.003, 2018.</u>
- 762 Collier, R. and Edmond, J.: The trace element geochemistry of marine biogenic particulate matter, Prog.
- 763 Oceanogr., 13(2), 113–199, doi:10.1016/0079-6611(84)90008-9, 1984.
- 764 Cullen, J. T., Chong, M. and Ianson, D.: British columbian continental shelf as a source of dissolved iron to the
- subarctic northeast Pacific Ocean, Global Biogeochem. Cycles, 23(4), 1–12, doi:10.1029/2008GB003326, 2009.

- 766 Cutter, G. A. and Bruland, K. W.: Rapid and noncontaminating sampling system for trace elements in global
- 767 ocean surveys, Limnol. Oceanogr. Methods, 10(JUNE), 425–436, doi:10.4319/lom.2012.10.425, 2012.
- 768 Dammshäuser, A., Wagener, T., Garbe-Schönberg, D. and Croot, P.: Particulate and dissolved aluminum and
- 769 titanium in the upper water column of the Atlantic Ocean, Deep. Res. Part I Oceanogr. Res. Pap., 73, 127–139,
- 770 doi:10.1016/j.dsr.2012.12.002, 2013.
- 771 Dehairs, F., Jacquet, S., Savoye, N., Van Mooy, B. A. S., Buesseler, K. O., Bishop, J. K. B., Lamborg, C. H.,
- 772 Elskens, M., Baeyens, W., Boyd, P. W., Casciotti, K. L. and Monnin, C.: Barium in twilight zone suspended
- 773 matter as a potential proxy for particulate organic carbon remineralization: Results for the North Pacific, Deep.
- 774 Res. Part II Top. Stud. Oceanogr., 55(14–15), 1673–1683, doi:10.1016/j.dsr2.2008.04.020, 2008.
- 775 Duarte, B., Silva, G., Costa, J. L., Medeiros, J. P., Azeda, C., Sá, E., Metelo, I., Costa, M. J. and Caçador, I.:
- 776 Heavy metal distribution and partitioning in the vicinity of the discharge areas of Lisbon drainage basins (Tagus
- 777 Estuary, Portugal), J. Sea Res., 93(February), 101 111, doi:10.1016/j.seares.2014.01.003, 2014.
- 778 Dutay, J. C., Tagliabue, A., Kriest, I. and van Hulten, M. M. P.: Modelling the role of marine particle on large
- 779 scale 231Pa, 230Th, Iron and Aluminium distributions, Prog. Oceanogr., 133, 66-72,
- 780 doi:10.1016/j.pocean.2015.01.010, 2015.
- 781 Dutkiewicz, A., Müller, R. D., O'Callaghan, S. and Jónasson, H.: Census of seafloor sediments in the world's
- 782 ocean, Geology, 43(9), 795–798, doi:10.1130/G36883.1, 2015.
- 783 Eittreim, S., Thorndike, E. M. and Sullivan, L.: Turbidity distribution in the Atlantic Ocean, Deep. Res. Oceanogr.
- 784 Abstr., 23(12), 1115–1127, doi:10.1016/0011-7471(76)90888-3, 1976.
- 785 Elderfield, H. and Schultz, A.: Mid-Ocean Ridge Hydrothermal Fluxes and the Chemical Composition of the
- 786 Ocean, Annu. Rev. Earth Planet. Sci., 24(1), 191–224, doi:10.1146/annurev.earth.24.1.191, 1996.
- 787 Ellwood, M. J., Nodder, S. D., King, A. L., Hutchins, D. A., Wilhelm, S. W. and Boyd, P. W.: Pelagic iron cycling
- 788 during the subtropical spring bloom, east of New Zealand, Mar. Chem., 160, 18–33,
- 789 doi:10.1016/j.marchem.2014.01.004, 2014.
- 790 Elrod, V. A., Berelson, W. M., Coale, K. H. and Johnson, K. S.: The flux of iron from continental shelf sediments:
- 791 A missing source for global budgets, Geophys. Res. Lett., 31(12), 2–5, doi:10.1029/2004GL020216, 2004.
- 792 Fitzwater, S. E., Johnson, K. S., Gordon, R. M., Coale, K. H. and Smith, W. O.: Trace metal concentrations in
- 793 athe Ross Sea and their relationship with nutrients and phytoplankton growth, Deep. Res. Part II Top. Stud.
- 794 Oceanogr., 47(15–16), 3159–3179, doi:10.1016/S0967-0645(00)00063-1, 2000.
- 795 Fragoso, G. M., Poulton, A. J., Yashayaev, I. M., Head, E. J. H., Stinchcombe, M. C. and Purdie, D. A.:
- 796 Biogeographical patterns and environmental controls of phytoplankton communities from contrasting
- 797 hydrographical zones of the Labrador Sea, Prog. Oceanogr., 141, 212-226, doi:10.1016/j.pocean.2015.12.007,
- 798 2016.

- 799 Frew, R. D., Hutchins, D. A., Nodder, S., Sanudo-Wilhelmy, S., Tovar-Sanchez, A., Leblanc, K., Hare, C. E. and
- 800 Boyd, P. W.: Particulate iron dynamics during FeCycle in subantarctic waters southeast of New Zealand, Global
- 801 Biogeochem. Cycles, 20(1), 1–15, doi:10.1029/2005GB002558, 2006.
- 802 García-Ibáñez, M. I., Pardo, P. C., Carracedo, L. I., Mercier, H., Lherminier, P., Ríos, A. F. and Pérez, F. F.:
- 803 Structure, transports and transformations of the water masses in the Atlantic Subpolar Gyre, Prog. Oceanogr., 135,
- 804 18–36, doi:10.1016/j.pocean.2015.03.009, 2015.
- 805 Gardner, W. D., Tucholke, B. E., Richardson, M. J. and Biscaye, P. E.: Benthic storms, nepheloid layers, and
- 806 linkage with upper ocean dynamics in the western North Atlantic, Mar. Geol., 385, 304-327,
- 807 doi:10.1016/j.margeo.2016.12.012, 2017.
- 808 Gardner, W. D., Richardson, M. J. and Mishonov, A. V.: Global assessment of benthic nepheloid layers and
- linkage with upper ocean dynamics, Earth Planet. Sci. Lett., 482, 126–134, doi:10.1016/j.epsl.2017.11.008, 2018.
- 810 Gerringa, L. J. A., Rijkenberg, M. J. A., Schoemann, V., Laan, P. and de Baar, H. J. W.: Organic complexation
- 811 of iron in the West Atlantic Ocean, Mar. Chem., 177, 434–446, doi:10.1016/j.marchem.2015.04.007, 2015.
- Hawkings, J. R., Wadham, J. L., Tranter, M., Raiswell, R., Benning, L. G., Statham, P. J., Tedstone, A., Nienow,
- 813 P., Lee, K. and Telling, J.: Ice sheets as a significant source of highly reactive nanoparticulate iron to the oceans,
- 814 Nat. Commun., 5(May), 1–8, doi:10.1038/ncomms4929, 2014.
- 815 Hwang, J., Druffel, E. R. M. and Eglinton, T. I.: Widespread influence of resuspended sediments on oceanic
- 816 particulate organic carbon: Insights from radiocarbon and aluminum contents in sinking particles, Global
- 817 Biogeochem. Cycles, 24(4), 1–10, doi:10.1029/2010GB003802, 2010.
- 818 Jeandel, C. and Oelkers, E. H.: The influence of terrigenous particulate material dissolution on ocean chemistry
- and global element cycles, Chem. Geol., 395, 50–66, doi:10.1016/j.chemgeo.2014.12.001, 2015.
- 820 Jeandel, C., Peucker-Ehrenbrink, B., Jones, M. T., Pearce, C. R., Oelkers, E. H., Godderis, Y., Lacan, F., Aumont,
- 821 O. and Arsouze, T.: Ocean margins: The missing term in oceanic element budgets?, Eos, Transactions American
- 822 Geophysical Union, 92(26), 217–224, doi: 10.1029/2011EO260001, 2011.
- 323 Jickells, T. D., An, Z. S., Andersen, K. K., Baker, A. R., Bergametti, C., Brooks, N., Cao, J. J., Boyd, P. W., Duce,
- 824 R. A., Hunter, K. A., Kawahata, H., Kubilay, N., LaRoche, J., Liss, P. S., Mahowald, N., Prospero, J. M., Ridgwell,
- 825 A. J., Tegen, I. and Torres, R.: Global iron connections between desert dust, ocean biogeochemistry, and climate,
- 826 Science (80-. )., 308(5718), 67–71, doi:10.1126/science.1105959, 2005.
- Jouanneau, J. M., Garcia, C., Oliveira, A., Rodrigues, A., Dias, J. A. and Weber, O.: Dispersal and deposition of
- 828 suspended sediment on the shelf off the Tagus and Sado estuaries, S.W. Portugal, Prog. Oceanogr., 42(1-4), 233-
- 829 257, doi:10.1016/S0079-6611(98)00036-6, 1998.
- 830 Labatut, M., Lacan, F., Pradoux, C., Chmeleff, J., Radic, A., Murray, J. W., Poitrasson, F., Johansen, A. M., Thil,
- 831 F., Lacan, F., Pradoux, C., Chmeleff, J., Radic, A., Murray, J. W., Poitrasson, F., Johansen, A. M. and Thil, F.:

- 832 Iron sources and dissolved particulate interactions in the seawater of the Western Equatorial Pacific, iron isotope
- 833 perspectives., Global Biogeochem CyclesGlobal Biogeochemical Cycles, 1044–1065,
- 834 doi:10.1002/2014GB004928, 2014.
- 835 Lam, P. J. and Bishop, J. K. B.: The continental margin is a key source of iron to the HNLC North Pacific Ocean,
- 836 Geophys. Res. Lett., 35(7), 1–5, doi:10.1029/2008GL033294, 2008.
- 837 Lam, P. J., Ohnemus, D. C. and Marcus, M. A.: The speciation of marine particulate iron adjacent to active and
- 838 passive continental margins, Geochim. Cosmochim. Acta, 80, 108–124, doi:10.1016/j.gca.2011.11.044, 2012.
- 839 Lam, P. J., Ohnemus, D. C. and Auro, M. E.: Size-fractionated major particle composition and concentrations
- from the US GEOTRACES North Atlantic Zonal Transect, Deep. Res. Part II Top. Stud. Oceanogr., 116, 303-
- 841 320, doi:10.1016/j.dsr2.2014.11.020, 2015.
- 842 Lam, P. J., Lee, J. M., Heller, M. I., Mehic, S., Xiang, Y. and Bates, N. R.: Size-fractionated distributions of
- 843 suspended particle concentration and major phase composition from the U.S. GEOTRACES Eastern Pacific Zonal
- 844 Transect (GP16), Mar. Chem., (April), 0–1, doi:10.1016/j.marchem.2017.08.013, 2017.
- 845 Lannuzel, D., Bowie, A. R., van der Merwe, P. C., Townsend, A. T. and Schoemann, V.: Distribution of dissolved
- 846 and particulate metals in Antarctic sea ice, Mar. Chem., 124(1-4), 134-146, doi:10.1016/j.marchem.2011.01.004,
- 847 2011
- 848 Lannuzel, D., Van der Merwe, P. C., Townsend, A. T. and Bowie, A. R.: Size fractionation of iron, manganese
- and aluminium in Antarctic fast ice reveals a lithogenic origin and low iron solubility, Mar. Chem., 161, 47–56,
- 850 doi:10.1016/j.marchem.2014.02.006, 2014.
- 851 Lee, J. M., Heller, M. I. and Lam, P. J.: Size distribution of particulate trace elements in the U.S. GEOTRACES
- 852 Eastern Pacific Zonal Transect (GP16), Mar. Chem., 201(September 2017), 108-123,
- 853 doi:10.1016/j.marchem.2017.09.006, 2017.
- Lemaître, N., planquette, H., Planchon, F., Sarthou, G., Jacquet, S., Garcia-Ibanez, M. I., Gourain, A., Cheize, M.,
- 855 Monin, L., Andre, L., Laha, P., Terryn, H., and Dehairs, F.: Particulate barium tracing significant mesopelagic
- as a carbon remineralisation in the North Atlantic, Biogeosciences Discussions, doi:10.5194/bg-15-2289-2018, 2018a.
- 857 Lemaitre, N., Planchon, F., Planquette, H., Dehairs, F., Fonseca-Batista, D., Roukaerts, A., Deman, F., Tang, Y.,
- 858 Mariez, C., and Sarthou G.: High variability of export fluxes along the North Atlantic GEOTRACES section
- 859 GA01: Particulate organic carbon export deduced from the 234Th method, Biogeosciences—Discuss.,
- 860 doi:10.5194/bg-2018-190, 2018b.
- 861 Le Roy, E., Sanial, V., Charette, M.A., Van Beek, P., Lacan, F., Jacquet, S.H., Henderson, P.B., Souhaut, M.,
- 862 García-Ibáñez, M.I., Jeandel, C. and Pérez, F.: The 226Ra-Ba relationship in the North Atlantic during
- B63 GEOTRACES-GA01, Biogeosciences Discussions, doi:10.5194/bg-2017-478, 2017.

- 864 Loring, D. H. and Asmund, G.: Geochemical factors controlling accumulation of major and trace elements in
- 865 Greenland coastal and fjord sediments, Environ. Geol., 28(1), 2 11, doi:10.1007/s002540050072, 1996.
- 866 Mahowald, N. M., Baker, A. R., Bergametti, G., Brooks, N., Duce, R. A., Jickells, T. D., Kubilay, N., Prospero,
- 867 J. M. and Tegen, I.: Atmospheric global dust cycle and iron inputs to the ocean, Global Biogeochem. Cycles,
- 868 19(4), doi:10.1029/2004GB002402, 2005.
- Marsay, C. M., Lam, P. J., Heller, M. I., Lee, J. M. and John, S. G.: Distribution and isotopic signature of ligand-
- 870 leachable particulate iron along the GEOTRACES GP16 East Pacific Zonal Transect, Mar. Chem., (November
- 871 2016), 1–14, doi:10.1016/j.marchem.2017.07.003, 2017.
- 872 Martin, J. H., Fitzwater, S. E., Michael Gordon, R., Hunter, C. N. and Tanner, S. J.: Iron, primary production and
- 873 carbon-nitrogen flux studies during the JGOFS North Atlantic bloom experiment, Deep. Res. Part II, 40(1-2),
- 874 115–134, doi:10.1016/0967-0645(93)90009-C, 1993.
- 875 McCave, I. N. and Hall, I. R.: Turbidity of waters over the Northwest Iberian continental margin, Prog. Oceanogr.,
- 876 52(2–4), 299–313, doi:10.1016/S0079-6611(02)00012-5, 2002.
- 877 Menzel Barraqueta, J.L., Schlosser, C., Planquette, H., Gourain, A., Cheize, M., Boutorh, J., Shelley, R., Pereira
- 878 Contreira, L., Gledhill, M., Hopwood, M.J. and Lherminier, P.: Aluminium in the North Atlantic Ocean and the
- 879 Labrador Sea (GEOTRACES GA01 section): roles of continental inputs and biogenic particle removal.
- Biogeosciences <u>Discussions</u>, 1-28, doi: 10.5194/bg-2018-39, 2018.
- 881 Milne, A., Schlosser, C., Wake, B. D., Achterberg, E. P., Chance, R., Baker, A. R., Forryan, A. and Lohan, M.
- 882 C.: Particulate phases are key in controlling dissolved iron concentrations in the (sub)tropical North Atlantic,
- 883 Geophys. Res. Lett., 44(5), 2377–2387, doi:10.1002/2016GL072314, 2017.
- Mudie, P. J., Keen, C. E., Hardy, I. A. and Vilks, G.: Multivariate analysis and quantitative paleoecology of
- 885 benthic foraminifera in surface and Late Quaternary shelf sediments, northern Canada, Mar. Micropaleontol.,
- 886 8(4), 283–313, doi:10.1016/0377-8398(84)90018-5, 1984.
- 887 Nuester, J., Shema, S., Vermont, A., Fields, D. M. and Twining, B. S.: The regeneration of highly bioavailable
- 888 iron by meso- and microzooplankton, Limnol Oceanogr-, 59(4), 1399–1409, doi:10.4319/lo.2014.59.4.1399,
- 889 2014.
- 890 Oelkers, E. H., Jones, M. T., Pearce, C. R., Jeandel, C., Eiriksdottir, E. S. and Gislason, S. R.: Riverine particulate
- material dissolution in seawater and its implications for the global cycles of the elements, Geosci., 344(11-12),
- 892 646–651, doi:10.1016/j.crte.2012.08.005, 2012.
- 893 Ohnemus, D. C. and Lam, P. J.: Cycling of lithogenic marine particles in the US GEOTRACES North Atlantic
- 894 transect, Deep. Res. Part II Top. Stud. Oceanogr., 116, 283–302, doi:10.1016/j.dsr2.2014.11.019, 2015.
- 895 Peers, G. and Price, N. M.: A role for manganese in superoxide dismutases and growth of iron-deficient diatoms,
- 896 Limnol. Oceanogr., 49(5), 1774–1783, doi:10.4319/lo.2004.49.5.1774, 2004.

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- 897 Planquette, H. and Sherrell, R. M.: Sampling for particulate trace element determination using water sampling
- 898 bottles: Methodology and comparison to in situ pumps, Limnol. Oceanogr. Methods, 10(5), 367-388,
- 899 doi:10.4319/lom.2012.10.367, 2012.
- 900 Planquette, H., Fones, G. R., Statham, P. J. and Morris, P. J.: Origin of iron and aluminium in large particles (>
- 901 53 µm) in the Crozet region, Southern Ocean, Mar. Chem., 115(1-2), 31-42, doi:10.1016/j.marchem.2009.06.002,
- 902 2009
- 903 Planquette, H., Sanders, R. R., Statham, P. J., Morris, P. J. and Fones, G. R.: Fluxes of particulate iron from the
- 904 upper ocean around the Crozet Islands: A naturally iron-fertilized environment in the Southern Ocean, Global
- 905 Biogeochem. Cycles, 25(2), doi:10.1029/2010GB003789, 2011.
- 906 Planquette, H., Sherrell, R. M., Stammerjohn, S. and Field, M. P.: Particulate iron delivery to the water column
- 907 of the Amundsen Sea, Antarctica, Mar. Chem., 153, 15–30, doi:10.1016/j.marchem.2013.04.006, 2013.
- 908 Radic, A., Lacan, F. and Murray, J. W.: Iron isotopes in the seawater of the equatorial Pacific Ocean: New
- 909 constraints for the oceanic iron cycle, Earth Planet. Sci. Lett., 306(1-2), 1-10, doi:10.1016/j.eps1.2011.03.015,
- 910 2011.
- 911 Raiswell, R., Benning, L. G., Tranter, M. and Tulaczyk, S.: Bioavailable iron in the Southern Ocean: The
- 912 significance of the iceberg conveyor belt, Geochem. Trans., 9(1), 7, doi:10.1186/1467-4866-9-7, 2008.
- 913 Rijkenberg, M. J. A., Middag, R., Laan, P., Gerringa, L. J. A., Van Aken, H. M., Schoemann, V., De Jong, J. T.
- 914 M. and De Baar, H. J. W.: The distribution of dissolved iron in the West Atlantic Ocean, PLoS One, 9(6), 1-14,
- 915 doi:10.1371/journal.pone.0101323, 2014.
- 916 Rutgers Van Der Loeff, M. M., Meyer, R., Rudels, B. and Rachor, E.: Resuspension and particle transport in the
- 917 benthic nepheloid layer in and near Fram Strait in relation to faunal abundances and 234Th depletion, Deep. Res.
- 918 Part I Oceanogr. Res. Pap., 49(11), 1941–1958, doi:10.1016/S0967-0637(02)00113-9, 2002.
- 919 Sanders, R., Henson, S. A., Koski, M., De La Rocha, C. L., Painter, S. C., Poulton, A. J., Riley, J., Salihoglu, B.,
- 920 Visser, A., Yool, A., Bellerby, R. and Martin, A. P.: The Biological Carbon Pump in the North Atlantic, Prog.
- 921 Oceanogr., 129(PB), 200–218, doi:10.1016/j.pocean.2014.05.005, 2014.
- 922 Sarthou, G., Lherminier, and the GEOVIDE team: Introduction to the French GEOTRACES North Atlantic
- 923 Transect (GA01): GEOVIDE cruise, Biogeosciences, 15, 7097-7109, https://doi.org/10.5194/bg-15-7097-2018,
- 924 <u>2018.</u>
- 925
- 926 Sarthou, G., Vincent, D., Christaki, U., Obernosterer, I., Timmermans, K. R. and Brussaard, C. P. D.: The fate of
- 927 biogenic iron during a phytoplankton bloom induced by natural fertilisation: Impact of copepod grazing, Deep.
- 928 Res. Part II Top. Stud. Oceanogr., 55(5–7), 734–751, doi:10.1016/j.dsr2.2007.12.033, 2008.

- 929 Schlosser, C., Schmidt, K., Aquilina, A., Homoky, W. B., Castrillejo, M., Mills, R. A., Patey, M. D., Fielding, S.,
- 930 Atkinson, A. and Achterberg, E. P.: Mechanisms of dissolved and labile particulate iron supply to shelf waters
- 931 and phytoplankton blooms off South Georgia, Southern Ocean, Biogeosciences Discuss., 0049(July), 1-49,
- 932 doi:10.5194/bg-2017-299, 2017.
- 933 Shelley, R. U., Landing, W. M., Ussher, S. J., Planquett, H. and Sarthou, G.: Characterisation of aerosol
- 934 provenance from the fractional solubility of Fe (Al, Ti, Mn, Co, Ni, Cu, Zn, Cd and Pb) in North Atlantic aerosols
- 935 (GEOTRACES GA01 and GA03), Biogeosciencess, submitted(November), 1-31, doi:10.5194/bg-2017-415,
- 936 2017.
- 937 Shelley, R. U., Landing, W. M., Ussher, S. J., Planquette, H. and Sarthou, G.: Regional trends in the fractional
- 938 solubility of Fe and other metals from North Atlantic aerosols (GEOTRACES cruises GA01 and GA03) following
- 939 a two-stage leach, Biogeosciences, 155194(1), 2271–2288, doi:10.5194/bg-15-2271-2018, 2018.
- 940 Sherrell, R. M., Field, P. M. and Gao, Y.: Temporal variability of suspended mass and composition in the
- 941 Northeast Pacific water column: Relationships to sinking flux and lateral advection, Deep. Res. Part II Top. Stud.
- 942 Oceanogr., 45(4–5), 733–761, doi:10.1016/S0967-0645(97)00100-8, 1998.
- 943 Spinrad, R. W., Zaneveld, J. R. and Kitchen, J.C.: A Study of the Optical Characteristics of the Suspended Particles
- 944 Benthic N epheloid Layer of the Scotian Rise, J. Geophys. Res., 88, 7641–7645, doi:0148-0227/83/003C, 1983.
- 945 Statham, P. J., Skidmore, M. and Tranter, M.: Inputs of glacially derived dissolved and colloidal iron to the coastal
- 946 ocean and implications for primary productivity, Global Biogeochem. Cycles, 22(3), 1-11,
- 947 doi:10.1029/2007GB003106, 2008.
- 948 Straneo, F., Pickart, R. S. and Lavender, K.: Spreading of Labrador sea water: An advective-diffusive study based
- 949 on Lagrangian data, Deep. Res. Part I Oceanogr. Res. Pap., 50(6), 701–719, doi:10.1016/S0967-0637(03)00057-
- 950 8, 2003.
- 951 Sunda, W. G. and Huntsman, S. A.: Effect of Competitive Interactions Between Manganese and Copper on
- 952 Cellular Manganese and Growth in Estuarine and Oceanic Species of the Diatom Thalassiosira, Limnol.
- 953 Oceanogr., 28(5), 924–934, doi:10.4319/lo.1983.28.5.0924, 1983.
- 954 Tagliabue, A., Bopp, L., Dutay, J. C., Bowie, A. R., Chever, F., Jean-Baptiste, P., Bucciarelli, E., Lannuzel, D.,
- 955 Remenyi, T., Sarthou, G., Aumont, O., Gehlen, M. and Jeandel, C.: Hydrothermal contribution to the oceanic
- 956 dissolved iron inventory, Nat. Geosci., 3(4), 252–256, doi:10.1038/ngeo818, 2010.
- 957 Tagliabue, A., Bowie, A. R., Boyd, P. W., Buck, K. N., Johnson, K. S. and Saito, M. A.: The integral role of iron
- 958 in ocean biogeochemistry, Nature, 543(7643), 51–59, doi:10.1038/nature21058, 2017.
- 959 Taylor, S.-, and McLennan, S.-.: The geochemical evolution of the continental crust, Rev. Geophys., 33(2), 241-
- 960 265, doi:10.1029/95RG00262, 1995.

- 961 Tebo, B. M. and Emerson, S. R.: Effect of Oxygen Tension Manganese (II) Concentration and Temperature on
- 962 the Microbially Catalyzed Manganese-Ii Oxidation Rate in a Marine Fjord, Appl. Environ. Microbiol., 50(5),
- 963 1268–1273, 1985.
- 964 Tebo, B. M., Nealson, K. H., Emerson, S. and Jacobs, L.: Microbial mediation of Mn(II) and Co(II) precipitation
- 965 at the o2/H2S interfaces in two anoxic fjords, 29(6), 1247-1258, 1984.
- 966 Tonnard, M., Planquette, H., Bowie, A. R., van der Merwe, P., Gallinari, M., Desprez de Gésincourt, F., Germain,
- 967 Y., Gourain, A., Benetti, M., Reverdin, G., Tréguer, P., Boutorh, J., Cheize, M., Menzel Barraqueta, J., Pereira-
- 968 Contreira, L., Shelley, R., Lherminier, P., and Sarthou, G.: Dissolved iron in the North Atlantic Ocean and
- 969 Labrador Sea along the GEOVIDE section (GEOTRACES section GA01), Biogeosciences Discuss.,
- 970 https://doi.org/10.5194/bg-2018-147, 2018
- 971 Trefry, J. H., Trocine, R. P., Klinkhammer, G. P. and Rona, P. A.: Iron and copper enrichment of suspended
- particles in dispersed hydrothermal plumes along the mid????\_Atlantic Ridge, Geophys. Res. Lett., 12(8), 506–
- 973 509, doi:10.1029/GL012i008p00506, 1985.
- 974 Ussher, S. J., Achterberg, E. P. and Worsfold, P. J.: Marine biogeochemistry of iron, Environ. Chem., 1(2), 67-
- 975 80, doi:10.1071/EN04053, 2004.
- 976 Ussher, S. J., Worsfold, P. J., Achterberg, E. P., Laës, A., Blain, S., Laan, P., de Baar, H. J. W.: Distribution and
- 977 redox speciation of dissolved iron on the European continental margin, Limnol. Oceanogr., 52(6), 2530-2539,
- 978 doi:10.4319/lo.2007.52.6.2530, 2007.
- 979 Van der Merwe, P., Lannuzel, D., Bowie, A. R., Mancuso Nichols, C. A. and Meiners, K. M.: Iron fractionation
- 980 in pack and fast ice in East Antarctica: Temporal decoupling between the release of dissolved and particulate iron
- 981 during spring melt, Deep. Res. Part II Top. Stud. Oceanogr., 58(9–10), 1222–1236,
- 982 doi:10.1016/j.dsr2.2010.10.036, 2011a.
- Van Der Merwe, P., Lannuzel, D., Bowie, A. R. and Meiners, K. M.: High temporal resolution observations of
- 984 spring fast ice melt and seawater iron enrichment in East Antarctica, J. Geophys. Res. Biogeosciences, 116(3), 1-
- 985 18, doi:10.1029/2010JG001628, 2011b.
- 986 Weinstein, S. E. and Moran, S. B.: Distribution of size-fractionated particulate trace metals collected by bottles
- 987 and in-situ pumps in the Gulf of Maine-Scotian Shelf and Labrador Sea, Mar. Chem., 87(3-4), 121-135,
- 988 doi:10.1016/j.marchem.2004.02.004, 2004.
- 989 Yashayaev, I.: Hydrographic changes in the Labrador Sea, 1960-2005, Prog. Oceanogr., 73(3-4), 242-276,
- 990 doi:10.1016/j.pocean.2007.04.015, 2007.
- 991 Yashayaev, I. and Loder, J. W.: Enhanced production of Labrador Sea Water in 2008, Geophys. Res. Lett., 36(1),
- 992 doi:10.1029/2008GL036162, 2009.

Zunino, P., Lherminier, P., Mercier, H., Daniault, N., García-Ibáñez, M. I., and Pérez, F. F.: The GEOVIDE cruise in May–June 2014 reveals an intense Meridional Overturning Circulation over a cold and fresh subpolar North Atlantic. Biogeosciences, 14(23), 5323, 2017.

Figure 1: Map of stations where suspended particle samples were collected with GO-FLO bottles during the GEOVIDE cruise (GA01). Biogeochemical provinces are indicated by red squares, IM: Iberian Margin, IAP: Iberian Abyssal Plain, WEB: Western European Basin, IcB: Iceland Basin, RR: Reykjanes Ridge, IrB: Irminger Basin, GS: Greenland Shelf, LB: Labrador Basin, NS: Newfoundland Shelf. This figure was generated by Ocean Data View (Schlitzer, R., Ocean Data View, odv.awi.de, 2017).

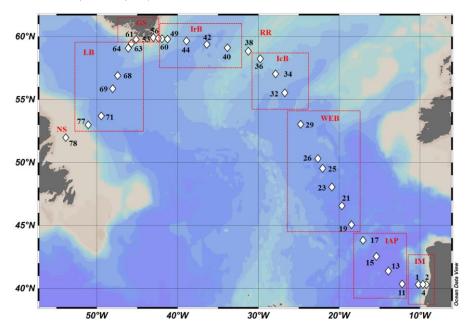


Figure 2: Salinity section during the GEOVIDE cruise. Water masses are indicated in black, MW: Mediterranean Water; NACW: North Atlantic Central Water; NEADW: North East Atlantic Deep Water; LSW: Labrador Sea Water; DSOW: Denmark Strait Overflow Water; ISOW: Iceland-Scotland Overflow Water; SAIW: Sub-Arctic Intermediate Water; IcSPMW: Iceland Sub-Polar Mode Water, IrSPMW: Irminger Sub-Polar Mode Water. Stations locations are indicated by the numbers. Biogeochemical provinces are indicated in blue font above station numbers. Contour of salinity = 35.8psu have been applyied to identify the Mediterranean Water. This figure was generated by Ocean Data View (Schlitzer, R., Ocean Data View, odv.awi.de, 2017).

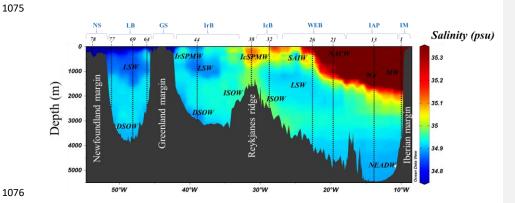
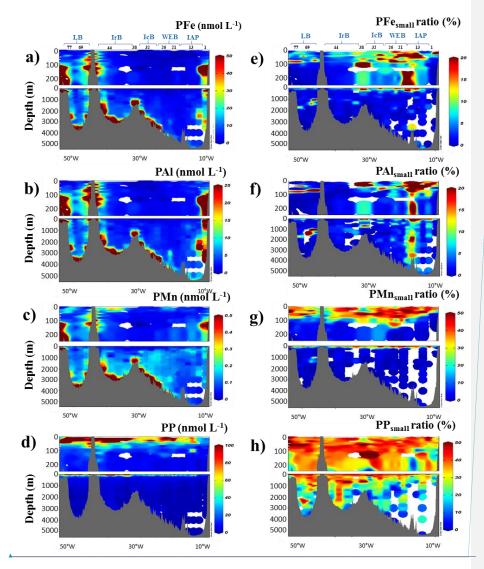


Figure 3: Left)- panel: Distribution of (a) total particulate iron (a, PFe), (b) aluminium (b, PAl), (c) manganese (e, PMn) and (d) phosphorus (d-PP) concentrations (in-nmol L-1) along the GEOVIDE section. Right panel; Contribution of the small size fraction (0,45-5  $\mu$ m) expressed as a percentage (%) of the total concentration of (e) PFe-(e), (f) PAl-(f), (g) PMn-(g) and (h) PP-(h). Station IDs and biogeochemical regions are indicated on top of section a. This figure was generated by Ocean Data View (Schlitzer, R., Ocean Data View, odv.awi.de, 2017).





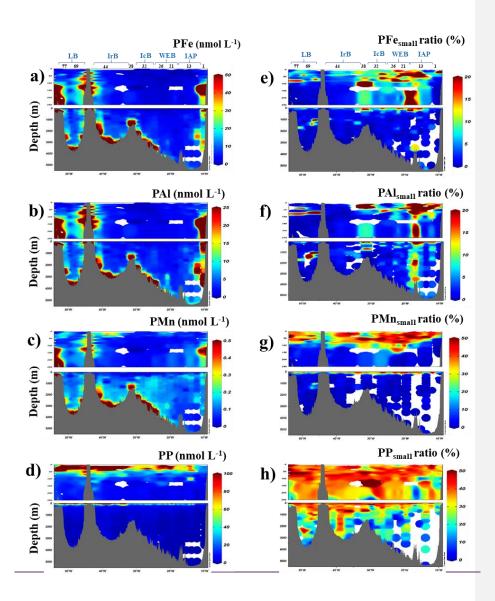


Figure 4: Section of derived contributions of sedimentary inputs (a) manganese bulk sediment proxy-(a) and (b) transmissometry (b) along the GA01 section. Station IDs and biogeochemical region are indicated above the section (a). This figure was generated by Ocean Data View (Schlitzer, R., Ocean Data View, odv.awi.de, 2017).

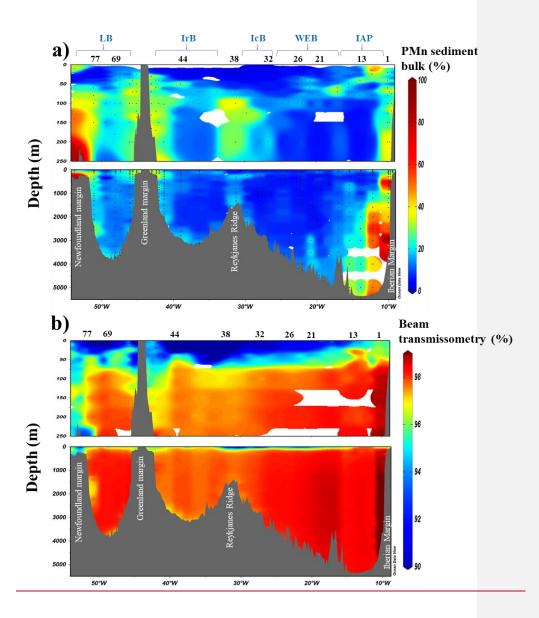


Figure 4: Boxplot figure of the particulate iron vertical profile (in nmol  $L^{-1}$ ) in the a) Iberian abyssal plain (IAP), b) Western European basin (WEB), c) Icelandic basin (IeB), d) Irminger basin (IrB) and c) Labrador basins (LB). Please note the change of PFe scale between the basins. The left boundary of the box represents the  $25^{th}$  percentile while the right boundary represents the  $75^{th}$  percentile, the line within the box marks the median value. Whiskers represent the

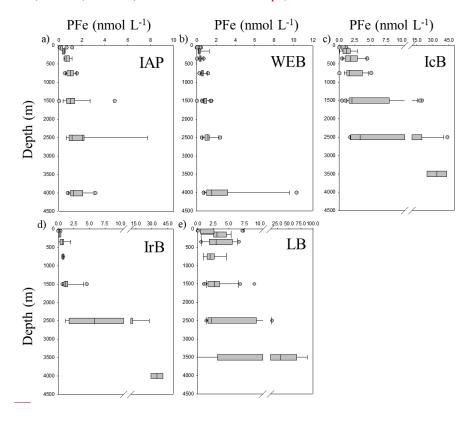
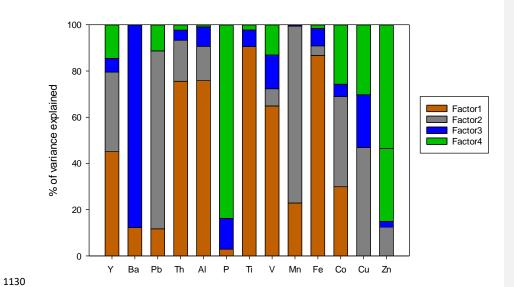
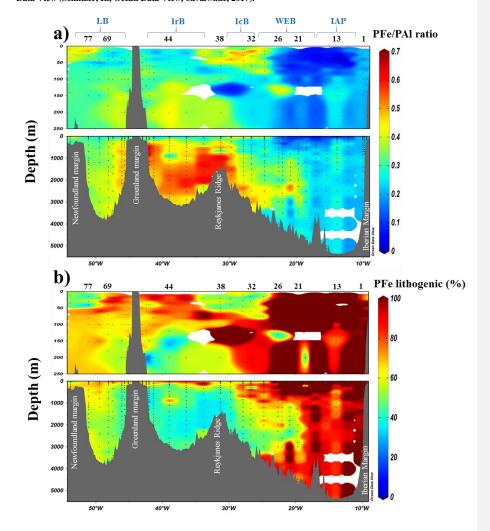


Figure 5: Factor fingerprint of the positive matrix factorisation. The four factors are represented in a stacked bar chart of the percentage of variance explained per element.

**Commented [RS1]:** It might be a good idea to list what each factor is dominated by, e.g. Factor 1 is dominated by the lithogenic elements, Ti, Fe, Al and Th, etc

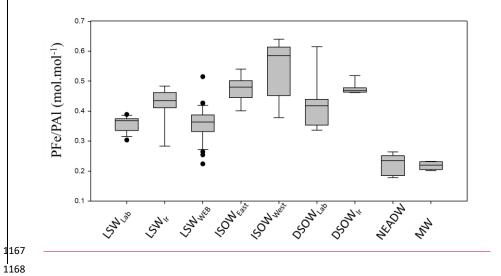


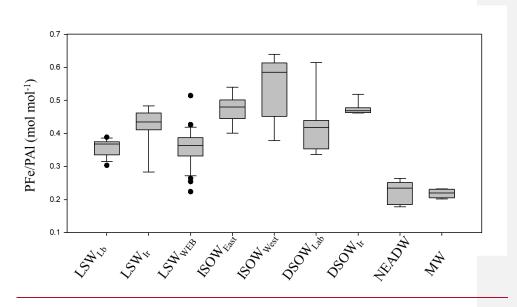


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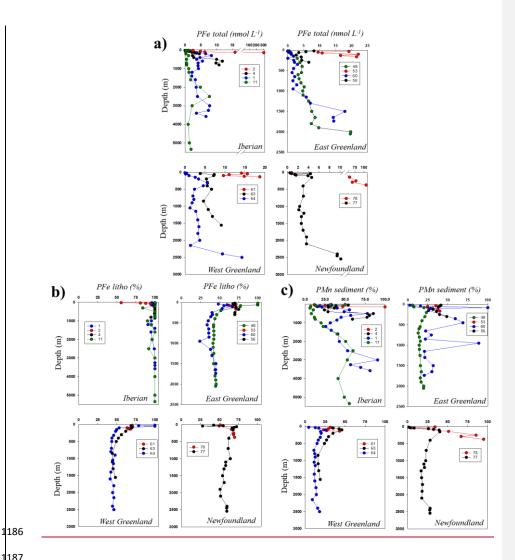
Water masses for which we had less than 5 data points for PFe/PAl were excluded from this test. As the differences in the median values among the treatment groups were greater than would be expected by chance; the difference in PFe/PAl between water masses is statistically significant (P = <0.001).

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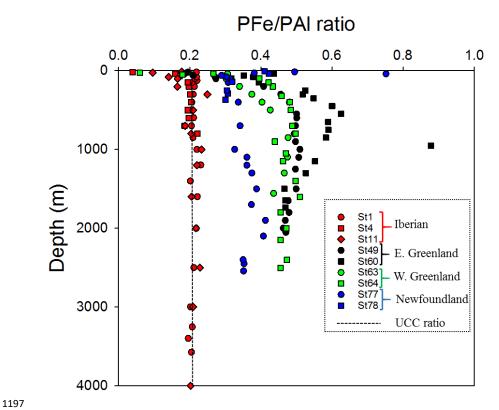


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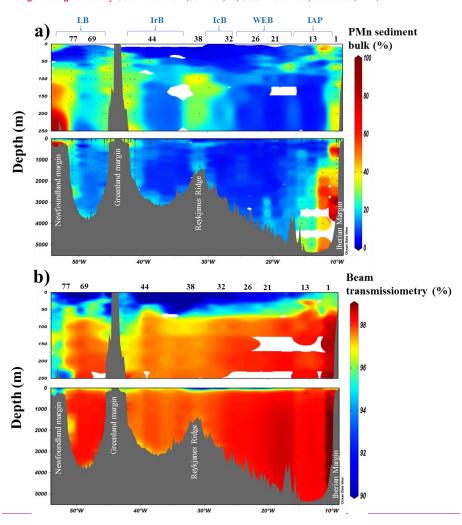
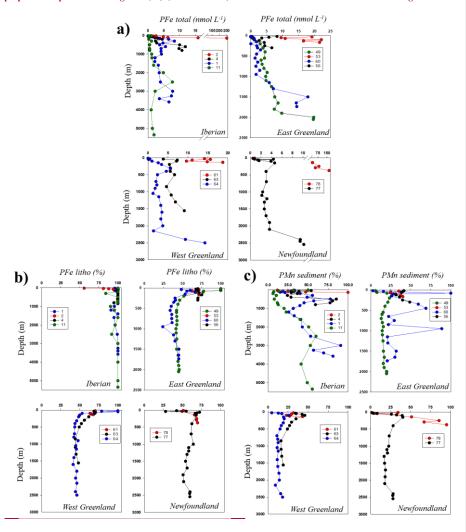
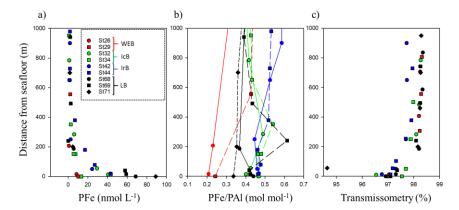


Figure 10: Vertical profiles of PFe (nmol  $L^4$ , a), lithogenic proportion of particulate iron (%, b) and sedimentary proportion of particulate manganese (%, e) at the Iberian, East-West Greenland and Newfoundland margins.





		Fe	Al	P	Mn
Pl 1 ( 11:1)	5μm filter 0.072		0.100	0.511	0.003
Blank (nmol L <sup>-1</sup> )	0.45µm filter	0.132	0.164	1.454	0.005
T: : C1	5µm filter	0.011	0.030	0.365	0.001
Limit of detection (nmol L <sup>-1</sup> )	0.45µm filter	0.026	0.046	1.190	0.001
	BCR-414 (n=10)	88 ± 7			$94 \pm 7$
Recovery CRM (%)	MESS-4 (n=5)	98 ± 14	$97 \pm 14$	$80\pm30$	$110\pm18$
	PACS-3 (n=8)	101 ± 9	$99 \pm 14$	$91 \pm 34$	$112\pm11$

 $\label{eq:continuous_continuous$ 

Author	Year	Fraction	Location	Depth range	PFe	PAI	PMn	PP
This study		>0.45µm	N. Atlantic (>40°N)	All	bdl-304	bdl-1544	bdl-3.5	bdl-402
Barrett et al.	2012	0.4um	N. Atlantic (25-60°N)	Upper 1000m	0.29-1.71	0.2-19.7		
Dammshauser et al.	2013	>0.2 µm	Eastern tropical N.A.	0-200		0.59-17.7		
Dammshauser et al.	2013	>0.2 µm	Meridional Atlantic	0-200		0.35-16.1		
Lam et al.	2012	1–51 um	Eastern tropical N.A.	0-600	ND-12			
Lannuzel et al.	2011	>0.2 µm	East Antarctic	Surface		0.02-10.67	0.01-0.14	
Lannuzel et al.	2014	>0.2 μm	East Antarctic	Fast ice	43-10385	121-31372	1-307	
Lee et al.	2017	>0.8 µm	Eastern tropical S.Pacific	All	bdl-159	bdl-162	bdl-8.7	bdl-983
Marsay et al.	2017	>0.4 µm	Ross Sea	All	0.68-57.3	ND-185	ND-1.4	5.4-404
Milne et al.	2017	>0.45µm	Sub-tropical N.A.	All	ND-140	ND-800		
Ohnemus et al.	2015	0.8–51 μm	N. Atlantic	All	0-938	0-3600		
Planquette et al.	2009	>53 μm	Southern Ocean	30-340	0.15-13.2	0.11-25.5		
Schlosser et al.	2017	>1 µm	South Georgia Shelf	All	0.87-267	0.6-195	0.01-3.85	
Sherrell et al.	1998	1-53um	Northeast Pacific	0-3557		0.0-54.2		
Weinstein et al.	2004	>53 µm	Labrador Sea	0-250	0.1-1.2	0.1-1.5		
Weinstein et al.	2004	0.4- 10um	Labrador Sea	0-250	2.5	3.6	0.05	
Weinstein et al.	2004	>0.4 µm	Gulf of Maine	0-300	34.8	109		

Table 2: Concentration (in nmol  $L^{-1}$ ) of trace elements (PFe, Pal, PMn and PP) in suspended particles collected in diverse regions of the world's ocean. Bdl: below detection limit, ND: non-determined.