

# Christian Schlosser reviews

Dear Christian Schlosser and Editor,

The reviewers are thanked for their insightful comments; these have helped to improve the manuscript considerably. Please see our detailed answers to the referees' comments below. Line numbers refer to the new version.

All the answers are attached as a supplementary file.

Best regards,

Tonnard et al.,

## Abstract

Please, note that we added François Lacan as a co-author.

Line 29ff: Air-sea interactions responsible for deep winter convection – Did you mean special cooling!

→ We have changed the sentence "Air-sea interactions were suspected to be responsible for the increase in DFe concentrations within subsurface waters of the Irminger Sea due to deep convection occurring the previous winter..." by "Enhanced air-sea interactions were suspected to be responsible for the increase in DFe concentrations within subsurface waters of the Irminger Sea due to deep convection occurring the previous winter..."

## Introduction

Page 2 Line 4: I would also include oxygen, the whole ventilation and redox state of the deep ocean depends on deep water formation in the North Atlantic and Weddell Sea

→ We removed this sentence.

Page 2 Line 6: "stores" is maybe the wrong term; I would rather go with "accumulates"

→ We have modified this part based on your comment and Reviewer#2 comment.

Page 2 Lines 2-33 Page 3 Lines 1-25: The North Atlantic Ocean is known for its pronounced spring phytoplankton blooms (Henson et al., 2009; Longhurst, 2007). Phytoplankton blooms induce the capture of aqueous carbon dioxide through photosynthesis, and conversion into particulate organic carbon (POC). This POC is then exported into deeper waters through the production of sinking biogenic particles and ocean currents. Via these processes, and in conjunction with the physical carbon pump, the North Atlantic Ocean is the largest oceanic sink of anthropogenic CO<sub>2</sub> (Pérez et al., 2013), despite covering only 15% of global ocean area (Humphreys et al., 2016; Sabine et al., 2004) and is therefore crucial for Earth's climate.

Indeed, phytoplankton must obtain, besides light and inorganic carbon, chemical forms of essential elements, termed nutrients to be able of photosynthesis. Indeed, Fe is a key element for a number of metabolic processes (e.g. Morel et al., 2008). The availability of these nutrients in the upper ocean frequently limits the activity and abundance of these organisms together with light conditions

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(Moore et al., 2013). In particular, winter nutrient reserves in surface waters set an upper limit for biomass accumulation during the annual spring-to-summer bloom and will influence the duration of the bloom (Follows and Dutkiewicz, 2001; Henson et al., 2009; Moore et al., 2013; 2008). Hence, nutrient depletion due to biological consumption is considered as a major factor in the decline of blooms (Harrison et al., 2013).

The extensive studies conducted in the North Atlantic Ocean through the Continuous Plankton Recorder (CPR) have highlighted the relationship between the strength of the westerlies and the displacement of the subarctic front (SAF), (which corresponds to the North Atlantic Oscillation (NAO) index (Bersch et al., 2007)), and the phytoplankton dynamics of the central North Atlantic Ocean (Barton et al., 2003). Therefore, the SAF not only delineates the subtropical gyre from the subpolar gyre but also two distinct systems in which phytoplankton limitations are controlled by different factors. In the North Atlantic Ocean, spring phytoplankton growth is largely light-limited within the subpolar gyre. Light levels are primarily set by freeze-thaw cycles of sea ice and the high-latitude extremes in the solar cycle (Longhurst, 2007). Simultaneously, intense winter mixing supplies surface waters with high concentrations of nutrients. In contrast, within the subtropical gyre, the spring phytoplankton growth is less impacted by the light regime and has been shown to be N and P-co-limited (e.g. Harrison et al., 2013; Moore et al., 2008). This is principally driven by Ekman downwelling with an associated export of nutrients out of the euphotic zone (Oschlies, 2002). Thus, depending on the location of the SAF, phytoplankton communities from the central North Atlantic Ocean will be primarily light or nutrient limited.

However, once the water column stratifies and phytoplankton are released from light limitation, seasonal high-nutrient, low chlorophyll (HNLC) conditions were reported at the transition zone between the gyres, especially in the Irminger Sea and Iceland Basin (Sanders et al., 2005). In these HNLC zones, trace metals are most likely limiting the biological carbon pump. Among all the trace metals, Fe has been recognized as the prime limiting element of North Atlantic primary productivity (e.g. Boyd et al., 2000; Martin et al., 1994; 1988; 1990). However, the phytoplankton community has been shown to become N and/or Fe-(co)-limited in the Iceland Basin and the Irminger Sea (e.g. Nielsdóttir et al., 2009; Painter et al., 2014; Sanders et al., 2005).

In the North Atlantic Ocean, dissolved Fe (DFe) is delivered through multiple pathways such as ice-melting (e.g. Klunder et al., 2012; Tovar-Sanchez et al., 2010), atmospheric inputs (Achterberg et al., 2018; Baker et al., 2013; Shelley et al., 2015; 2017), coastal runoff (Rijkenberg et al., 2014), sediment inputs (Hatta et al., 2015), hydrothermal inputs (Achterberg et al., 2018; Conway and John, 2014) and by water mass circulation (vertical and lateral advections, e.g. Laes et al., 2003). Dissolved Fe can be regenerated through biological recycling (microbial loop, zooplankton grazing, e.g. Boyd et al., 2010; Sarthou et al., 2008). Iron is removed from the dissolved phase by biological uptake, export and scavenging along the water column and precipitation (itself a function of salinity, pH of seawater and ligand concentrations).

Although many studies investigated the distribution of DFe in the North Atlantic Ocean, much of this work was restricted to the upper layers (< 1000 m depth) or to one basin. Therefore, uncertainties

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remain on the large-scale distribution of DFe in the North Atlantic Ocean and more specifically within the subpolar gyre where few studies have been undertaken, and even fewer in the Labrador Sea. In this biogeochemically important area, high-resolution studies are still lacking for understanding the processes influencing the cycle of DFe.

The aim of this paper is to elucidate the sources and sinks of DFe, its distribution regarding water masses and assesses the links with biological activity along the GEOVIDE (GEOTRACES-GA01) transect. This transect spanned several biogeochemical provinces including the West European Basin, the Iceland Basin, the Irminger and the Labrador Seas (Fig. 1). In doing so we hope to constrain the potential long-range transport of DFe through the Deep Western Boundary Current (DWBC) via the investigation of the local processes effecting the DFe concentrations within the three main water masses that constitute it: Iceland Scotland Overflow Water (ISOW), Denmark Strait Overflow Water (DSOW) and Labrador Sea Water (LSW).

## MM

Page 3 Line 11: Remove “the” from “. . .aboard the N/O. . .”

→ We have modified this part as suggested

Page 3 Line 24ff: Two different filtration techniques were applied, 0.2 and 0.45  $\mu\text{m}$ . Did you test that both approaches deliver the same result? I know water is restricted and sometimes sampling techniques need to be changed, however, please indicate why you did this and that swapping between both filtration techniques did not cause problems (offset, etc.)!

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→ We have added precision as suggested. Please note that there was no station where both filtration techniques were used.

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Page 4 Lines 10-14: Samples were either taken from the filtrate of particulate samples (collected on polyethersulfone filters, 0.45  $\mu\text{m}$  supor<sup>®</sup>, see Gourain et al., this issue) or after filtration using 0.2  $\mu\text{m}$  filter cartridges (Sartorius SARTOBRAN<sup>®</sup> 300) due to water budget restriction (Table 1). No significant difference was observed between DFe values filtered through 0.2  $\mu\text{m}$  and 0.45  $\mu\text{m}$  filters (p-value > 0.2, Wilcoxon test) for most stations. Differences were only observed between profiles of stations 11 and 13 and, 13 and 15.

Page 3 Line 26: exchange “on” by “using”. By the way, did you apply pressured air to the Go-Flo’s to filter your samples. If so, please state that!

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→ We have added precision as suggested

Page 3 Line 30: You did you use 0.2% HCl to acidify your water, or? It reads like that! I assume you used concentrated HCl and the dilution with the seawater was than 0.2%.

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→ We have modified the text for clarification

Page 4 Lines 17-18: Samples were then acidified to ~ pH 1.7 with HCl (Ultrapur<sup>®</sup> Merck, 2 %<sub>v/v</sub>) under a class 100 laminar flow hood inside the clean container.

Page 4 Line 2: The first sentence does not fit here; first you preconcentrated your sample using a SeaFAST system. Then the eluent was introduced via a PFA nebulizer and cyclonic spray chamber into your instrument (please indicate what kind of instrument you used, Element?). Please clarify!

→ We have modified this part as suggested

Page 4 Lines 27-30: Seawater samples were preconcentrated using a SeaFAST-pico™ (ESI, Elemental Scientific, USA) and the eluent was directly introduced via a PFA-ST nebulizer and a cyclonic spray chamber in an Element XR Sector Field Inductively Coupled Plasma Mass Spectrometry (Element XR SF-ICP-MS, Thermo Fisher Scientific Inc., Omaha, NE), following the protocol of Lagerström et al. (2013).

Note that we have also changed part of the reagent text, as we found out there were mistakes.

Page 4 Lines 31-33, Page 5 Lines 1-4: High-purity grade solutions and water (Milli-Q) were used to prepare the following reagents: the acetic acid-ammonium acetate buffer (CH<sub>3</sub>COO<sup>-</sup> and NH<sub>4</sub><sup>+</sup>) was made of 140 mL acetic acid (> 99% NORMATOM® - VWR chemicals) and ammonium hydroxide (25%, Merck Suprapur®) in 500 mL PTFE bottles and was adjusted to pH 6.0 ± 0.2 for the on-line pH adjustment of the samples. The eluent was made of 1.4 M nitric acid (HNO<sub>3</sub>, Merck Ultrapur®) in Milli-Q water by a 10-fold dilution and spiked with 1 µg L<sup>-1</sup> <sup>115</sup>In (SCP Science calibration standards) to allow for drift correction. Autosampler and column rinsing solutions were made of HNO<sub>3</sub> 2.5% (v/v) (Merck Suprapur®) in Milli-Q water. The carrier solution driven by the syringe pumps to move the sample and buffer through the flow injection system was made in the same way.

Page 4 Line 11: gravimetrically is perhaps not the right word, you used a balance, right!

→ We did not changed our sentence here since gravimetrically means by weighting the standards.

Page 4 Line 13ff: please include "...in-house standard seawater..", was this seawater acidified in the same way?

→ We have modified this part as suggested and added at the end of section 2.1 the precision on the sampling and acidification methods of the in-house standard seawater.

In section 2.1:

Page 4 lines 20-25: Large volumes of seawater sample (referred hereafter as the in-house standard seawater) were also collected using a towed fish at around 2-3 m deep and filtered in-line inside a clean container through a 0.2 µm pore size filter capsule (Sartorius SARTOBAN® 300) and was stored unacidified in 20-30 L LDPE carboys (Nalgene™). All the carboys were cleaned following the guidelines of the GEOTRACES Cookbook (Cutter et al., 2017). This in-house standard seawater was used for calibration on the SeaFAST-pico™ - SF-ICP-MS (see Section 2.2) and was acidified to ~ pH 1.7 with HCl (Ultrapur® Merck, 2 % v/v) at least 24h prior to analysis.

Page 4 Line 16ff: Please include the analytical precision, the blank, detection limit of the analytical method. And how many samples did you run normally and how much samples were between each calibration curve? Please also include, how you calculated your errors, standard deviation of the three slopes? Or just the s.d. of the Element?

→ We have added precision as suggested.

Page 5 Lines 11-20: Data were blank-corrected by subtracting an average acidified Milli-Q blank that was pre-concentrated on the SeaFAST-pico™ in the same way as the samples and seawater

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standards. Each analytical session consisted of about fifty samples and two calibrations, one at the beginning and another one at the end of each analytical session. The errors associated to each sample were calculated as the standard deviation for five measurements of low-Fe seawater samples. The mean Milli-Q blank was equal to  $0.08 \pm 0.09 \text{ nmol L}^{-1}$  ( $n = 17$ ). The detection limit, calculated for a given run as 3 times the standard deviation of the Milli-Q blanks, was on average  $0.05 \pm 0.05 \text{ nmol L}^{-1}$  ( $n = 17$ ). Reproducibility was assessed through the standard deviation of replicate samples (every 10th sample was a replicate) and the average of the in-house standard seawater, and was equal to 17% ( $n = 84$ ). Accuracy was determined from the analysis of consensus (SAFE S, GSP) and certified (NASS-7) seawater matrices (see Table 2) and in-house standard seawater ( $D\text{Fe} = 0.42 \pm 0.07 \text{ nmol L}^{-1}$ ,  $n = 84$ ).

Page 4 Line 21: The CTD sensors were deployed on a stainless steel rosette. Correct? Please indicate and correct throw-out the rest of the text.

→ We have modified this part as suggested

Page 6 Lines 11-15: Potential temperature ( $\theta$ ), Salinity ( $S$ ), dissolved oxygen ( $O_2$ ) and beam attenuation data were retrieved from the CTD sensors (CTD SBE911 equipped with a SBE-43) that were deployed on a stainless steel rosette. Nutrient and pigment samples were obtained from the stainless steel rosette casts and analysed according to Aminot and Kerouel (2007) and Ras et al. (2008), respectively. We used the data from the stainless steel rosette casts that were deployed immediately before or after our TMR casts. All these data are/will be available on the LEFE/CYBER database (<http://www.obs-vlfr.fr/proof/php/geovide/geovide.php>).

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Page 4 Line 28: Name the parameter  $\Delta\sigma$

▪ We have added this precision (Page 6 Line 18): "...where  $Z_m$  is defined as an absolute change in the density of seawater at a given temperature ( $\Delta\sigma_p \geq 0.125 \text{ kg m}^{-3}$ )..."

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Page 4 Line 30: What do you mean with perturbation, at which depth, please indicate in Table 1, for which station this was the case.

→ We have changed the word "perturbation" by "disturbance" for clarification. In addition, we have reported in Table 1 the precision on whether temperature and salinity profiles were uniform or disturbed with an asterisk symbol next to stations where profiles were not uniform and we added the following sentence in the legend of Table 1: "Note that the asterisk next to station numbers refers to disturbed temperature and salinity profiles as opposed to uniform profiles."

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Page 5 Line 2ff: Please indicate for which data you applied statistics on?

→ We have modified the text as suggested.

Page 6 Lines (23-24): All statistical approaches, namely the comparison between the pore size used for filtration, correlations and Principal Component Analysis (PCA), were performed using the R statistical software (R development Core Team 2012).

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Page 5 Line 3: You did not measure the p-value, you maybe determined or calculated the value.

→ We have modified this part as suggested (Page 6 Line 25),

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Page 5 Line 15: Include "...540 data points. . .

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→ We have modified this part as suggested (Page 7 Lines 8-9).

Page 5 Line 19: Exchange “The complete relational database. . .” by “The complete data set. . .”

→ We have modified this part as suggested (Page 7 Line 12)

## Results

Page 5 Line 27: I would swap the two sentences “For a schematic of water masses, currents and pathways, see Danialt et al. (2016).” and “Hereafter we summarise the main features (Fig. 1 and 2).”

→ We have modified this part as suggested (Page 7 Lines 19-20)

Page 6 Line 1: Give a depth range of the “Upper waters (0 – 800 m)” or so! Please also include this to the Intermediate and Deep waters.

→ We have added precisions as suggested.

Page 6 Line 5: Did you mean with central water the Subarctic intermediate water (SAIW). Please clarify! Please also increase the letter size in Fig. 2. It is really hard to see on a normal A4 print out! There are no currents in Fig. 2, either you somehow include them or remove the caption.

→ By central waters, we meant ENACW as defined in the first sentence of the paragraph and therefore changed “Cnetral Waters” by “ENACW”. We have removed the currents from the figure caption in Fig. 2 and we have increased the font size.

Page 6 Line 18: Please rewrite “..Labrador Sea Water (LSW).

→ We have modified this part as suggested (Page 8 Line 9)

Page 6 Line 29ff: I do not understand the sentence, starting with “During GA01,. . .”

→ We have rewritten this part

Page 8 Lines 19-20: During GEOVIDE, LSW formed by deep convection the previous winter was found at several stations from the Labrador Sea (68, 69, 71 and 77).

Page 6 Line 30ff, I am not sure about, explaining the different flow paths, It is really hard to follow without any drawing. Other question, is it really important, since you are just interested in water masses and their DFe signal, and not about currents! I would remove that!

→ We have modified this part to make it shorter.

Page 8 Lines 20-22: After convecting, LSW splits into three main branches with two main cores separated by the Reykjanes Ridge (stations 1-32, West European and Iceland Basins; stations 40-60, Irminger Sea), and the last one entering the West European Basin (Zunino et al., 2017).

Page 7 Line 8: I do not see any silicic acid and nitrate data, please indicate concentrations and where they can be found.

→ We have added the averages and SD for silicic acid and nitrate concentrations (Page 8, Line 26) and the reference where data can be found.

Page 8 Lines 24-27: North East Atlantic Deep Water (NEADW,  $1.98 < \theta < 2.50^{\circ}\text{C}$ ,  $34.895 < S < 34.940$ ) was the dominant water mass in the West European Basin at stations 1-29 from 2000 m depth to the

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bottom and is characterized by high silicic acid ( $42 \pm 4 \mu\text{mol L}^{-1}$ ), nitrate ( $21.9 \pm 1.5 \mu\text{mol L}^{-1}$ ) concentrations and lower oxygen concentration ( $\text{O}_2 \approx 252 \mu\text{mol kg}^{-1}$ ) (see Sarthou et al., 2018).

Page 7 Line 9ff: It is hard to understand what you mean with "PIW is in contact with the atmosphere once a year (?) during the time of winter convection.." All together there is a lot of water mass information, that can be found elsewhere in the special issue, I would rather shorten that part of the result section.

→ We have modified this part to make it shorter

Page 8 Lines 28-32: Polar Intermediate Water (PIW,  $\theta \approx 0^\circ\text{C}$ ,  $S \approx 34.65$ ) is a ventilated, dense, low-salinity water intrusion to the deep overflows within the Irminger and Labrador Seas that is formed at the Greenland shelf. PIW represents only a small contribution to the whole water mass pool (up to 27%) and was observed over the Greenland slope at stations 53 and 61 as well as in surface waters from station 63 (from 0 to ~ 200 m depth), in intermediate waters of stations 49, 60 and 63 (from ~ 500 to ~ 1500 m depth) and in bottom waters of stations 44, 68, 69, 71 and 77 with a contribution higher than 10%.

Page 7 Line 30: Cannot check if this is correct! No nitrate data available.

→ The location of these data was already precised "Sarthou et al. (this issue)". However, we have changed this reference by the accurate one and added the reference of the SEANOE data base and associated paper: García-Ibáñez et al., 2018; Pérez et al., 2018; Sarthou et al., 2018 Please note that in this manuscript, Nitrate data are changed for RFe/N data, therefore we did not added the nitrate data.

Page 8 Line 11-12: This is school book knowledge, that is why we are using sensors! Remove the two sentences! However this entire section 3.2.2 needs an overhaul.

→ We have removed this part as suggested.

Page 9 Lines 24-28: Overall, most of the phytoplankton biomass was localised above 100 m depth with lower total chlorophyll-*a* (TChl-*a*) concentrations South of the Subarctic Front and higher at higher latitudes (Fig. 3). While comparing TChl-*a* maxima considering all stations, the lowest value ( $0.35 \text{ mg m}^{-3}$ ) was measured within the West European Basin (station 19, 50 m depth) while the highest values were measured at the Greenland (up to  $4.9 \text{ mg m}^{-3}$ , 30 m depth, station 53 and up to  $6.6 \text{ mg m}^{-3}$ , 23 m depth, station 61) and Newfoundland (up to  $9.6 \text{ mg m}^{-3}$ , 30 m depth, station 78) margins.

Page 8 Line 20ff: You can delete the first three sentences, they do not contain any important data!

→ We have removed this part as suggested

Page 8 Line 29: Also station 61 and 78 are high, at least this is shown by your plot!

→ We were talking about enhanced DFe at the surface compared to deeper DFe values, and this is only the case for stations 2, 4 and 56.

And replace "...were around..." by "...ranged from..."

→ We have modified the text as suggested.

Generally, I would merge section 3.3, 3.3.1 and 3.3.2.

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→ We have modified the text as suggested.

Figure 4: Are you sure that single elevated values at site 40 (1500m) and at site 44 (500m) are correct. They just seem like outliers to me! Do we really need Fig.5 and 6, we see everything already in Fig. 4.

→ These enhanced DFe concentrations are consistent with high ligand concentrations measured during this study. In addition, these samples were analysed during 3 separated analytical sessions on the seaFAST SF-ICP-MS. Regarding Figs 5 and 6, we agree that Fig. 6 is repetitive and therefore we removed it from the ms and included it into the supplementary material. However, we think Fig. 5 is helpful to understand the section 4.2.2 on high latitudes meteoric water and sea-ice processes.

Page 9 Line 1ff: rewrite sentence, hard to read!

→ We have rewritten the sentence

Page 10 Lines 7-9: Considering the four oceanic basins, mean vertical profiles (supplementary material Fig. S2) showed increasing DFe concentrations down to 3000 m depth followed by decreasing DFe concentrations down to the bottom. Among deep-water masses, the lowest DFe concentrations were measured in the West European Basin.

Page 9 Line 6ff: Please provide numbers for surface waters.

→ We have added this precision.

Page 10 Lines 12-14: Overall, surface DFe concentrations were higher ( $0.36 \pm 0.18 \text{ nmol L}^{-1}$ ) in the North Atlantic Subpolar gyre (above  $52^{\circ}\text{N}$ ) than in the North Atlantic Subtropical gyre ( $0.17 \pm 0.05 \text{ nmol L}^{-1}$ ).

Page Line 9ff: But also at station 21 the DFe value is high. I do not think they are significantly different from the others, s.d. is  $\pm 20\%$  and higher.

→ We agree with you and removed this sentence.

Page 9 Line 17: NEADW was very similar to the median GEOVIDE voyage but compared to test of deep waters lower, please rewrite! But the DSOW in the Labrador Sea was similar.

→ We have modified the text accordingly

Page 11 Lines 8-11: The DFe concentrations in the NEADW were relatively similar to the DFe median value of the GEOVIDE voyage (median DFe =  $0.75 \text{ nmol L}^{-1}$ , Figs. 4 and 7) with an average value of  $0.74 \pm 0.16 \text{ nmol L}^{-1}$  ( $n=18$ ) and presented relatively low median DFe concentrations (median DFe =  $0.71 \text{ nmol L}^{-1}$ ) compared to other deep water masses.

I am not sure Fig. 7 is really required. It just comprises what we already see in Fig. 4. And apart from some outliers (hydrothermal? Any Mn data), surface waters, NADW and waters from the Labrador Sea, concentrations are around 1nM. And as numerous times shown, it is impossible to fingerprint water masses with DFe.

→ We agree with this suggestion and Figure 7 was removed from the MS and added to supplementary material.

Page 10 Line 9: Others showed also elevated concentrations, for instance, station 44. However I understand why the authors decided to explain both station! For myself station 1 is not a problem, it

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is very close to the continental margin and influenced by lateral water mass transport than the other stations farther off-shore. However, site 17 is a bit more tricky. Did you reanalyze that station, that would confirm that the analysis was alright and you do not face just a strange offset. Anyway, I would discuss station 17, but please rephrase some sentences, it was really hard to grasp the issue you wanted to bring across. From the first sentence it should be clear what the issue is, than explain (eg. Concentrations are irregularly high).

→ We have modified the text as suggested

Page 11 Lines 20-28: Considering the entire section, two stations (stations 1 and 17) showed irregularly high DFe concentrations ( $> 1 \text{ nmol L}^{-1}$ ) throughout the water column, thus suggesting analytical issues. However, these two stations were analysed twice and provided similar results, therefore discarding any analytical issues. This means that these high values originated either from genuine processes or from contamination issues. If there had been contamination issues, one would expect a more random distribution of DFe concentrations and less consistence throughout the water column. It thus appears that contamination issues were unlikely to happen. Similarly, the influence of water masses to explain these distributions was discarded as the observed high homogenized DFe concentrations were restricted to these two stations. Station 1, located at the continental shelf-break of the Iberian Margin, also showed enhanced PFe concentrations from lithogenic origin suggesting a margin source (Gourain et al., 2018).

Page 10 Line 23ff: Please provide the numbers from the other studies. Would it be possible to plot the surface DFe concentration and put the graph in the sup material. Than you can relate to that!

→ We have provided the numbers from other studies, updated Table 3 with the DFe values from Achterberg et al., 2018 and we added the following plot to the supplementary material:

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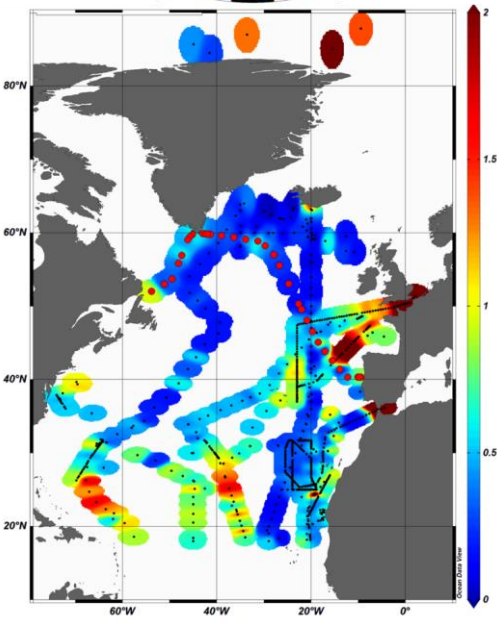
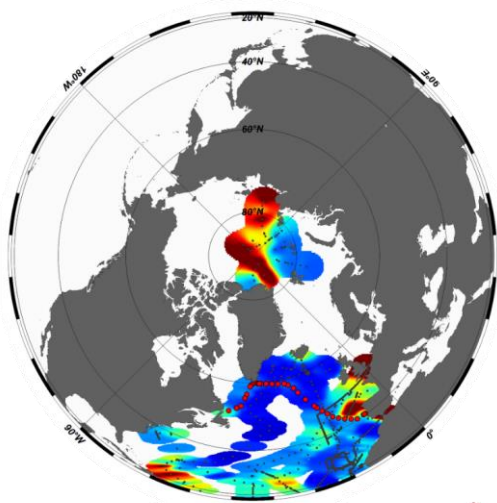


Figure S4: Surface layer of DFe concentrations, new measurements are shown in red dots (GEOVIDE voyage), while previous studies are displayed in black (Achterberg et al., 2018; Bergquist et al., 2007; Blain et al., 2004; Boye et al., 2006, 2003; de Jong et al., 2007; Gledhill et al., 1998; Hatta et al., 2015; Klunder et al., 2012; Laës et al., 2003; Martin et al., 1993; Measures et al., 2008; Mills et al., 2008; Mohamed et al., 2011; Nédélec et al., 2007; Nielsdóttir et al., 2009; Pohl et al., 2011; Rijkenberg et al., 2014; Sarthou et al., 2007, 2003; Sedwick et al., 2005; Ussher et al., 2013; Witter and Luther III, 1998; Wu and Boyle, 2002; Wu and Luther III, 1996, 1994; Wu et al., 2001).

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→ We also changed the text as we made a mistake in this section. Indeed, low DFe concentrations were previously measured in the central Irminger Sea. When we first wrote this part we considered stations that were closed to land likely impacted by sea-ice melting.

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Page 12 Lines 3-7: Among the four distinct basins described in this paper, the Irminger Sea exhibited the highest DFe concentrations within the surface waters (from 0 to 250 m depth) with values ranging from 0.23 to 1.3 nmol L<sup>-1</sup> for open-ocean stations. Conversely, low DFe concentrations were previously reported in the central Irminger Sea by Rijkenberg et al. (2014) (April-May, 2010) and Achterberg et al. (2018) (April-May and July-August, 2010) with DFe concentrations ranging from 0.11 to 0.15 and from ~ 0 to 0.14 nmol L<sup>-1</sup>, respectively (see supplementary material Fig. S4 and Table S2).

Page 10 Line 29: Please include an opening sentence, what you think is the reason (something similar to the last sentence). It is quite a step from Fe distribution to the original of water mass mixing.

→ We have added an opening sentence

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Page 12 Lines 9-11: Indeed, enhanced surface DFe concentrations measured during GEOVIDE in the Irminger Sea could be due to intense wind forcing events that would deepen the winter Z<sub>m</sub> down to the core of the Fe-rich LSW.

Page 11 Line 5: Explain what tip jets are!

→ We have added a definition

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Page 12 Lines 19-22: Moore (2003) and Piron et al. (2016) described low-level westerly jets centred northeast of Cape Farewell, over the Irminger Sea, known as tip jet events, whose structure depends upon the splitting occurring as the flow encounter the orographic features from Cape Farewell, and that are strong enough to induce deep convective mixing (Bacon et al., 2003; Pickart et al., 2003).

Page 11 Line 10ff: This process is called winter entrainment (Tagliabue et al. 2014). Rephrase sentence and delete the last one (You just repeat yourself).

→ We have modified the text as suggested

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Page 12 Lines 26-28: Such winter entrainment was likely the process involved in the vertical supply of DFe within surface waters fuelling the spring phytoplankton bloom with DFe values close to those found in LSW.

Page 11 Line 16: Also contaminated waters are introduced!

→ Yes, we completely agree. However, since this section is dedicated to atmospheric deposition we did not specify this.

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Page 11 Line 18: What is a stratification period? Be preciss!

→ We have changed the text

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Page 12 Line 32: "During the summer, when thermal stratification occurs, ..."

Page 11 Line 218ff: You can not compare the Mediterranean surface waters with MOW. Rewrite! DA1 and DFe behave entirely different in the water column (residence time, organic complexation,

concentrations, etc.), but both of them are likely to be scavenged from particles. So when a dust storm hits, both elements should decrease, do they actually do this in the water column of the Mediterranean sea. However, I am not too much surprised to see no DFe signal in the MOW. However, I suggest you have a look for DFe literature values from deep Mediterranean waters (GA04 is not available, a pity).

→ Yes, we agree that DAI and DFe behave differently in the water column depending on organic complexation and that they are both likely to be scavenged from particles. However, DAI and DFe originating from dust deposition are not scavenged by the same type of particles. Indeed, Wuttig et al. (2013) reported that after a single dust deposition event DAI loss rates was highly affected by the concentrations of biogenic particles while DFe was removed by sinking dust particles. The same authors highlighted that the following dust deposition event were likely inducing the dissolution of Fe from dust particles depending on the amounts of Fe-binding organic ligands. Therefore, both elements should not necessarily decrease.

→ We have changed the text as suggested

Page 13 Lines 2-14: After atmospheric deposition, the fate of Fe will depend on the nature of aerosols, vertical mixing, biological uptake and scavenging processes (Bonnet and Guieu, 2006; Wuttig et al., 2013). During GEOVIDE, MOW was observed from stations 1 to 29 between 1000 and 1200 m depth and associated with high dissolved aluminium (DAI, Menzel Barraqueta et al., 2018) concentrations (up to 38.7 nmol L<sup>-1</sup>), confirming the high atmospheric deposition in the Mediterranean region. In contrast to Al, no DFe signature was associated with MOW (Figs. 2 and 3). This feature was also reported in some studies (Hatta et al., 2015; Thuróczy et al., 2010), while others measured higher DFe concentrations in MOW (Gerringa et al., 2017; Sarthou et al., 2007). However, MOW coincides with the maximum Apparent Oxygen Utilization (AOU) and it is not possible to distinguish the MOW signal from the remineralisation one (Sarthou et al., 2007). On the other hand, differences between studies are likely originating from the intensity of atmospheric deposition and the nature of aerosols. Indeed, Wagener et al. (2010) highlighted that large dust deposition events can accelerate the export of Fe from the water column through scavenging. As a result, in seawater with high DFe concentrations and where high dust deposition occurs, a strong individual dust deposition event could act as a sink for DFe. It thus becomes less evident to observe a systematic high DFe signature in MOW despite dust inputs.

Page 12 Line 1: The entire section 4.1.3 is highly speculative. I agree elevated DFe in the Irminger Basin needs to come from somewhere, however, just looking at your Chl a data it is a very productive site, so presumably PFe concentrations are elevated as well, if so you should mention that, than it is just elevated remineralization and intense deep mixing during winter time that is responsible. However, you need to rewrite that section, to make it less speculative, look for existing data!

→ We have changed the text as suggested.

Page 13 Lines 16-33 and Page 14 Lines 1-16: As described in Section 3.1, the LSW exhibited increasing DFe concentrations from its source area, the Labrador Sea, toward the other basins with the highest DFe concentrations observed within the Irminger Sea, suggesting that the water mass was enriched in DFe either locally in each basin or during its flow path (Fig. 7). These DFe sources could originate from a combination of high export of PFe and its remineralisation in the mesopelagic area and/or the dissolution of sediment.

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The Irminger and Labrador Seas exhibited the highest averaged integrated TChl-a concentrations ( $98 \pm 32 \text{ mg m}^{-2}$  and  $59 \pm 42 \text{ mg m}^{-2}$ ) compared to the West European and Iceland Basins ( $39 \pm 10 \text{ mg m}^{-2}$  and  $53 \pm 16 \text{ mg m}^{-2}$ ), when the influence of margins was discarded. Stations located in the Irminger (stations 40-56) and Labrador (stations 63-77) Seas, were largely dominated by diatoms (>50% of phytoplankton abundances) and displayed the highest chlorophyllid- $\alpha$  concentrations, a tracer of senescent diatom cells, likely reflecting post-bloom condition (Tonnard et al., in prep.). This is in line with the highest POC export data reported by Lemaitre et al. (2018) in these two oceanic basins. This likely suggests that biogenic PFe export was also higher in the Labrador and Irminger Seas than in the West European and Iceland Basins. Although, Gourain et al. (2018) highlighted a higher biogenic contribution for particles located in the Irminger and Labrador Seas with relatively high PFe:PAI ratios ( $0.44 \pm 0.12 \text{ mol:mol}$  and  $0.38 \pm 0.10 \text{ mol:mol}$ , respectively) compared to particles from the West European and Iceland Basins ( $0.22 \pm 0.10$  and  $0.38 \pm 0.14 \text{ mol:mol}$ , respectively, see Fig. 6 in Gourain et al., 2018), they reported no difference in PFe concentrations between the four oceanic basins (see Fig. 12A in Gourain et al., 2018) when the influence of margins was discarded, which likely highlighted the remineralisation of PFe within the Irminger and Labrador Seas. Indeed, Lemaitre et al. (2017) reported higher remineralisation rates within the Labrador (up to  $13 \text{ mmol C m}^{-2} \text{ d}^{-1}$ ) and Irminger Seas (up to  $10 \text{ mmol C m}^{-2} \text{ d}^{-1}$ ) using the excess barium proxy (Dehairs et al., 1997), compared to the West European and Iceland Basins (ranging from 4 to  $6 \text{ mmol C m}^{-2} \text{ d}^{-1}$ ). Therefore, the intense remineralisation rates measured in the Irminger and Labrador Seas likely resulted in enhanced DFe concentrations within LSW.

Higher DFe concentrations were, however, measured in the Irminger Sea compared to the Labrador Sea and coincided with lower transmissometer values (i.e. 98.0-98.5% vs. >99%), thus suggesting a particle load of the LSW. This could be explained by the reductive dissolution of Newfoundland Margin sediments. Indeed, Lambelet et al. (2016) reported high dissolved neodymium (Nd) concentrations (up to  $18.5 \text{ pmol.kg}^{-1}$ ) within the LSW at the edge of the Newfoundland Margin ( $45.73^\circ\text{W}$ ,  $51.82^\circ\text{N}$ ) as well as slightly lower Nd isotopic ratio values relative to those observed in the Irminger Sea. They suggested that this water mass had been in contact with sediments approximately within the last 30 years (Charette et al., 2015). Similarly, during GA03, Hatta et al. (2015) attributed the high DFe concentrations in the LSW to continental margin sediments. Consequently, it is also possible that the elevated DFe concentrations from the three LSW branches which entered the West European and Iceland Basins and Irminger Sea was supplied through sediment dissolution (Measures et al., 2013) along the LSW pathway.

The enhanced DFe concentrations measured in the Irminger Sea and within the LSW were thus likely attributed to the combination of higher productivity, POC export and remineralisation as well as a DFe supply from reductive dissolution of Newfoundland sediments to the LSW along its flow path.

Page 12 Line 25: the elevated concentration on station 44, is not this just a single point?

➔ No, the elevated DFe concentrations at station 44 concerned three data points.

Page 12 Line 26ff: Replace "above" by "at", and what are i) sediment inputs (these are particles), and ii) intrusion of an Fe-rich water mass, please be more specific!

➔ We have changed the sentence as suggested and added precision.

Page 14 Lines 19-20: "... i) vertical diffusion from local sediment, ii) lateral advection of a water mass displaying enhanced DFe concentrations, and iii) local dissolution of Fe from particles."

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Page 12 Line 33: How often have you analyzed the samples below 2.500 m at site 44. For me this is just one outlier, the two other samples from cast 44 in Fig 8A are not that out of the range.

→ The full station 44 was analysed during two separated analytical sessions on the seaFAST SF-ICP-MS from different sampling bottles with a good agreement between results. Therefore, we do not think that this data point is an outlier.

Page 13 Line 10ff: Your argument is based on four data points, I could also put a straight line through, with a similar R2. However this entire paragraph is highly speculative! In an earlier paragraph you mention that DFe do not fingerprint different water masses, and now they do? You should remove this section!

→ We agree on the fact that the polynomial fitting could also be a linear fitting. However, with either a polynomial or a linear fitting on the 5 data points, the conclusion would be the same with apparently, the dissolution of Fe-rich particles.

→ We reformulated this section as we considered it too speculative.

Page 15 Lines 2-20: However, considering the short residence time of DFe and the circulation of water masses in the Irminger Sea, it is possible that instead of being attributed to one specific water mass, these enhanced DFe concentrations resulted from lateral advection of the deep waters. Figure 8B) shows the concentrations of both DFe and PFe for the mixing line between DSOW/PIW and ISOW at station 44 and considering 100% contribution of ISOW for the shallowest sample (2218 m depth) and of DSOW/PIW for the deepest (2915 m depth), as these were the main water masses. This figure shows increasing DFe concentrations as DSOW/PIW mixed with ISOW. In addition, Le Roy et al. (2018) reported for the GEOVIDE voyage at station 44 a deviation from the conservative behaviour of  $^{226}\text{Ra}$  reflecting an input of this tracer centred at 2500 m depth, likely highlighting diffusion from deep-sea sediments and coinciding with the highest DFe concentrations measured at this station. Although the transmissometer values were lower at the sediment interface than at 2500 m depth, Deng et al. (2018) reported a stronger scavenged component of the  $^{230}\text{Th}$  at the same depth range, likely suggesting that the mixture of water masses were in contact with highly reactive particles. If there is evidence that the enhanced DFe concentrations observed at station 44 coincided with lateral advection of water masses that were in contact with particles, the difference of behaviour between DFe and  $^{230}\text{Th}$  remains unsolved. The only parameter that would explain without any ambiguity such differences of behaviour between DFe and  $^{230}\text{Th}$  would be the amounts of Fe-binding organic ligands for these samples. Indeed, although PFe concentrations decreased from the seafloor to the above seawater, this trend would likely be explained by a strong vertical diffusion alone and not necessarily from the dissolution of particles that were laterally advected.

Therefore, the high DFe concentrations observed might be inferred from local processes as ISOW mixes with both PIW and DSOW with a substantial load of Fe-rich particles that might have dissolved in solution due to Fe-binding organic ligands.

Page 13 Line 22: unpublished sources? You need to explain that! Did you look through your Mn and Pb data, when they are also high, we talk about a hydrothermal input of trace metals.

→ We have changed the text for clarification

Page 15 Lines 22-33 and page 16 Lines 1-12: Hydrothermal activity was assessed over the Mid Atlantic Ridge, namely the Reykjanes Ridge, from stations 36 to 42. Indeed, within the interridge database (<http://www.interridge.org>), the Reykjanes Ridge is reported to have active hydrothermal

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sites that were either confirmed (Baker and German, 2004a; German et al., 1994; Olafsson et al., 1991; Palmer et al., 1995) close to Iceland or inferred (e.g. Chen, 2003; Crane et al., 1997; German et al., 1994; Sinha et al., 1997; Smallwood and White, 1998) closer to the GEOVIDE section as no plume was detected but a high backscatter was reported potentially corresponding to a lava flow. Therefore, hydrothermal activity at the sampling sites remains unclear with no elevated DFe concentrations or temperature anomaly above the ridge (station 38). However, enhanced DFe concentrations (up to  $1.5 \pm 0.22 \text{ nmol L}^{-1}$ , station 36, 2200 m depth) were measured east of the Reykjanes Ridge (Fig. 4). This could be due to hydrothermal activity and resuspension of sunken particles at sites located North of the section and transported through the ISOW towards the section (Fig. 7). Indeed, Achterberg et al. (2018) highlighted at  $\sim 60^\circ\text{N}$  and over the Reykjanes Ridge a southward lateral transport of an Fe plume of up to 250-300 km. In agreement with these observations, previous studies (e.g. Fagel et al., 1996; Fagel et al., 2001; Lackschewitz et al., 1996; Parra et al., 1985) reported marine sediment mineral clays in the Iceland Basin largely dominated by smectite (> 60%), a tracer of hydrothermal alteration of basaltic volcanic materials (Fagel et al., 2001; Tréguer and De La Rocha, 2013). Hence, the high DFe concentrations measured east of the Reykjanes Ridge could be due to a hydrothermal source and/or the resuspension of particles and their subsequent dissolution.

West of the Reykjanes Ridge, a DFe-enrichment was also observed in ISOW within the Irminger Sea (Figs. 4 and 7). The low transmissometer values within ISOW in the Irminger Sea compared to the Iceland Basin suggest a particle load. These particles could come from the Charlie Gibbs Fracture Zone (CGFZ,  $52.67^\circ\text{N}$  and  $34.61^\circ\text{W}$ ) and potentially Bight Fracture Zone (BFZ,  $56.91^\circ\text{N}$  and  $32.74^\circ\text{W}$ ) (Fig. 1) (Lackschewitz et al., 1996; Zou et al., 2017). Indeed, hydrographic sections of the northern valley of the CGFZ showed that below 2000 m depth the passage through the Mid-Atlantic Ridge was mainly filled with the ISOW (Kissel et al., 2009; Shor et al., 1980). Shor et al. (1980) highlighted a total westward transport across the sill, below 2000 m depth of about  $2.4 \times 10^6 \text{ m}^3 \text{ s}^{-1}$  with ISOW carrying a significant load of suspended sediment ( $25 \mu\text{g L}^{-1}$ ), including a 100-m-thick benthic nepheloid layer. It thus appears that the increase in DFe within ISOW likely came from sediment resuspension and dissolution as the ISOW flows across CGFZ and BFZ.

→ Note that for Pb, no particular hydrothermal signal was observed during GEOVIDE (Zurbrick et al., 2018). For Mn, data are analysed but not yet processed.

Page 14 Line 3ff: There are no elevated DFe values farther east from the ridge!

→ We have changed the text for clarification (see above). The DFe enrichment east of Reykjanes Ridge corresponded to the section on top of this sentence while further downstream corresponded to west of the Reykjanes Ridge.

Where is the CGFC and BFC. Questions over questions!

→ These two features are now added to Fig. 1

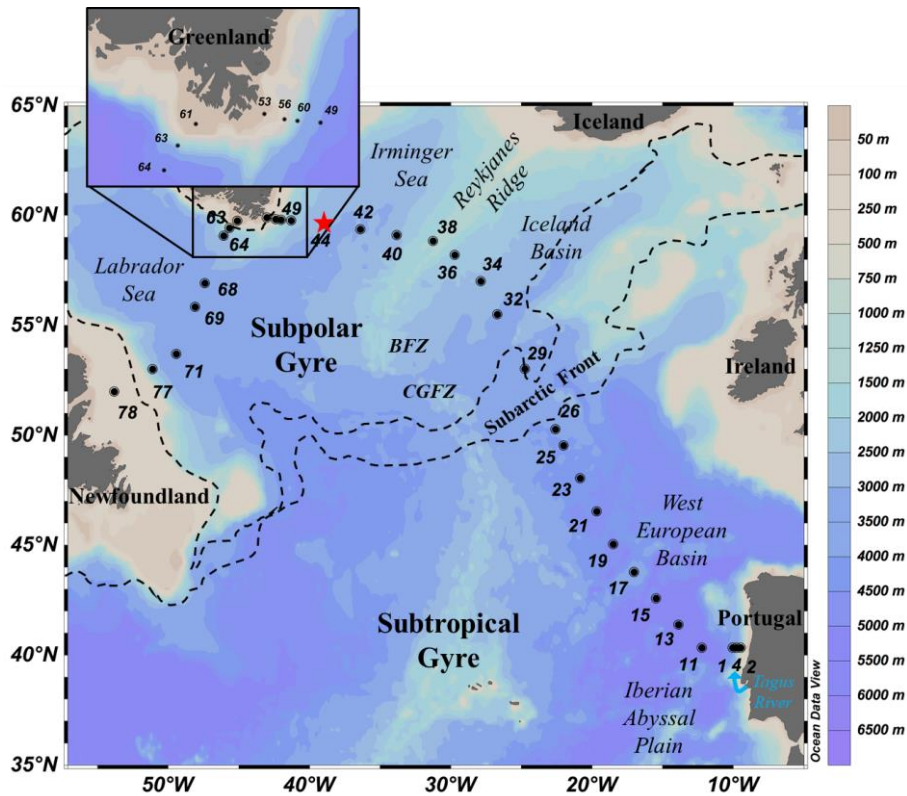
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**Figure 1: Map of the GEOTRACES GA01 voyage plotted on bathymetry as well as the major topographical features and main basins. Crossover station with GEOTRACES voyage (GA03) is shown as a red star. (Ocean Data View (ODV) software, version 4.7.6, R. Schlitzer, <http://odv.awi.de>, 2016). BFZ: Bight Fracture Zone, CGFZ: Charlie-Gibbs Fracture Zone.**

Page 14 Line 13ff: I am confused. Do we talk about station 40 and 1.75nM at 1500 m, this is a single high value for me, and not located in ISOW waters.

→ Yes, we talk about station 40 (one point) and station 42 (three points) that are all located in the ISOW (see Fig. 7).

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Page 14 Line 26ff: The DFe/DAI ratio in seawater can not compared with the Fe/Al ratio of dust particles. Both elements have different fractional solubility's. So the ratio is always different! Remove!

→ We agree with the reviewer and have changed the text as suggested and have added some information

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Page 16 lines 25-28: Our SML DFe inventories were about three times higher at station 1 (~ 1 nmol L<sup>-1</sup>) than those calculated during the GA03 voyage (~ 0.3 nmol L<sup>-1</sup>, station 1) during which atmospheric



deposition were about one order of magnitude higher (Shelley et al., 2018; Shelley et al., 2015), the atmospheric source seemed to be minor.

Page 15 Line 1: Remove most of them does not add to the story!

→ We agree and have removed most of them.

Page 16 Line 32 and Page 17 Lines 1-2: Many types of industry (e.g. heavy metallurgy, ore processing, chemical industry) release metals including Fe, which therefore result in high levels recorded in surface sediments, suspended particulate matter, water and organisms in the lower estuary (Santos-Echeandia et al., 2010).

Page 15 Line 5: What do you mean with “..below ground biomass..” In general I do not understand, why you excluded sediments, that could be an additional source.

→ We did not intend to exclude the sediment source and have change the text for clarification.

Page 16 lines 28-30 Consequently, the Tagus River appears to be the most likely source responsible for these enhanced DFe concentrations, either as direct input of DFe or indirectly through Fe-rich sediment carried by the Tagus River and their subsequent dissolution.

Page 15 Line 14ff: Fronts refer to temperature and salinity changes in surface waters, such as the Polar Front, not in the water column. Call it different; just use the term “fresh water lens”. Why multi-year-sea ice?

→ We have changed the text as suggested

Page 17 Lines 9-10: The presence of this freshwater lens suggests that sediment derived enrichment to these surface waters was unlikely.

→ We talked about multiyear sea ice because of drainage processes and the release of brines (see below)

Page 15 Line 18ff: But glacial sources and land ice sheet is the same, just call, it “ . . .freshwater induced by meteoric water and sea-ice melt.” Than all is clear.

→ We have made the correction as suggested (Page17 Lines 10-11)

Page 15 Line 27: Where do get the sea-ice fractions from, and explain how it works, include references! And what have brines to do with it, either ice forms or not! Brines are not part of your story, so far I can tell. Brines always from when sea-ice is formed, or in the desert by evaporation. And in line 31 you switch back to sea-ice formation, please stay with that term.

→ We have included a section in the method on how these fractions were calculated.

Page 5 Lines 23-32, Page 6 Lines 1-9: We separated the mass contributions to samples from stations 53, 61 and 78 in Sea-Ice Melt (SIM) Meteoric Water (MW) and saline seawater inputs using the procedure and mass balance calculations that are fully described in Benetti et al. (2016) (Fig. 5D), E) and F)). Hereafter, we describe briefly the principle. We considered two types of seawater, namely the Atlantic Water (AW) and the Pacific Water (PW). After estimating the relative proportions of AW ( $f_{AW}$ ) and PW ( $f_{PW}$ ) and their respective salinity and  $\delta^{18}O$  affecting each samples, the contribution of SIM and MW can be determined using measured salinity ( $S_m$ ) and  $\delta^{18}O$  ( $\delta O_m^{18}$ ). The mass balance calculations are presented below:

$$f_{AW} + f_{PW} + f_{MW} + f_{SIM} = 1 \text{ (eq.1)}$$

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$$f_{AW} \cdot S_{AW} + f_{PW} \cdot S_{PW} + f_{MW} \cdot S_{MW} + f_{SIM} \cdot S_{SIM} = S_m \text{ (eq.2)}$$

$$f_{AW} \cdot \delta O_{AW}^{18} + f_{PW} \cdot \delta O_{PW}^{18} + f_{MW} \cdot \delta O_{MW}^{18} + f_{SIM} \cdot \delta O_{SIM}^{18} = \delta O_m^{18} \text{ (eq.3)}$$

where  $f_{AW}$ ,  $f_{PW}$ ,  $f_{MW}$ ,  $f_{SIM}$  are the relative fraction of AW, PW, MW, and SIM. To calculate the relative fractions of AW, PW, MW and SIM we used the following end-members:  $S_{AW} = 35$ ,  $\delta O_{AW}^{18} = +0.18\text{‰}$  (Benetti et al., 2016);  $S_{PW} = 32.5$ ,  $\delta O_{PW}^{18} = -1\text{‰}$  (Cooper et al., 1997; Woodgate and Aagaard, 2005);  $S_{MW} = 0$ ,  $\delta O_{MW}^{18} = -18.4\text{‰}$  (Cooper et al., 2008);  $S_{SIM} = 4$ ,  $\delta O_{SIM}^{18} = +0.5\text{‰}$  (Melling and Moore, 1995).

In Figure 5 D), E) and F), negative sea-ice fractions indicated a net brine release while positive sea-ice fractions indicated a net sea-ice melting. Note that for stations over the Greenland Shelf, we assumed that the Pacific Water (PW) contribution was negligible for the calculations, supported by the very low PW fractions found at Cape Farewell in May 2014 (see Figure B1 in Benetti et al., 2017), while for station 78, located on the Newfoundland shelf, we used nutrient measurements to calculate the PW fractions, following the approach from Jones et al. (1998) (the data are published in Benetti et al., 2017).

→ Regarding brines, they can originate from two different processes: either as a result of multiyear sea-ice melting or during sea-ice formation. Indeed, during the early melting season, multiyear sea-ice has a higher porosity and gravitational drainage of brine occur. These two processes of brine release might lead to different TM signatures in brine originating from sea-ice formation and brine originating from early melting of multiyear sea-ice (Petrich and Eicken, 2010; Wadhams, 2000).

Page 15 Line 33: But brines usually sink, because they are heavier than the surrounding water!!! It is really hard to follow your argumentation here.

We agree with the reviewer in the fact that brines sink due to higher density. However, after reaching neutral buoyancy, they will stop sinking.

Page 16 Line 11: You have to explain how you produced these numbers, a citation in an earlier paragraph is not enough!

→ We have included a section in the method on how these fractions were calculated (see above)

Page 16 Line 15: How do you lose a sample! Generally first you talk about the contribution of MW and then you switch to biological uptake of DFe, that in the same paragraph? You lose the reader here; this entire section needs an overhaul.

→ We have reorganised this section.

Page 18 Lines 3-18: Surface waters (from 0 to ~ 100 m depth) from station 53 and 61 were characterized by high MW fractions (ranging from 8.3 to 7.4% and from 7.7 to 7.3%, respectively, from surface to ~100 m depth, Figs. 5D and E). These high MW fractions were both enriched in PFe and DFe (except station 53 for which no data was available close to the surface) compared to seawater located below 50 m depth, thus suggesting a MW source. These results are in line with previous observations, which highlighted strong inputs of DFe from a meteoric water melting source in Antarctica (Annett et al., 2015). Although the ability of MW from Greenland Ice Sheet and runoffs to deliver DFe and PFe to surrounding waters has previously been demonstrated (Bhatia et al., 2013; Hawkings et al., 2014; Schroth et al., 2014; Statham et al., 2008), both Fe fractions were lower at the sample closest to the surface, then reached a maximum at ~ 50 m depth and decreased at ~ 70 m depth, for station 61 (Fig. 4D). The surface DFe depletion was likely explained by phytoplankton uptake, as indicated by the high TChl- $\alpha$  concentrations (up to 6.6 mg m<sup>-3</sup>) measured from surface to

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about 40 m depth, drastically decreasing at ~ 50 m depth to 3.9 mg m<sup>-3</sup> (Fig. 4D). Hence, it seemed that meteoric water inputs from the Greenland Margin likely fertilized surface waters with DFe, enabling the phytoplankton bloom to subsist. The profile of PFe can be explained by two opposite plausible hypotheses: 1) MW inputs did not released PFe, as if it was the case, one should expect higher PFe concentrations at the surface (~25 m depth) than the one measured at 50 m depth due to both the release from MW and the assimilation of DFe by phytoplankton 2) MW inputs can release PFe in a form that is directly accessible to phytoplankton with subsequent export of PFe as phytoplankton died. The latter solution explains the PFe maximum measured at ~ 50 m depth and is thus the most plausible.

Page 16 Line 32: “.. decreasing from surface to depth.” Which depth, down to the bottom in 400 m depth? Be precise

→ We have added this precision.

Page 18 Lines 21-22: Newfoundland shelf waters (station 78) were characterized by high MW fractions (up to 7%), decreasing from surface to 200 m depth (~2%).

Page 17 Line 15-25: What has the tropical and subtropical North Atlantic to do with your work! I assume very little, please delete or at least reduce the text.

→ We agree with the reviewer and removed the part on tropical North Atlantic.

Page 18 Lines 32-33 and Page 19 Lines 1-4: On a regional scale, the North Atlantic basin receives the largest amount of atmospheric inputs due to its proximity to the Saharan Desert (Jickells et al., 2005), yet even in this region of high atmospheric deposition, inputs are not evenly distributed. Indeed, aerosol Fe loading measured during GEOVIDE (Shelley et al., 2017) were much lower (up to four orders of magnitude) than those measured during studies from lower latitudes in the North Atlantic (e.g. Baker et al., 2013; Buck et al., 2010; and for GA03, Shelley et al., 2015), but atmospheric inputs could still be an important source of Fe in areas far from land.

Page 17 Line 30: I would rather suggest to say: “Shelley et al. concluded that. . .” because without any trajectories here I can check, and more or less all this work was already published.

→ We have removed this part.

Page 18 Line 13: Do you mean DOM? Or organic material OM. However, you talk about DOM for 7 lines, and then you don't have the data. Once sentence should be enough to point out the importance of DOM.

→ We have removed this part.

Page 18 Line 16: This entire paragraph is very poor! It is interesting to compare elemental ratios of seawater with the soluble fraction of dust. But the reasoning here “. . . whether there was enough atmospheric input to sustain the SML DFe concentrations. . .” without any flux numbers, residence times is unscientific. Even more strange, at the end of the paragraph you don't even say, whether there is enough or not. Similar to the above, this needs serious work to make it worthwhile reading. There is too much hand waving, and too few data, sorry! I suggest you look up the actual flux numbers and then compare them with your data.

→ We agree with the reviewer and removed this section to replace it by Turnover Times relative to Atmospheric Deposition (TTADs) as defined in Guieu et al. (2014).

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Page 19 Lines 5-17: In an attempt to estimate whether there was enough atmospheric input to sustain the SML DFe concentrations, we calculated Turnover Times relative to Atmospheric Deposition (TTADs, Guieu et al., 2014). To do so, we made the following assumptions: 1) the aerosol concentrations are a snapshot in time but are representative of the study region, 2) the aerosol solubility estimates based on two sequential leaches are an upper limit of the aerosol Fe in seawater and 3) the water column stratified just before the deposition of atmospheric inputs, so MLD DFe will reflect inputs from above. Thus, the TTADs were defined as the integrated DFe concentrations in the SML for each station divided by the contribution of soluble Fe contained in aerosols averaged per basin to the water volume of the SML. Although, TTADs were lower in the West European and Iceland Basins with an average of  $\sim 9 \pm 3$  months compared to other basins ( $7 \pm 2$  years and  $5 \pm 2$  years for the Irminger and Labrador Seas, respectively) (Fig. 6) they were about three times higher than those reported for areas impacted by Saharan dust inputs ( $\sim 3$  months, Guieu et al., 2014). Therefore, the high TTADs measured in the Irminger and Labrador Seas and ranging from 2 to 15 years provided further evidence that atmospheric deposition were unlikely to supply Fe in sufficient quantity to be the main source of DFe (see Sections 4.2.1 and 4.3.2) while in the West European and Iceland Basins they played an additional source, perhaps the main source of Fe especially at station 36 which displayed TTAD of 3 months.

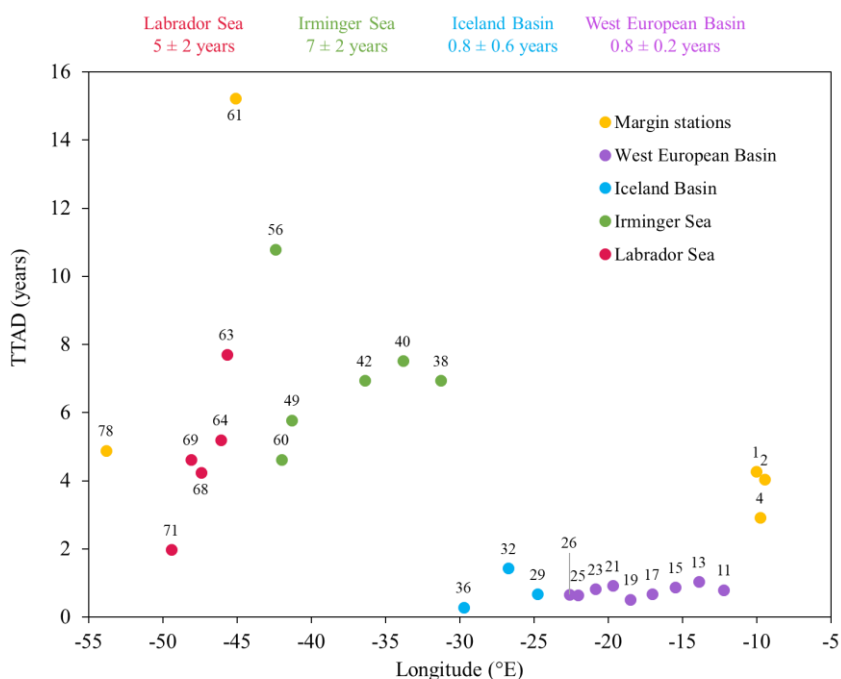


Figure 6: Plot of dissolved Fe (DFe) Turnover Times relative to Atmospheric Deposition (TTADs) calculated from soluble Fe contained in aerosols estimated from a two-stage sequential leach (UHP water, then 25% HAC, Shelley et al., this issue). Note that numbers on top of data points represent station numbers and that the colour coding refers to different region with in yellow, margin stations; in purple, the West European Basin; in blue, the Iceland Basin; in green, the Irminger Sea and in red,

the Labrador Sea. The numbers on top of the plot represent TTADs averaged for each oceanic basin and their standard deviation.

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Page 19 Line 5ff: replace “on” by “in”. And which similar pattern followed the station. Be precise! Sentence stating in Line 6 makes no sense, please rewrite!

→ We have corrected the text and rephrased the next sentence as suggested.

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Page 19 Lines 20-23: DFe concentration profiles from all coastal stations (stations 2, 4, 53, 56, 61 and 78) are reported in Figure 5. To avoid surface processes, only depths below 100 m depth will be considered in the following discussion. DFe and PFe followed a similar pattern at stations 2, 53, 56, and 78 with increasing concentrations towards the sediment, suggesting that either the sources of Fe supplied both Fe fractions (dissolved and particulate) or that PFe dissolution from sediments supplied DFe.

Page 19 Line 11: What has the composition of sediments to do with your PFe value? Nothing. . .

→ We have changed the text for clarification.

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Page 19 Lines 25-28: DFe:PFe ratios ranged from 0.01 (station 2, bottom sample) to 0.27 (station 4, ~ 400 m depth) mol:mol with an average value of  $0.11 \pm 0.07$  mol:mol (n = 23, Table 4), highlighting a different behaviour of Fe between margins. This could be explained by the different nature of the sediments and/or different sediment conditions (e.g. redox, organic content).

Page 19 Line 15: “Intermediate behavior” of what? And then Chla? This paragraph is very hard to follow, what is the message you want to bring across, I can’t tell!

→ We removed this sentence for clarification.

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Page 19 Lines 28-33: Based on particulate and dissolved Fe and dissolved Al data (Gourain et al., 2018; Menzel Barraqueta et al., 2018, Table 4), three main different types of margins were reported (Gourain et al., 2018) with the highest lithogenic contribution observed at the Iberian Margin (stations 2 and 4) and the highest biogenic contribution at the Newfoundland Margin (station 78). These observations are consistent with higher TChl- $\alpha$  concentrations measured at the Newfoundland Margin and to a lesser extent at the Greenland Margin and the predominance of diatoms relative to other functional phytoplankton classes at both margins (Tonnard et al., in prep.).

Page 19 Line 26: “respect”? I respect you as a person, but samples usually don’t respect anything?

→ We have changed the word “respected” by “followed”.

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Page 19 Line 30ff: How do you know its manganese oxide, just use particulate Mn. And why you do not include the transmissometer data. That is what you wanted to show, or not that resuspended sediments control you particulate fraction.

→ We agree with the reviewer in the way that we are not sure these are manganese oxides as they were estimated as the fraction from the PMn that was not originating from a lithogenic fraction using Mn:Ti UCC ratio. Therefore, a biological source or a co-precipitation source without oxidation were not considered. We thus agree with the reviewer and we have changed the MnOx data towards PMn data in the PCA.

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→ We did not include the transmissometer data as we do not have true values for all samples and used the interpolated data.

Page 20 Line 3ff: You did not do a PCA for dFe, so how can you be sure that the dim1 controls DFe? I cannot follow.

→ Before performing the PCA, a huge number of variables were considered and we only kept the one that were correlated to DFe to build the PCA.

→ We have changed the text for clarification.

Page 20 Lines 6-16; Samples associated with high levels of particles (transmissometer < 99%) and below 500 m depth displayed a huge variability in DFe concentrations. From the entire dataset, 66 samples (~13% of the entire dataset) followed this criterion with 3 samples from the Iberian Margin (station 4), 14 samples from the West European Basin (station 1), 4 samples from the Iceland Basin (stations 29, 32, 36 and 38), 43 samples from the Irminger Sea (stations 40, 42, 44, 49 and 60) and 2 samples from the Labrador Sea (station 69). To determine which parameter was susceptible to explain the variation in DFe concentrations in these nepheloid layers, a Principal Component Analysis (PCA) on these samples. The input variables of the PCA were the particulate Fe, Al, and particulate manganese (PMn) (Gourain et al., 2018), the DAI (Menzel Barraqueta et al., 2018) and the Apparent Oxygen Utilization (AOU) and were all correlated to DFe concentrations explaining all together 93% of the subset variance (Fig. 11). The first dimension of the PCA was represented by the PAI, PFe and PMn concentrations and explained 59.5% of the variance, while the second dimension was represented by the DAI and the AOU parameters, explaining 33.2% of the variance. The two sets of variables were nearly at right angle from each other, indicating no correlation between them.

Page 20 Line 24: You did not show any evident information that would suggest that DFe is controlled by OM (you did not even show any data) and PMn. Like the others this paragraph needs more work!

→ We have reorganised the section and added complementary information.

Page 20 Lines 6-34, Page 21 lines 1-6:

#### 4.4.2 Nepheloid layers:

Samples associated with high levels of particles (transmissometer < 99%) and below 500 m depth displayed a huge variability in DFe concentrations. From the entire dataset, 63 samples (~13% of the entire dataset) followed this criterion with 14 samples from the West European Basin (station 1), 4 samples from the Iceland Basin (stations 29, 32, 36 and 38), 43 samples from the Irminger Sea (stations 40, 42, 44, 49 and 60) and 2 samples from the Labrador Sea (station 69). To determine which parameter was susceptible to explain the variation in DFe concentrations in these nepheloid layers, a Principal Component Analysis (PCA) on these samples. The input variables of the PCA were the particulate Fe, Al, and particulate manganese (PMn) (Gourain et al., 2018), the DAI (Menzel Barraqueta et al., 2018) and the Apparent Oxygen Utilization (AOU) and were all correlated to DFe concentrations explaining all together 93% of the subset variance (see supplementary material Fig. S6). The first dimension of the PCA was represented by the PAI, PFe and PMn concentrations and explained 59.5% of the variance, while the second dimension was represented by the DAI and the AOU parameters, explaining 33.2% of the variance. The two sets of variables were nearly at right angle from each other, indicating no correlation between them.

The variations in DFe concentrations measured in bottom samples from stations 32, 36 (Iceland Basin), 42 and 44 (Irminger Sea) and 69 (Labrador Sea) were mainly explained by the first dimension of the PCA (see supplementary material Fig. S6). Therefore, samples characterized by the lowest DFe concentrations (stations 32 and 69) were driven by particulate Al and Mn concentrations and resulted in an enrichment of Fe within particles. These results are in agreement with previous

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studies showing that the presence of Mn within particles can induce the formation of Fe-Mn oxides, contributing to the removal of Fe and Mn from the water column (Kan et al., 2012; Teng et al., 2001).

Low DFe concentrations (bottom samples from stations 42 and 1) were linked to DAL inputs and associated with lower AOU values. The release of Al has previously been observed from Fe and Mn oxide coatings on resuspended sediments under mildly reducing conditions (Van Beusekom, 1988). Conversely, higher DFe concentrations were observed for stations 44 and 49 and to a lesser extent station 60 coinciding with low DAL inputs and higher oxygen levels. This observation challenges the traditional view of Fe oxidation with oxygen, either abiotically or microbially induced. Indeed, remineralisation can decrease sediment oxygen concentrations, promoting reductive dissolution of PFe oxyhydroxides to DFe that can then diffuse across the sediment water interface as DFe(II) colloids (Homoky et al., 2011). Such processes will inevitably lead to rapid Fe removal through precipitation of nanoparticulate or colloidal Fe (oxyhydr)oxides, followed by aggregation or scavenging by larger particles (Boyd and Ellwood, 2010; Lohan and Bruland, 2008) unless complexation with Fe-binding organic ligands occurs (Batchelli et al., 2010; Gerringa et al., 2008). There exist, however, another process that is favoured in oxic benthic boundary layers (BBL) with low organic matter degradation and/or low Fe oxides, which implies the dissolution of particles after resuspension, namely the non-reductive dissolution of sediment (Homoky et al., 2013; Radic et al., 2011). In addition, these higher oxygenated samples were located within DSOW, which mainly originate (75% of the overflow) from the Nordic Seas and the Arctic Ocean (Tanhua et al., 2005), in which the ultimate source of Fe was reported by Klunder et al. (2012) to come from Eurasian river waters. The major Arctic rivers were highlighted by Slagter et al. (2017) to be a source of Fe-binding organic ligands that are then further transported via the TPD across the Denmark Strait. Hence, the enhanced DFe concentrations measured within DSOW might result from Fe-binding organic ligand complexation that were transported to the deep ocean as DSOW formed rather than the non-reductive dissolution of sediment.

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Page 20 Line 27ff: Include "some" in front of "maxima, Please tell me the difference between "the relationship between DFe and biological uptake" and "Did DFe concentrations potentially limit phytoplankton growth?" This sounds to me very connected with each other! Why not discussing that in the follow up paper?

We have corrected the first sentence as suggested and re-wrote the end of the paragraph for clarification. Note that we wanted to keep this discussion in this paper as it summarises the different processes discussed in this MS.

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Page 21 Lines 8-11: Overall, almost all the stations from the GEOVIDE voyage displayed DFe minima in surface water associated with some maxima of TChl-*a* (Fig. 3). In the following section, we specifically address the question of whether DFe concentrations potentially limit phytoplankton growth. Note that macronutrients and DFe limitations relative to phytoplankton functional classes are dealt in Tonnard et al. (in prep.).

Page 20 Line 31: Include mean or average Plus standard deviation

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→ We have included the average and SD and corrected few mistakes.

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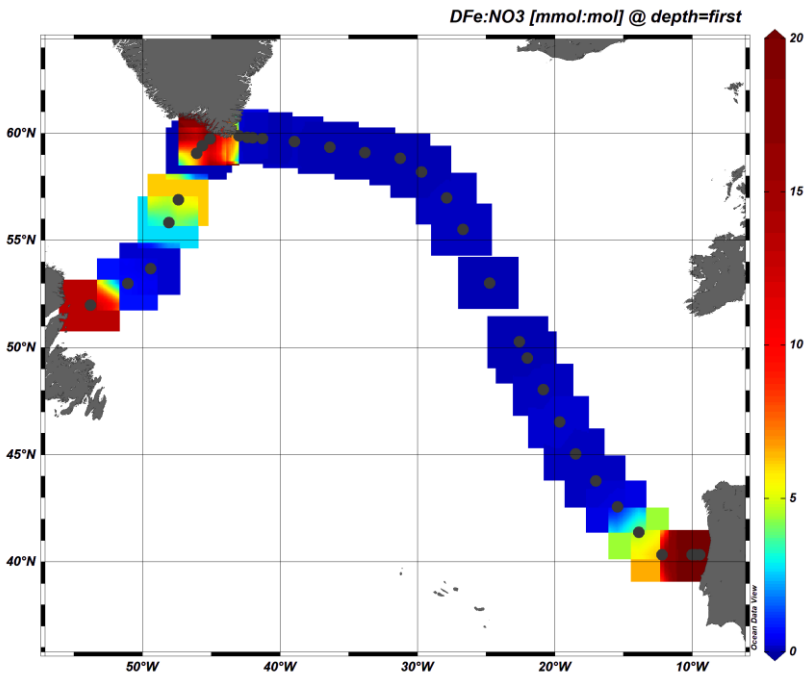
Page 21 Lines 13-14: The DFe:NO<sub>3</sub><sup>-</sup> ratios in surface waters varied from 0.02 (station 36) to 38.6 (station 61) mmol:mol with an average of 5 ± 10 mmol:mol (see supplementary material Fig. S7).

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Page 21 Line 4: Please include numbers! Following the text in the paragraph, it is very hard to follow, you jump between F:N ratios, water masses and Chl a. Try to keep it short and weed out unnecessary details. Otherwise you will lose the reader!

→ We have changed the text accordingly and added a surface map of DFe:NO<sub>3</sub><sup>-</sup> ratios.



Page 21 Lines 12--33 Page 22 Lines 1-34, Page 23 Lines 1-3: A key determinant for assessing the significance of a DFe source is the magnitude of the DFe:macronutrient ratio supplied, since this term determines to which extent DFe will be utilised. The DFe:NO<sub>3</sub><sup>-</sup> ratios in surface waters varied from 0.02 (station 36) to 38.6 (station 61) mmol:mol with an average of  $5 \pm 10$  mmol:mol (see supplementary material Fig. S7). Values were typically equal or lower than 0.28 mmol mol<sup>-1</sup> in all basins except at the margins and at stations 11, 13, 68, 69 and 77. The low nitrate concentrations observed at the eastern and western Greenland and Newfoundland Margins reflected a strong phytoplankton bloom which had reduced the concentrations as highlighted by the elevated integrated TChl-*a* concentrations ranging from 129.6 (station 78) to 398.3 (station 61) mg m<sup>-2</sup>. At the Iberian Margin, they likely reflected the influence of the N-limited Tagus River (stations 1, 2 and 4) with its low TChl-*a* integrated concentrations that ranged from 31.2 (station 1) to 46.4 (station 4) mg m<sup>-2</sup>. The high DFe:NO<sub>3</sub><sup>-</sup> ratios determined at those stations, which varied from 13.4 (station 78) to 38.6 (station 61) mmol:mol, suggested that waters from these areas, despite having the lowest NO<sub>3</sub><sup>-</sup> concentrations, were relatively enriched in DFe compared to waters from Iceland Basin and Irminger Sea.

In our study, DFe:NO<sub>3</sub><sup>-</sup> ratios displayed a gradient from the West European Basin to Greenland (supplementary material S7 and S8). This trend only reverses when the influence of Greenland was encountered, as also observed by Painter et al. (2014). The remineralisation of organic matter is a

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major source of macro and micronutrients in subsurface waters (from 50 to 250 m depth). Remineralisation is associated with the consumption of oxygen and therefore, Apparent Oxygen Utilization (AOU) can provide a quantitative estimate of the amount of material that has been remineralised. While no relationship was observed below 50 m depth for  $\text{NO}_3^-$  or DFe and AOU considering all the stations, a significant correlation was found in the Subpolar gyre when removing the influence of margins (stations 29-49, 56, 60, 63-77) ( $\text{AOU} = 3.88 \text{ NO}_3^- - 39.32$ ,  $R^2=0.79$ ,  $n=69$ ,  $p$ -value  $< 0.001$ ). This correlation indicates that remineralisation of Particulate Organic Nitrogen (PON) greatly translates into Dissolved Inorganic Nitrogen (DIN) and that  $\text{NO}_3^-$  can be used as a good tracer for remineralisation in the studied area. Within these Subpolar gyre waters, there was a significant correlation between DFe and AOU ( $\text{AOU} = 22.6 \text{ DFe}$ ,  $R^2=0.34$ ,  $n=53$ ,  $p$ -value  $< 0.001$ ). The open-ocean stations from Subpolar gyre also exhibited a good linear correlation between DFe and  $\text{NO}_3^-$  ( $R^2=0.42$ ,  $n=51$ ,  $p$ -value  $< 0.05$ ). The slope of the relationship, representing the typical remineralisation ratio, was  $R_{\text{Fe:N}} = 0.07 \pm 0.01 \text{ mmol mol}^{-1}$ . The intercept of the regression line was  $-0.4 \pm 0.2 \text{ nmol L}^{-1}$ , reflecting possible excess of preformed  $\text{NO}_3^-$  compare to DFe in these water masses. These significant correlations allow us to use the  $\text{Fe}^*$  tracer to assess where DFe concentrations potentially limit phytoplankton growth by subtracting the contribution of organic matter remineralisation from the dissolved Fe pool, as defined by Rijkenberg et al. (2014) and Parekh et al. (2005) for  $\text{PO}_4^{3-}$ , and modified here for  $\text{NO}_3^-$  as follow:

$$\text{Fe}^* = [\text{DFe}] - R_{\text{Fe:N}} \times [\text{NO}_3^-] \text{ (eq. 4)}$$

where  $R_{\text{Fe:N}}$  refers to the average biological uptake ratio Fe over nitrogen, and  $[\text{NO}_3^-]$  refers to nitrate concentrations in seawater. Although, we imposed a fixed biological  $R_{\text{Fe:N}}$  of  $0.05 \text{ mmol mol}^{-1}$ , it is important to note that the biological uptake ratio of  $\text{DFe}:\text{NO}_3^-$  is not likely to be constant. Indeed, this ratio has been found to range from 0.05 to 0.9  $\text{mmol mol}^{-1}$  depending on species (Ho et al., 2003; Sunda and Huntsman, 1995; Twining et al., 2004). The ratio we choose is thus less drastic to assess potential Fe limitation and more representative of the average biological uptake of DFe over  $\text{NO}_3^-$  calculated for this study (i.e.  $R_{\text{Fe:N}} = 0.07 \pm 0.01 \text{ mmol mol}^{-1}$ , for Subpolar waters). Negative values of  $\text{Fe}^*$  indicate the removal of DFe that is faster than the input through remineralisation or external sources and positive values suggest input of DFe from external sources (Fig. 7). Consequently, figure 7 shows that phytoplankton communities with very high Fe requirements relative to  $\text{NO}_3^-$  ( $R_{\text{Fe:N}} = 0.9$ ) will only be able to grow above continental shelves where there is a high supply of DFe as previously reported by Nielsdóttir et al. (2009) and Painter et al. (2014). All these results are corroborating the importance of the Tagus River (Iberian Margin, see section 4.2.1), glacial inputs in the Greenland and Newfoundland Margins (see section 4.2.2) and to a lesser extent atmospheric inputs (see section 4.2.3) in supplying Fe with  $\text{Fe}:\text{N}$  ratios higher than the average biological uptake/demand ratio. Figure 7 (see also supplementary material S7, S8, S9 and S10) also highlights the Fe limitation for the low-Fe requirement phytoplankton class ( $R_{\text{Fe:N}} = 0.05$ ) within the Iceland Basin, Irminger and Labrador Seas. The Fe deficiency observed in surface waters ( $> 50 \text{ m}$  depth) from the Irminger and Labrador Seas might be explained by low atmospheric deposition for the ICPMW and the LSW (Shelley et al., 2017). Low atmospheric Fe supply and sub-optimal  $\text{Fe}:\text{N}$  ratios in winter overturned deep water could favour the formation of the High-Nutrient, Low-Chlorophyll (HNLC) conditions. The West European Basin, despite exhibiting some of the highest  $\text{DFe}:\text{NO}_3^-$  ratios within surface waters (see supplementary material Fig. S8), displayed the strongest Fe-depletion from 50 m depth down to the bottom, suggesting that the main source of Fe was coming from dust deposition and/or riverine inputs.

Similarly as for the West European Basin, the pattern displayed in the surface map of  $\text{DFe}:\text{NO}_3^-$  ratios (supplementary material S8) extended to about 50 m depth, after which the trend reversed (Fig. 7

and supplementary material Fig. S7). Below 50 m depth, the Fe\* tracer (Fig. 7) was positive in the Irminger Sea and overall negative in the other basins. In the Irminger Sea positive Fe\* values were likely the result of the winter entrainment of Fe-rich LSW (see section 4.2.1) coinciding with high remineralised carbon fluxes in this area (station 44; Lemaître et al., 2017) (see section 4.2.2). The largest drawdown in DFe:NO<sub>3</sub><sup>-</sup> ratios was observed between stations 34 and 38 and was likely due to the intrusion of the ICSPMW, this water mass exhibiting low DFe and high in NO<sub>3</sub><sup>-</sup> (from 7 to 8 μmol L<sup>-1</sup>) concentrations. Similarly, the SAIW exhibited high NO<sub>3</sub><sup>-</sup> concentrations. Both the ICSPMW and the SAIW sourced from the NAC. The NAC as it flows along the coast of North America receives atmospheric depositions from anthropogenic sources (Shelley et al., 2017; 2015) which deliver high N relative to Fe (Jickells and Moore, 2015) and might be responsible for the observed ranges.

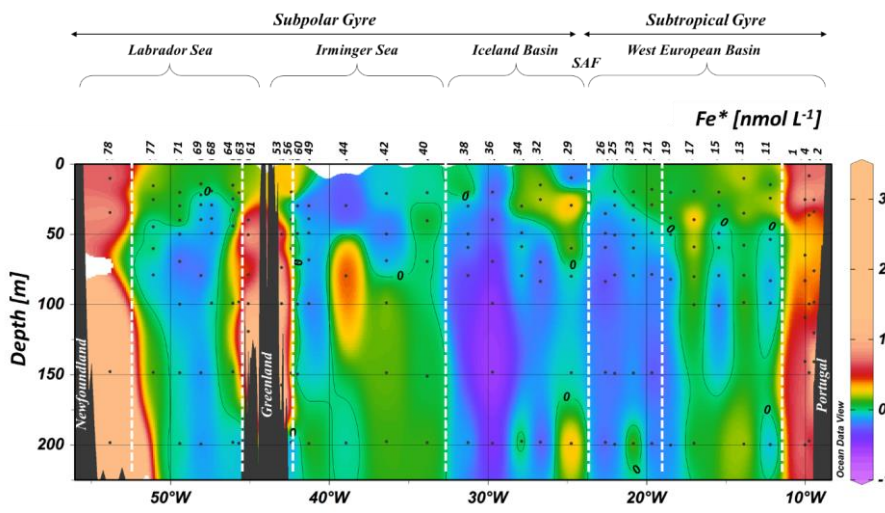
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Page 21 Line 20ff: Can you explain to me why you calculate Fe\* for the entire water body (Fig. 13) and explain DFe limitation of the phytoplankton community. They live in the first 100-200 m. Same fro Fig.14.

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- ➔ We calculated the Fe\* for the entire water body as water mass circulation and/or processes such as deep convection/upwelling, ... can homogenized deep water masses with surface water masses. Thus, looking at DFe:NO<sub>3</sub><sup>-</sup> ratios in these water masses appeared for us to be as important as just looking at the surface where phytoplankton live.
- ➔ However, we understand the reviewer's opinion and decided to restrict this section to the top 250 meters. Consequently, we did a new plot for Fig. 13 with only the upper water column and removed Fig. 14.

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Reading the last sentence of the section on page 22 "However, atmospheric loading (and especially Fe) was higher within the subtropical gyre than elsewhere in the GEOVIDE section mainly due to the proximity to mineral dust source (i.e. the Sahara Desert)." I feel I am still stuck in the Atmospheric chapter. Please shorten the paragraph and just say what you can prove with data.

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- ➔ We have removed the last two sentences to shorten the paragraph.

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Page 22 Line 14ff: The entire conclusion needs an overhaul!

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→ We have modified the conclusion to be more specific.

Page 23 Lines 5-31 Page 24 Lines 1-2: The DFe concentrations measured during this study were in good agreement with previous studies that spanned the West European Basin. However, within the Irminger Basin the DFe concentrations measured during this study were up to 3 times higher than the ones measured by Rijkenberg et al. (2014) in deep waters (> 1000 m depth) that was likely explained by the different water masses encountered (i.e. the Polar Intermediate Water, ~ 2800 m depth) and by a stronger signal of the Iceland Scotland Overflow Water (ISOW) from 1200 to 2300 m depth. This corresponded to the most striking feature of the whole section with DFe concentrations reaching up to 2.5 nmol L<sup>-1</sup> within the ISOW, Denmark Strait Overflow Water (DSOW) and Labrador Sea Water (LSW), three water masses that are part of the Deep Western Boundary Current and was likely the result of a lateral advection of particles in the Irminger. However, as these water masses reached the Labrador Sea, lower DFe levels were measured. These differences could be explained by different processes occurring within the benthic nepheloid layers, where DFe was sometimes trapped onto particles due to Mn-sediment within the Labrador Sea (Gourain et al., 2018) and sometimes released from the sediment potentially as a result of interactions with dissolved organic matter. Such Fe-binding organic ligands could have also been produced locally due to the intense remineralisation rate reported by Lemaître et al. (2017) of biogenic particles (Boyd et al., 2010; Gourain et al., 2018). The LSW exhibited increasing DFe concentrations along its flow path, likely resulting from sediment inputs at the Newfoundland Margin. Although DFe inputs through hydrothermal activity were expected at the slow spreading Reykjanes Ridge (Baker and German, 2004b; German et al., 1994), our data did not evidence this specific source as previously pointed by Achterberg et al. (2018) further north (~60°N) from our section.

In surface waters several sources of DFe were highlighted especially close to lands, with riverine inputs from the Tagus River at the Iberian margin (Menzel Barraqueta et al., 2018) and meteoric inputs (including coastal runoff and glacial meltwater) at the Newfoundland and Greenland margins (Benetti et al., 2016). Substantial sediment inputs were observed at all margins but with different intensity. The highest DFe sediment input was located at the Newfoundland margin, while the lowest was observed at the eastern Greenland margin. These differences could be explained by the different nature of particles with the most lithogenic located at the Iberian margin and the most biogenic, at the Newfoundland margin (Gourain et al., 2018). Although previous studies (e.g. Jickells et al., 2005; Shelley et al., 2015) reported that atmospheric inputs substantially fertilized surface waters from the West European Basin, in our study only stations located in the West European and Iceland Basins exhibited enhanced SML DFe inventories with lower TTADs. However, these TTADs were about three times higher than those reported for Saharan dust inputs and thus atmospheric deposition appeared to be a minor source of Fe at the sampling period. Finally, there was evidence of convective inputs of the LSW to surface seawater caused by long tip jet event (Piron et al., 2016) that deepened the winter mixed layer down to ~ 1200 m depth (Zunino et al., 2017), in which Fe was in excess of nitrate and where thus Fe was not limiting at the sampling period.

## Figures:

Figure 1: great;

Figure 2: increase letter size, it is hard to read;

Figure 3: I am not sure that white contour lines for DFe help to understand Chl a. I would remove DFe and include this figure in the supplementary material.

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Figure 4, 5 and 6(?): great

Figure 7, 8, 9, 10: can go in the supplementary material, maybe Fig. 9 you can keep

Figure 11 -14: in the sup mat.,

Maybe Figure 13 for the first 200 m can stay!

Table 3 and 4: belongs into the sup. Material

→ As you suggested we only kept Figs 1, 2, 4, 5, 9, 10 (the new one) and 13 (with your suggestions), all other Figures are now in the supplementary material. Tables 3 is also in the supplementary material.

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