Anonymous Referee#2 reviews

Dear Referee#2 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.,

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

Page 1, Line 35: "in the Denmark Straight. . ."

➔ We have changed the text accordingly.

Page 1, Line 35: explain what types of particles you are talking about and briefly explain the differences observed (which ones scavenge and which ones release dFe)

➔ We have added precision

Page 1 Lines 35-37: Finally, the nepheloid layers located in the different basins and at the Iberian Margin were found to act as either a source or a sink of DFe depending on the nature of particles with organic particles likely releasing DFe and Fe-Mn oxides scavenging DFe.

Page2, Line 4: The reasoning is not flowing properly here. You need to say that (1) high productivity leads to high atmospheric carbon capture and that (2) Deep water formation leads to sequestration of this carbon into deeper waters, where carbon is stored for longer. The last sentence comes a little out of the blue, needs to be better linked – instead close the paragraph highlighting why it is important to study trace metals in this area.

➔ We have reorganised the full introduction as suggested.

Page 2 Lines 1-33, Page 3 Lines 1-25:

1 Introduction

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_2 (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 photosynthesise. 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

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Moved up [1]: Page 1, Line 35: explain what types of particles you are talking about and briefly explain the differences observed (which ones scavenge and which ones release dFe) ¶

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frequently limits the activity and abundance of these organisms together with light conditions (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).

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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 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).

Page 2, Line10: I can not follow the reasoning in this paragraph. A little bit of a muddle of all the phytoplankton limiting factors (light, nutrients, wind, temp) without a clear insight what factor limits where. Needs to be better explained

➔ We have reorganised the full introduction as suggested (see above).

Page 2, Line 15: the connection between light limitation and nutrient limitation is not clearly explained

➔ We have reorganised the full introduction as suggested (see above).

Page 2, Line26: what about soluble Fe? Is this not considered the most bioavailable?

➔ Yes, we absolutely agree. Therefore to avoid confusion we have decided to remove the following sentence "…and within its physical speciation, its dissolved form (DFe) is considered to be the most available form for phytoplankton (Morel, 2008; Morel et al., 2008).".

Page 3, Line5: Aims of this paper are a bit poor. Add better understanding of the biogeochemical cycling of dFe in the oceans - inform biogeochemical models – and why this is important, what you expect to achieve. . ..

➔ We have reorganised the full introduction as suggested (see above).

Page 3, Line 20: remove "national"

➔ Removed

Page 3, line 30: do you mean concentrated HCl?

➔ We have changed the text for clarification

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

Page 3, Line25: why different filtering methods? Have you compared the Fe concentrations in those fractions? i.e., have you collected the same sample with both filtration cut-offs and checked there is no significant difference?

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➔ We added precision for clarification. Note that we did not collected the same samples with the two different techniques.

Page 4 Lines 10-14: Samples were either taken from the filtrate of particulate samples (collected on polyethersulfone filters, 0.45 µm supor®, see Gourain et al., this issue) or after filtration using 0.2 µm filter cartridges (Sartorius SARTOBRAN® 300) due to water budget restriction (Table 1). No significant difference was observed between DFe values filtered through 0.2 µm and 0.45 µm 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 4, Line 4: remove “daily basis”

➔ We have changed the text “High-purity grade solutions and water (Milli-Q) were used on a daily basis to prepare the following reagents:” by “High-purity grade solutions and water (Milli-Q) were used to prepare the following reagents each day:” (Page 4 Line 31)

Page 4, Line 14: replace “run” with “analytical session”

➔ We have changed the text as suggested (Page 5 Line 10)

Page 4, line 19: replace “in nmol L-1” with “to nmol L-1”

➔ We have changed the text as suggested (Page 5 Line 21)

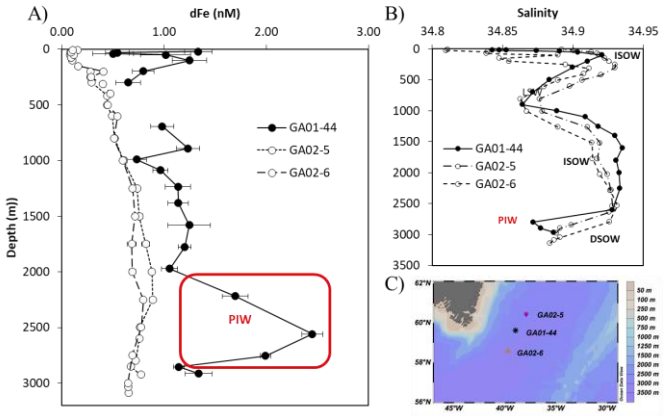
Page 4, Line 19: would it not be more correct to multiply by the specific density of each sample? If you do not have this data (normally this is a standard parameter obtained from temperature and salinity.

➔ Yes, we agree with the reviewer. However, the converting factor used for consensus materials has always been 1.025 kg L^{-1}, which is why we used this converting factor.

Page 4, Line 19: Please show a comparison of dFe data at the crossover-station with GA02, as an intercalibration exercise.

➔ We did not include an intercalibration exercise with the GA02 voyage as DFe was determined on board by FIA-CL during GA02, which thus means that some refractory DFe was not measured on board with only a short time of acidification (Chever et al., 2010).

➔ Just for your information, here after you will find the plot comparing both voyages:



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Page 4, Line25: Why did you not use the CTD data from the trace metal casts? Please explain

➔ We did not use the CTD data from the trace metal casts as the O2 data could not be calibrated. We therefore decided to use all the parameters from the same rosette, i.e. the stainless steel rosette.

Page 5, Line 12: awkward sentence, difficult to follow, please rewrite!

➔ We have changed the text for clarification

Page 7 Lines 4-5: Using this water mass determination, DFe concentrations were considered as representative of a specific water mass only when the contribution of this specific water mass was higher than 60% of the total water mass pool.

Page 6, Line 5: which central waters? Names?

➔ The central waters are Eastern North Atlantic Central Waters (ENACW). The acronym was added in the text there page 7 line 26.

Page 6, Line 9: to keep consistency, keep "stations 49 and 60" out of the parenthesis. Rephrase the end of the sentence to do so.

➔ We have modified the text as suggested

Page 7 Lines 27-30, Page 8 Lines 1: West of the Subarctic Front, Iceland SubPolar Mode Waters (IcSPMW, $7.07 < \theta < 8°C$, $35.16 < S < 35.23$, $280 < O_2 < 289$ µmol kg^{-1}) was encountered from stations 34-40 (accounting for more than 45% of the water mass pool from 0 to ~ 800 m depth) and Irminger SubPolar Mode Waters (IrSPMW, $\theta \approx 5°C$, $S \approx 35.014$) from stations 42-44 (contributing to 40% of the water mass pool from 0 to ~ 250 m depth) and stations 49 and 60 (accounting for 40% of the water mass pool down to 1300 m depth).

Page 6, Line10: specify which stations

➔ We have changed the text as suggested.

Page 8 Lines 1-2: The IcSPMW was also observed within the Subtropical gyre (stations 11-26), subducted below ENACW until ~ 1000 m depth.

Page 6, Line 14: remove "The" from start of sentence

➔ We changed the text accordingly.

Page 6, Line 19: what is that contribution? 40 %? Please specify!

➔ We have changed the text as suggested.

Page 8 Lines 9-10: The LSW was also observed in surface waters of station 44 with a similar contribution than IrSPMW (~ 40%).

Page 6, Line 28: ... “was” sourced from...

➔ We have corrected the text accordingly.

Page 6, Line 30: and “in the” Labrador Sea

➔ We have corrected the text accordingly.

Page 6, Line 34: I am getting a little lost with all those branches, not sure when you’re talking about the same one and when you change talking about another one. Not clear, please rephrase this section.

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➔ We have changed the text for clarification.

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 5: delete "The" from beginning of sentence. In this entire section please remove “the” in front of water masses. The text should be revised by one of the English speaking co-authors before submission.

➔ We have changed the text accordingly

Page 7, Line7: “lower” oxygen. . .

➔ We have changed the text accordingly.

Page 7, Line 4: what do you mean by dense shelf? Do you mean the water masses have higher density? Please rephrase

➔ We have changed the text for clarification

Page 8 Lines 28-32: Polar Intermediate Water (PIW, θ ≈ 0°C, S ≈ 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 16: “the” Charlie Gibbs. . .

➔ We have changed the text as suggested.

Page 7, Line 19: mixing with... remove "of the overflow"

➔ These lines have been removed according to Referee #1 comments

Page 7, Line 21: Which stations?

➔ The stations numbers were added for the Iceland Basin.

Page 9 Lines 4-6: ISOW was observed from 1500 m depth to the bottom of the entire Iceland Basin (stations 29-38) and from 1800 to 3000 m depth within the Irminger Sea (stations 40-60)

Page 7, Line 30: At least put a nitrate section figure in the supplementary file; otherwise text hard to follow. Data not yet available on the site you referenced

➔ Data are now available on the Sarthou et al., 2018 paper. 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 4: how do you define the "most open ocean station" for the transect? Deepest? Furthest away from land masses? Are you sure this is st 23?

➔ We have change the text for clarification and corrected the station number.

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Page 9 Lines 16-17: The low surface NO_3^- concentrations (lower than 6 µmol L^{-1}) in the West European Basin extended from station 2 (closest station to continental land mass) to station 25 (open ocean station) with concentrations ranging from 0.02 (station 11) to 3.9 (station 25) µmol L^{-1}.

Page 8, Line 4: it is unclear when you switch to talk about non-surface nitrate concentrations. Please rephrase this section to make this clearer.

➔ We have changed the section for clarification as suggested

Page 9 Lines 12-22: Surface nitrate (NO_3^-) concentrations (García-Ibáñez et al., 2018; Pérez et al., 2018; Sarthou et al., 2018) ranged from 0.01 to 10.1 µmol L^{-1} (stations 53 and 63, respectively). There was considerable spatial variability in NO_3^- surface distributions with high concentrations found in the Iceland Basin and Irminger Sea (higher than 6 µmol L^{-1}), as well as at stations 63 (10.1 µmol L^{-1}) and 64 (5.1 µmol L^{-1}), and low concentrations observed in the West European Basin, in the Labrador Sea and above continental margins. The low surface concentrations in the West European Basin ranged from 0.02 (station 11) to 3.9 (station 25) µmol L^{-1}. Station 26 delineating the extreme western boundary of the West European Basin exhibited enhanced NO_3^- concentrations as a result of mixing between ENACW and IcSPMW, although these surface waters were dominated by ENACW. In the Labrador Sea (stations 68-78) low surface concentrations were observed with values ranging from 0.04 (station 68) to 1.8 (station 71) µmol L^{-1}. At depth, the lowest concentrations (lower than 15.9 µmol L^{-1}) were measured in ENACW (~ 0 - 800 m depth) and DSOW (> 1400 m depth), while the highest concentrations were measured within NEADW (up to 23.5 µmol L^{-1}), and in the mesopelagic zone of the West European and Iceland Basins (higher than 18.4 µmol L^{-1}).

Page 8, Line 6: which depths are you talking about?

➔ We added precision as suggested (see above)

Page 8, Line 12: isn't the fluorometer calibrated with the Chl-a measurements?

➔ We removed this part as suggested by Referee#1. However, the fluorometer was calibrated separately from the Chl-*a* measurements. To do so, 6 stations (including 6 different water masses) at 6 depths (surface, chlorophyll-max down to the base of the euphotic zone) were sampled. These samples included early morning, late evening and daytime samples to account for non-photochemical quenching, which causes a decrease of fluorescence signal at the surface during day-time. Therefore, all profiles were calibrated using night-time dependency and corrected day-time surface data for non-photochemical quenching.

Page 8, Line 13: Specify which depth range you are considering for looking at min/max Chl-a concentrations. Evidently, minimum Chl-a concentrations are found in the deep ocean

➔ As you said, the minimum Chl-*a* concentrations are found in the deep ocean and therefore we only reported min and max values for the maximum Chl-*a* concentrations considering all stations. We have changed the text for clarification

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 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 mg m^{-3}, 30 m depth, station 53 and up to 6.6 mg m^{-3}, 23 m depth, station 61) and Newfoundland (up to 9.6 mg m^{-3}, 30 m depth, station 78) margins.

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Page 8, Line 19: remind the reader here here that all the dFe data can be found in the supplementary file.

➔ We have added this precision as suggested. Note that Referee#1 suggested to gather subsections 3.3.1 and 3.3.2, to remove the first lines from section 3.3 (Page 8 Lines 20-25) and to change section 3.3.3 for section 3.4, which is what we did.

Page 10 Lines 2-3: Dissolved Fe concentrations (see supplementary material) ranged from 0.09 ± 0.01 nmol L^{-1} (station 19, 20 m depth) to 7.8 ± 0.5 nmol L^{-1} (station 78, 371 m depth) (see Fig. 3).

Page 9, Line 13: when describing the regions, go in same order as in Figure, otherwise confusing (start Labrador Sea and end WEB)

➔ We have made the corrections accordingly.

Page 10 Lines 19-32 and Page 11 Lines 1-11: In the Labrador Sea, IrSPMW exhibited an average DFe concentration of 0.61 ± 0.21 nmol L^{-1} (n=14). DFe concentrations in the LSW were the lowest in this basin, with an average value of 0.71 ± 0.27 nmol L^{-1} (n=53) (Fig. 7). Deeper, ISOW displayed slightly higher average DFe concentrations (0.82 ± 0.05 nmol L^{-1}, n=2). Finally, DSOW had the lowest average (0.68 ± 0.06 nmol L^{-1}, n=3, Fig. 7) and median (0.65 nmol L^{-1}) DFe values for intermediate and deep waters.

In the Irminger Sea, surface waters were composed of SAIW (0.56 ± 0.24 nmol L^{-1}, n=4) and IrSPMW (0.72 ± 0.32 nmol L^{-1}, n=34). The highest open-ocean DFe concentrations (up to 2.5 ± 0.3 nmol L^{-1}, station 44, 2600 m depth) were measured within this basin. In the upper intermediate water LSW was identified only at stations 40 to 44, and had the highest DFe values with an average of 1.2 ± 0.3 nmol L^{-1} (n=14). ISOW showed higher DFe concentrations than in the Iceland Basin (1.3 ± 0.2 nmol L^{-1}, n=4). At the bottom, DSOW was mainly located at stations 42 and 44 and presented the highest average DFe values (1.4 ± 0.4 nmol L^{-1}, n=5) as well as the highest variability from all the water masses presented in this section (Fig. 7).

In the Iceland Basin, SAIW and IcSPMW displayed similar averaged DFe concentrations (0.67 ± 0.30 nmol L^{-1}, n=7 and 0.55 ± 0.34 nmol L^{-1}, n=22, respectively). Averaged DFe concentrations were similar in both LSW and ISOW, and higher than in SAIW and IcSPMW (0.96 ± 0.22 nmol L^{-1}, n=21 and 1.0 ± 0.3 nmol L^{-1}, n=10, respectively, Fig. 7).

Finally, in the West European Basin, DFe concentrations in ENACW were the lowest of the whole section with an average value of 0.30 ± 0.16 nmol L^{-1} (n=64). MOW was present deeper in the water column but was not characterized by particularly high or low DFe concentrations relative to the surrounding Atlantic waters (Fig. 7). The median DFe value in MOW was very similar to the median value when considering all water masses (0.77 nmol L^{-1}, Figs. 4 and 7). LSW and IcSPMW displayed slightly elevated DFe concentrations compared to the overall median with mean values of 0.82 ± 0.08 (n=28) and 0.80 ± 0.04 (n=8) nmol L^{-1}, respectively. The DFe concentrations in NEADW were relatively similar to the DFe median value of the GEOVIDE voyage (median DFe = 0.75 nmol L^{-1}, Figs. 4 and 7) with an average value of 0.74 ± 0.16 nmol L^{-1} (n=18) and presented relatively low median DFe concentrations (median DFe = 0.71 nmol L^{-1}) compared to other deep water masses.

Page 9, Line 13: remove “the” before MOW and in front of LSW (line 15), SAIW (line 24) and IrSPMW (line 24).

➔ We have removed “the” in front of each water masses throughout the section

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Page 9, Line 20: this is confusing, specify that you mean similar averages and not ranges (the range is larger for IcSPMW)

➔ We have added this precision as suggested (see above)

Page 9, Line 22: LSW and ISOW averages are also similar, combine sentences.

➔ We have changed the text accordingly (see above).

Page 9: Line 24: "composed of" instead of "characterised by"

➔ We have changed the text as suggested (see above).

Page 9, Line 32: Delete "compared to other ones"

➔ We have changed as suggested (see above)

Page 10, Line 2: "lowest average dFe value" (DSOW also shows the highest deep water dFe concentrations)

➔ We have changed the text as suggested (see above).

Page 10, Line 9: it is a little odd to start explaining what cannot be included in any of your sub-sections. You start introducing the general structure of your discussion and then you go into much detail explaining dFe trends all the sudden. This is totally out of place here. You should add this paragraph to the end of the discussion or in a new section.

➔ We have added a new section for this paragraph but we kept this new section there in the MS, as we think it would be odd to let the reader know at the end of the MS what we did not include.

Page 11 Lines 13-31:

4 Discussion

In the following sections, we will first discuss the high DFe concentrations observed throughout the water column of stations 1 and 17 located in the West European Basin (Section 4.1), then, the relationship between water masses and the DFe concentrations (Section 4.2) in intermediate (Section 4.2.2 and 4.2.3) and deep (Section 4.2.4 and 4.2.5) waters. We will also discuss the role of wind (Section 4.2.1), rivers (Section 4.3.1), meteoric water and sea-ice processes (Section 4.3.2), atmospheric deposition (Section 4.3.3) and sediments (Section 4.4) in delivering DFe. Finally, we will discuss the potential Fe limitation using DFe:NO_3^- ratios (Section 4.5).

4.1 High DFe concentrations at station 1 and 17

Considering the entire section, two stations (stations 1 and 17) showed irregularly high DFe concentrations (> 1 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). Conversely, no relationship was observed between DFe and PFe nor transmissometry for station 17. However, Ferron et al. (2016) reported a

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strong dissipation rate at the Azores-Biscay Rise (station 17) due to internal waves. The associated vertical energy fluxes could explain the homogenized profile of DFe at station 17, although such waves are not clearly evidenced in the velocity profiles. Consequently, the elevated DFe concentrations observed at station 17 remain unsolved.

Page 10, Line 20: I don't understand the aim of discussing dFe with water masses? I'd rather focus on the sources and sinks of dFe along the section. Discuss then the role of water masses in distributing the dFe signals. This means completely changing the focus of this section

➔ We decided to discuss DFe with the main water masses, i.e. LSW, ISOW and DSOW as they all constitute the Deep Western Boundary Current (DWBC).

Page 11, Line 2: Flow of thoughts not clear, reasons of deep winter mixing scattered; bits of information thrown in a little randomly. You should start off by saying that deep winter mixing is an important mechanism supplying nutrients to the surface ocean in the North Atlantic Ocean; then say how this deep winter mixing is produced (from what I can understand in your text are you trying to say this is due to the effects of wind + convective mixing+ subduction/upwelling; am I right? This was not clear); then say what the specific conditions were in the year you sampled. I am still giving you corrections on the section, which you can incorporate in your rewritten discussion if fitting. I think you can recycle some parts of your discussion.

➔ We have modified the text as suggested for clarification.

Page 12 Lines 12-22: In the North Atlantic Ocean, the warm and salty water masses of the upper limb of the MOC are progressively cooled and become denser, and subduct into the abyssal ocean. In some areas of the SubPolar North Atlantic, deep convective winter mixing provides a rare connection between surface and deep waters of the MOC thus constituting an important mechanism in supplying nutrients to the surface ocean (de Jong et al., 2012; Louanchi and Najjar, 2001). Deep convective winter mixing is triggered by the effect of wind and a pre-conditioning of the ocean in such a way that the inherent stability of the ocean is minimal. Pickart et al. (2003) demonstrated that these conditions are satisfied in the Irminger Sea with the presence of weakly stratified surface water, a close cyclonic circulation, which leads to the shoaling of the thermocline and intense winter air-sea buoyancy fluxes (Marshall and Schott, 1999). 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 7: "events" and "with a positive NAO"

➔ We have corrected as suggested.

Page 11, Line 8: "The winter mixed layer depth"

➔ We have corrected as suggested.

Page 11, Line 9: instead of " and were" use "which was"

➔ We have corrected as suggested.

Page 11, Line 11: "close to those found in LSW"

➔ We have corrected as suggested.

Page 11, Line 13: sentence incomprehensive, please rephrase

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➔ This sentence has been removed according to Referee#1 comments and gather with previous sentence.

Page 12 Lines 26-27: 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 24: need to improve a little the flow of thoughts in this paragraph. Start by saying why you see no MOW dFe signal, then support/contradict that argument(s) by what has been seen in other studies.

➔ We have modified this section according to your comments and Referee#1 comments.

Page 12 Lines 29-32 and Page 13 Lines 1-14: On its northern shores, the Mediterranean Sea is bordered by industrialized European countries, which act as a continuous source of anthropogenic derived constituents into the atmosphere, and on the southern shores by the arid and desert regions of north African and Arabian Desert belts, which act as sources of crustal material in the form of dust pulses (Chester et al., 1993; Guerzoni et al., 1999; Martin et al., 1989). During the summer, when thermal stratification occurs, DFe concentrations in the SML can increase over the whole Mediterranean Sea by 1.6-5.3 nmol L^{-1} in response to the accumulation of atmospheric Fe from both anthropogenic and natural origins (Bonnet and Guieu, 2004; Guieu et al., 2010; Sarthou and Jeandel, 2001). 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 (DAl, Menzel Barraqueta et al., 2018) concentrations (up to 38.7 nmol L^{-1}), confirming the high atmospheric deposition in the Mediterranean region. In contrast to Al, no DFe signature was associated with MOW (Figs. 2 and 4). 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 3: "suggesting that the water mass is enriched in dFe during its flow path"

➔ We have changed the text as suggested taking into account Referee#1 comments.

Page 13 Lines 16-18: 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 (see supplementary material Fig. S3).

Page 12, Line 4: start by saying what those sources are and then support with the available literature.

➔ We have reorganised this section based on your comments and Reviewer#1 comments.

Page 13 Lines 16-33, 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

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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.

The Irminger and Labrador Seas exhibited the highest averaged integrated TChl-a concentrations (98 ± 32 mg m^{-2} and 59 ± 42 mg m^{-2}) compared to the West European and Iceland Basins (39 ± 10 mg m^{-2} and 53 ± 16 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 chlorophillid-*a* 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:PAl ratios (0.44 ± 0.12 mol:mol and 0.38 ± 0.10 mol:mol, respectively) compared to particles from the West European and Iceland Basins (0.22 ± 0.10 and 0.38 ± 0.14 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, Lemaître et al. (2017) reported higher remineralisation rates within the Labrador (up to 13 mmol C m^{-2} d^{-1}) and Irminger Seas (up to 10 mmol C m^{-2} d^{-1}) using the excess barium proxy (Dehairs et al., 1997), compared to the West European and Iceland Basins (ranging from 4 to 6 mmol C m^{-2} 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 transmissometry 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 pmol.kg^{-1}) within the LSW at the edge of the Newfoundland Margin (45.73°W, 51.82°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 6: change “the ones” for “those”

➔ We have corrected as suggested (see above).

Page 12, Line 12: Provide a brief description of how the remineralisation rates were measured...

➔ We have added this precision (see above).

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Page 12, Line 18: You are repeating yourself!

➔ We removed this sentence.

Page 12, Line 19: conspitious? clearly visible? change this word

➔ We have changed the text for clarification (see above).

Page 12, Line 21: confusing between remineralisation and bacteria mediated ligand production. Please be clear.

➔ We have reformulated this sentence (see above).

Page 12, Line 27: "Hereafter"

➔ We have corrected the text.

Page 13, Line 2: you also need to consider vertical/lateral inputs (think in 3D), so sedimentary Fe could be coming from further north, for example, within nepheloid layers. You can't discard sedimentary inputs just by looking at vertical gradients.

➔ We have reorganised the idea of this paragraph and wehave considered your suggestions (see below).

Page 13, Line 3: remove "the" before "Polar Intermediate Water" and "PIW" (line 6)

➔ We have corrected the text accordingly.

Page 13, Line 9: instead of thinking that one water mass carries a certain dFe concentration (think of the short Fe residence times), this water mass might have "picked up" some dFe from, e.g., the sediments, on its pathway or It might have picked up particles in suspension which dissolve over time, etc!

➔ We have reorganised the idea of this paragraph and we have considered your suggestions.

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}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}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}Th remains unsolved. The only parameter that would explain without any ambiguity such differences of behaviour between DFe and ^{230}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.

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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 11: instead of "seawater" use "water masses"

➔ We have reformulated this sentence (see above)

Page 13, Line 11: specify what you mean; 100 % contribution of which water mass at which depth

➔ We have changed the text as suggested (see above).

Page 13, Line 18: watch out, this leads to miss-understanding as the reader thinks you are saying that particulate Fe is sustained by organic ligands. I don't think that is what you want to say.

➔ We have changed the text as suggested (see above).

Page 13, Line 19: restructure this paragraph, say what you think the reasons are behind the high dFe concentrations, then support that by correlations, graphs etc and then compare to literature to confirm or dispute your theory. end the section with a stronger statement of what you think is happening in these deep water masses

➔ We have changed the text as suggested (see above).

Page 13, Line 26: your explanation is a little long winded. Say that Mid Ocean Ridges can be a source of dFe but that this has not been found in the Reykjanes Ridge so far

➔ We have changed the text according to your comments and referee#1 comments.

Page 15 Lines 22-30: 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 sites that were either confirmed (Baker and German, 2004a; German et al., 1994; Olaffson 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).

Page 13, Line 30: you are repeating you pattern again; please first explain the signals you see and then compare to the literature

➔ We have changed the text according to your comments and Referee#1 comments.

Page 15 Lines 28-33, Page 16 Lines 1-3: However, enhanced DFe concentrations (up to 1.5 ± 0.22 nmol L^{-1}, station 36, 2200 m depth) were measured east of the Reykjanes Ridge (Fig. 3). 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 (see supplementary material Fig. S3). Indeed, Achterberg et al. (2018) highlighted at ~60°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

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could be due to a hydrothermal source and/or the resuspension of particles and their subsequent dissolution.

Page 14, Line 4-6: are these located on your transect? If not, you need to give their locations (coordinates) and explain where they are relative to your section. Are these GEOVIDE hydrographic sections (I don't think so) or from other cruises? Specify which ones. Cite appropriate Figures, and supporting literature.

➔ We have changed the text as suggested. Note that we also added the location of CGFZ and BFZ on Figure 1.

Page 16 Lines 4-12: 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°N and 34.61°W) and potentially Bight Fracture Zone (BFZ, 56.91°N and 32.74°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 x 10^6 m^3 s^{-1} with ISOW carrying a significant load of suspended sediment (25 µg 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.

Page 14, Line 11: complete this section with more recent references. There have been previous studies in this area

➔ We have removed the following sentence "The seabed of this area has been identified as a depositional environment with patches of ripples and rock fragments surrounded by moat." Based on Referee#1 comments and have changed the text (see above).

Page 14, Line 20: restructure, again start by saying what you see, explaining the reason of these signals showing correlations, and then supporting literature

➔ We have changed the text as suggested.

Page 16 Lines 19-32, Page 17 Lines 1-2: Enhanced DFe surface concentrations (up to 1.07 ± 0.12 nmol L^{-1}) were measured over the Iberian Margin (stations 1-4) and coincided with salinity minima (~ <35) and enhanced DAl concentrations (up to 31.8 nmol L^{-1}, Menzel Barraqueta et al., 2018). DFe and DAl concentrations were both significantly negatively correlated with salinity (R^2 = ~1 and 0.94, respectively) from stations 1 to 13 (Fig. 5). Salinity profiles from station 1 to 4 showed evidence of a freshwater source with surface salinity ranging from 34.95 (station 1) to 35.03 (station 4). Within this area, only two freshwater sources were possible: 1) wet atmospheric deposition (4 rain events, Shelley, pers. comm.) and 2) the Tagus River, since the ship SADCP data revealed a northward circulation (P. Lherminier and P. Zunino, Ifremer Brest, pers. comm.). Our SML DFe inventories were about three times higher at station 1 (~ 1 nmol L^{-1}) than those calculated during the GA03 voyage (~ 0.3 nmol L^{-1}, 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. Consequently, the Tagus River appears as 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. The Tagus estuary is the largest in the western European coast and very industrialized (Canário et al., 2003; de Barros, 1986; Figueres et al., 1985; Gaudencio et al., 1991; Mil-Homens et al., 2009), extends through an area of 320 km^2 and is

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characterized by a large water flow of 15.5 10^9 m^3 y^{-1} (Fiuza, 1984). 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 14, Line 26: You can't say "et al." in a personal communication. All people should be mentioned. Also State first name and affiliation in a personal communication.

➔ We have modified the text to fulfil this comment (P. Lherminier and P. Zunino, Ifremer Brest, pers. comm.)

Page 14, Line 26: Very difficult to follow; why "however"? Start by explaining the typical ratios of different sources before you give the observed ratios in your study. Then discuss the What dFe:dAl ratio is expected from a river source? And from an atmospheric source?

➔ We have removed this sentence according to your comment and Referee#1 comment and we have changed the text for clarification (see above).

Page 14, Line 29: Difficult to understand; you need to present each option that could lead to the enhanced dFe signal, discuss and then accept or discard

➔ We have changed the text as suggested (see above).

Page 15, Line 18: I don't understand this "extended as close as 200 km from our Greenland stations". Please rephrase. Also avoid parenthesis next to parenthesis, and explain clearly what can be found in the link and what info you got from there.

➔ We reword the sentence for clarification.

Page 17 Lines 10-17: The most plausible sources would be freshwater induced by meteoric water and sea-ice melt. Conversely, deeper in the water column, brine signals were calculated at stations 53 (100 m depth, Fig. 5D) 61 (100 m depth, Fig. 5E) and 78 (30 m depth, Fig. 5F). The release of brines could originate from two different processes: the sea-ice formation or the early melting of multiyear sea ice due to gravitational drainage and subsequent brine release (Petrich and Eicken, 2010; Wadhams, 2000). Indeed, during the winter preceding the GEOVIDE voyage, multiyear sea ice extended 200 km far from our Greenland stations (http://nsidc.org/arcticseaicenews/).

Page 15, Line 20: how were these calculated? give equations

➔ 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 $\delta^{18}O$ (δO^{18}_m). The mass balance calculations are presented below:

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

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

$$f_{AW}.\delta O^{18}_{AW} + f_{PW}.\delta O^{18}_{PW} + f_{MW}.\delta O^{18}_{MW} + f_{SIM}.\delta O^{18}_{SIM} = \delta O^{18}_m \text{ (eq.3)}$$

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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, δO^{18}_{AW} = +0.18‰ (Benetti et al., 2016); S_{PW} = 32.5, δO^{18}_{PW} = -1‰ (Cooper et al., 1997; Woodgate and Aagaard, 2005); S_{MW} = 0, δO^{18}_{MW} = -18.4‰ (Cooper et al., 2008); S_{SIM} = 4, δO^{18}_{SIM} = +0.5‰ (Melling and Moore, 1995).

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).

Page 15, Line 27: I suggest instead of describing the Figures, you should use them to support your discussion. So try to avoid starting sentences and paragraphs describing Figures! You do this very often

➔ We have changed the text as suggested.

Page 17 Lines 20-33, Page 18 Lines 1-2: Considering the sampling period at stations 53 (16 June 2014) and 61 (19 June 2014), sea-ice formation unlikely explained the brine signals calculated as this period coincides with summer melting in both the Central Arctic and East Greenland (Markus et al., 2009). However, it is possible that the brines observed in our study could originates from sea-ice formation, which occurred during the previous winter(s) at 66°N (and/or higher latitudes). The residence time can vary from days (von Appen et al., 2014) to 6-9 months (Sutherland et al., 2009). Due to our observed strong brine signal at station 61 we suggest that the residence time was potentially longer than average. Given that the brine signal was higher at station 61 than at station 53 (which was located upstream in the EGC), we suggest that station 53 was exhibiting a freshening as a result of the transition between the freezing period toward the melting period. This would result in a dilution of the brine signal at the upstream station. Consequently, the salinity of this brine signal may reflect sea ice formation versus melting which may have an effect on the trace metal concentration within this water (Hunke et al., 2011). The associated brine water at station 61 (100 m depth) was slightly depleted in both DFe and PFe, which may be attributed to sea ice formation processes. Indeed, Janssens et al. (2016) highlighted that as soon as sea ice forms, sea salts are efficiently flushed out of the ice while PFe is trapped within the crystal matrix and DFe accumulates, leading to an enrichment factor of these two Fe fractions compared to underlying seawater. Conversely, the brine signal observed at station 53 (100 m depth) showed slight enrichment in DFe, which may be attributed to brine release during early sea ice melting and the associated release of DFe into the underlying water column as the brine sinks until reaching neutral buoyancy due to higher density.

Page 15, Line 27: how is sea-ice fraction calculated? please provide equations (if you calculated it yourself) or references where this data is published. Provide clear info so the reader can understand

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

Page 15, Line 30: explain why you compare to dAl in these profiles.

➔ We have removed DAl distribution.

Page 15, Line 33: "originates"

➔ We have corrected accordingly.

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Page 16, Line 7: I can't believe that sea-ice can "uptake" Fe! The concentration of Fe in the newly formed ice and in the remaining water should stay the same. You need to find another explanation!

➔ We reword the sentence and add a reference for clarification (see above).

Page 16, Line 10: you mean sea-ice formation? Hence release of brine? Also explain that the brine sinks because it is denser (this is why it is observed below the surface)

➔ We have changed the text as suggested (see above).

➔ 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 16, Line 10: "release of dFe"

➔ We have corrected accordingly.

Page 16, Line 10: "underlying water column"

➔ We have corrected accordingly.

Page 16, Line 11: split this sentence, it is too long

➔ We have changed the text as suggested.

Page 16 Lines 29-32: Surface waters (from 0 to ~ 50 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 7.4% , respectively, from surface to ~50 m depth). Within these surface waters, station 53 exhibited substantial sea-ice melting contribution (1.5%, 4 m depth) while station 61 exhibited low contribution (0.6%) from brine release that was linearly increasing with depth (1.3% at 50 m depth and 2.2 % at 100 m depth).

Page 16, Line 15: instead of describing the data (this is more appropriate for results section) you should say what correlates with what (e.g., low Fe with low MW). difficult to follow flow of thoughts

➔ We have changed the text as suggested.

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-*a* concentrations (up to 6.6 mg m^{-3}) measured from surface to about 40 m depth, drastically decreasing at ~ 50 m depth to 3.9 mg m^{-3} (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

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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 17: remind the reader in which figure information can be found (Chl-a, ect)

➔ We added these precisions throughout this section.

Page 16, Line 24: results in agreement with the capacity of. . .? weird sentence, please change

➔ We have changed the text as suggested.

Page 18 Lines 7-11: Although the ability of MW from Greenland Ice Sheet and runoffs to deliver DFe and PFe to surrounding waters have 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).

Page 16, Line 27: so far you have not talked about dFe:dAl ratio in meteoric water. Explain if this ratio is used to trace MW inputs and what you see in your profiles. should explain at the start.

➔ We have added a sentence at the beginning of the section to fulfil this comment and removed the DAl data.

Page 17 Lines 15-17: In the following sections, we discuss the potential for meteoric water supply, sea-ice formation and sea-ice melting to affect DFe distribution.

Page 16, Line 28: change "noting that" for "although"

➔ We changed the text as suggested.

Page 17, Line 6: or maybe the brine conditions (pH, salinity etc) make the Fe more bioavailable; or maybe this peak is not Fe related, but related to the release of other TM, or a phytoplankton group that thrives in brine and does not require much Fe? Since this shelf is further south, the environmental conditions may be more favourable for phytoplankton to grow and hence consuming all the dFe more rapidly. You should explore all the possibilities

➔ We have changed the text as suggested.

Page 18 Lines 28-30: This either suggests that the brine likely contained important amounts of Fe (dissolved and/or particulate Fe) that were readily available for phytoplankton and consumed at the sampling period by potentially sea-ice algae themselves (Riebesell et al., 1991) or that another nutrient was triggering the phytoplankton bloom.

Page 17, Line 19: You repeat the word "dust" too much

➔ We have changed the text according to your comment and Referee#1 comment.

Page 18 Lines 1-2 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

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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 20, "proportions"

➔ We have changed the text (see above).

Page 18, Line 3: you are going into too much detail here about aerosols, which is part of the Shelley papers

➔ We have removed this part.

Page 18, Line 6: Meskhidze et al. (2017) is not in your reference list

➔ We added the reference to the reference list.

Page 18, Line 10: information is a little randomly thrown in... what is your point? This is not a review paper

➔ We have removed this part.

Page 18, Line 13: What does OM mean? write abbreviations out first time. Do you mean organic matter?

➔ Yes, we meant organic matter but we have removed this part.

Page 18, Line 15: So all this background information to lastly say that you don't comment on this? Rephrase, make some assumptions or delete some detail

➔ We have removed this part.

Page 18, Line 26: of those stations, station 40 is most similar to total aerosol dFe:dAl ratios. station 26 is closer to the soluble than the total composition, so I don't understand what you are saying

➔ We removed all this part and changed it for turnover times relative to atmospheric deposition as defined in Guieu et al., 2014.

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 ~ 9 ± 3 months compared to other basins (7 ± 2 years and 5 ± 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 (~ 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.

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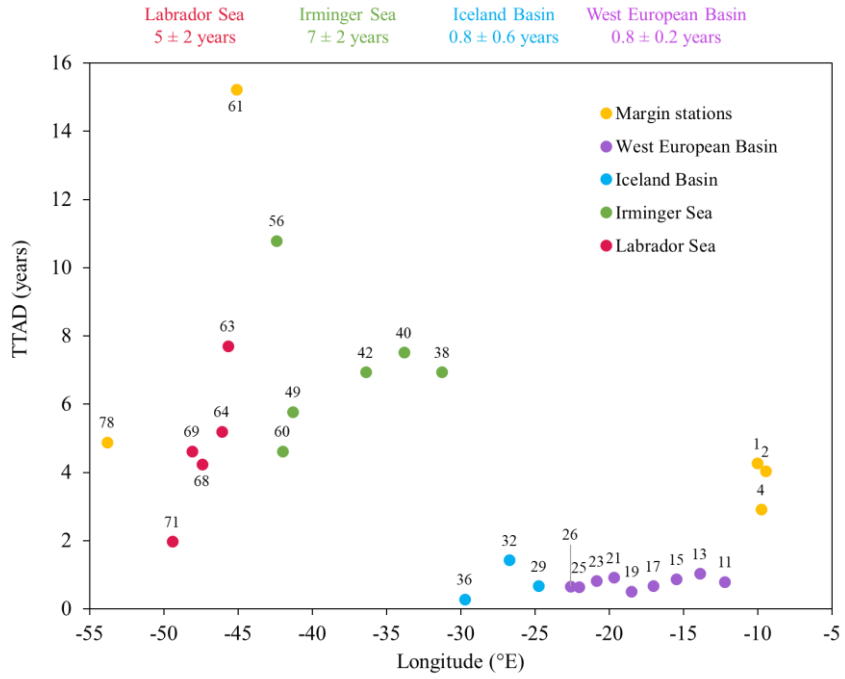

Figure 6: Plot of dissolved Fe (DFe) Turnover Times relative to Atmospheric Deposition (TTADs) calculated from soluble Fe contained in aerosols estimated from a two-stage sequential leach (UHP water, then 25% HAc, Shelley et al., this issue). Note that numbers on top of data points represent station 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.

Page 18, Line 28: And what about all the other stations where the ratios are different? I would say this is more of a "coincidence" that these data fall onto the black line. The multiple reactions occurring as Fe enters the ocean change this Fe:Al ratio rapidly

➔ Same as above, this part has been removed.

Page 18, Line 30: remove “time”

➔ This part has been removed (see above)

Page 19, Line 2: I don’t understand.... these station points on your figure 10 are similar to those of other stations. What makes you think they have a higher atmospheric influence??

➔ This part has also been removed (see above)

Page 19, Line 4: in this section you should mention that bottom water dFe concentrations were significantly higher on the Newfoundland margin than on the Greenland margins

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➔ We have included this precision.

Page 19 Lines 20-24: DFe concentration profiles from all coastal stations (stations 2, 4, 53, 56, 61 and 78) are reported on Figure 5. To avoid surface processes, only depths below 100 m depth will be considered in the following discussion. Stations where DFe and PFe followed a similar pattern are stations 2, 53, 56, and 78, suggesting that either the sources of Fe supplied both Fe fractions (dissolved and particulate) or that PFe dissolution from sediments supplied DFe. Among the different margins, the Newfoundland Margin exhibited the highest deep-water DFe concentrations.

Page 19, Line 10: "mol:mol" to stay consistent

➔ We have changed the text as suggested.

Page 19, Line 10: what is the average useful for? Show a plot with dFe:pFe ratios or a table

➔ Because the SD is relatively high thus conferring distinct signature throughout the water column (below 100 m depth) of each station. This allows the comparison station by station instead of comparing each samples. These ratios were added to Table 3.

Page 19, Line 11: as well as different sediment compositions, this could be also due to different supply mechanisms? Different sediment conditions (redox, organic content, temp, etc)

➔ We added this precision in the text.

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 ± 0.07 mol:mol (n = 23, Table 3), 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 17: Where is the predominance of diatoms?

➔ We changed the sentence for clarification.

Page 19 Lines 31-33: These observations are consistent with the high TChl-*a* 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 21: I think this section is great, but you need to organise the ideas clearly. Now difficult to follow. Also name this section "nepheloid layers"

➔ We changed the section title as suggested and reorganised the flow of ideas.

Page 19, Line 26: explain the criterion first. Explain briefly the PCA and the results you show in figure 11. Information is thrown in a little randomly. Please organise you paragraphs

➔ We reorganised the paragraph as suggested.

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 DAl (Menzel Barraqueta et al., 2018) and

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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.

Page 20, Line 1: I would not call it AOU "concentrations" find another way to express this

➔ We changed "concentrations" for parameters (Page 20 Line 20)

Page 20, Line 13: you should look into non-reducing dissolution of lithogenic material. You are missing out on a big topic! Radic et al., 2011; Labatut et al., 2014; Abadie et al., 2017

➔ We included this topic in the text as suggested.

Page 20 Lines 26-34, Page 21 Lines 1-6: 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 complexion 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.

Page 20, Line 14: "lead"

➔ We changed the text accordingly.

Page 20, Line 16: instead of "these" use "this sediment-derived. . ."

➔ We have changed the text accordingly.

Page 20, Line 17: do not state this as facts... these are assumptions

➔ We have changed the text as suggested(see above).

Page 20, Line 25: very poor sum-up, please improve

➔ We have changed the full section according to your comment (see above).

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Page 20, Line 26: You should also compare surface dFe data to AOU to look at dFe released from remineralisation. You have done this for >500 m depth, but it will be worth looking at this more closely below the surface mixed layer, where remineralisation occurs (below 100 m depth).

➔ We have included this just before Fe* section.

Page 21 Lines 25-33, Page 22 Lines 1-34, Page 23 Lines 1-3: The remineralisation of organic matter is a 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 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) (AOU = 3.88 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 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 (AOU = 22.6 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 NO_3^- (R^2=0.42, n=51, p-value < 0.05). The slope of the relationship, representing the typical remineralisation ratio, was $R_{Fe:N}$ = 0.07 ± 0.01 mmol mol^{-1}. The intercept of the regression line was -0.4 ± 0.2 nmol L^{-1}, reflecting possible excess of preformed NO_3^- compare to DFe in these water masses. These significant correlations allow us to use the 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 PO_4^{3-}, and modified here for NO_3^- as follow:

$$Fe^* = [DFe] - R_{Fe:N} \times [NO_3^-] \quad \text{(eq. 4)}$$

where $R_{Fe:N}$ refers to the average biological uptake ratio Fe over nitrogen, and [NO_3^-] refers to nitrate concentrations in seawater. Although, we imposed a fixed biological $R_{Fe:N}$ of 0.05 mmol mol^{-1}, it is important to note that the biological uptake ratio of DFe:NO_3^- is not likely to be constant. Indeed, this ratio has been found to range from 0.05 to 0.9 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 NO_3^- calculated for this study (i.e. $R_{Fe:N}$ = 0.07 ± 0.01 mmol mol^{-1}, for Subpolar waters). Negative values of 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 NO_3^- ($R_{Fe:N}$ = 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 Fe: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 ($R_{Fe:N}$ = 0.05) within the Iceland Basin, Irminger and Labrador Seas. The Fe deficiency observed in surface waters (> 50 m depth) from the Irminger and Labrador Seas might be explained by low atmospheric deposition for the IcSPMW and the LSW (Shelley et al., 2017). Low atmospheric Fe supply and sub-optimal Fe: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

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DFe: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 DFe: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_3^- 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_3^- (from 7 to 8 µmol L^{-1}) concentrations. Similarly, the SAIW exhibited high NO_3^- 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.

Page 20, Line 30: change the ending of this sentence, not properly expressed

➔ We have split and changed the sentence (see above).

Page 20, Line 31: First explain why you talk about Fe:nitrate ratios. THis comes out of the blue! Also cite Fig 12.

➔ We have changed the text as suggested (see above).

Page 21, Line 3: what do you mean by influence of the river, and the currents? specify what you mean

➔ We have changed the text for clarification (see above).

Page 21, Line 6: Can you provide a different kind of plot to help visualise this gradient you are talking about. In figure 12 this is impossible

➔ We have included a surface map of DFe:NO3- ratios.

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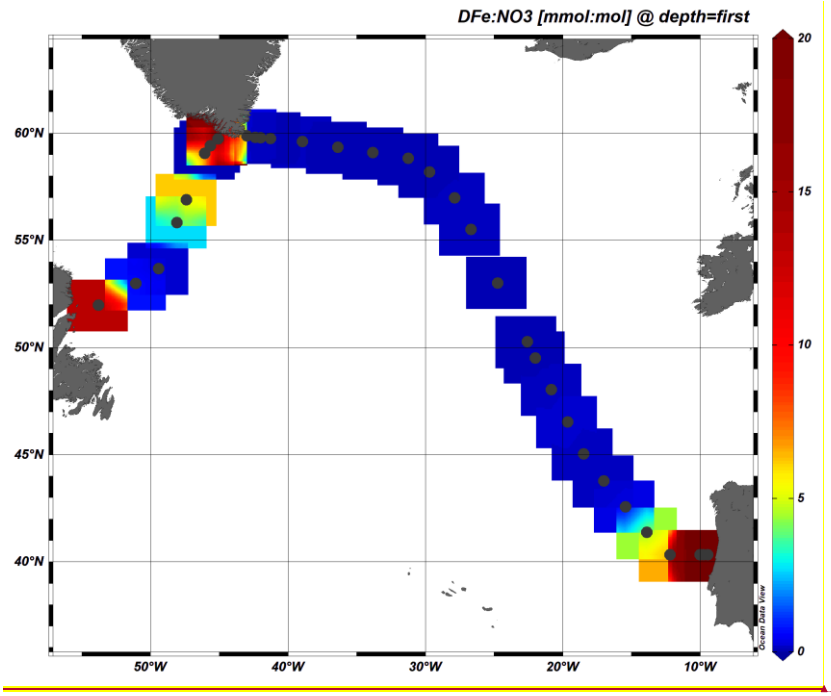

Page 21, Line 18: “disequilibrium” sounds a bit odd, better use the word “ranges”

➔ We have changed the text as suggested (see above).

Page 21, Line 23: do you assume that all the nitrate in seawater comes from remineralisation? Better explain what assumptions this equation relies on

➔ We have modified the text accordingly (see above the response to your comment “Page 20, Line 26”)

Page 21, Line 26: Rather, negative values of 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

➔ We have changed the text as suggested.

Page 21, Line 27: remove "out"

➔ We have changed the text as suggested.

Page 21, Line 28: you talk about surface waters here but the calculations are done below 100 m depth. I would keep discussion on the external sources of dFe and then link to inputs of dFe rich water masses to surface waters above

➔ We have reorganised the section according to your comments and Reviewer#1 comments. Therefore we only focus the discussion on the top 200 m depth of the section.

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

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term determines to which extent DFe will be utilised. The DFe:NO_3^- 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). Values were typically equal or lower than 0.28 mmol mol^{-1} 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^{-2}. 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^{-2}. The high DFe:NO_3^- 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_3^- concentrations, were relatively enriched in DFe compared to waters from Iceland Basin and Irminger Sea.

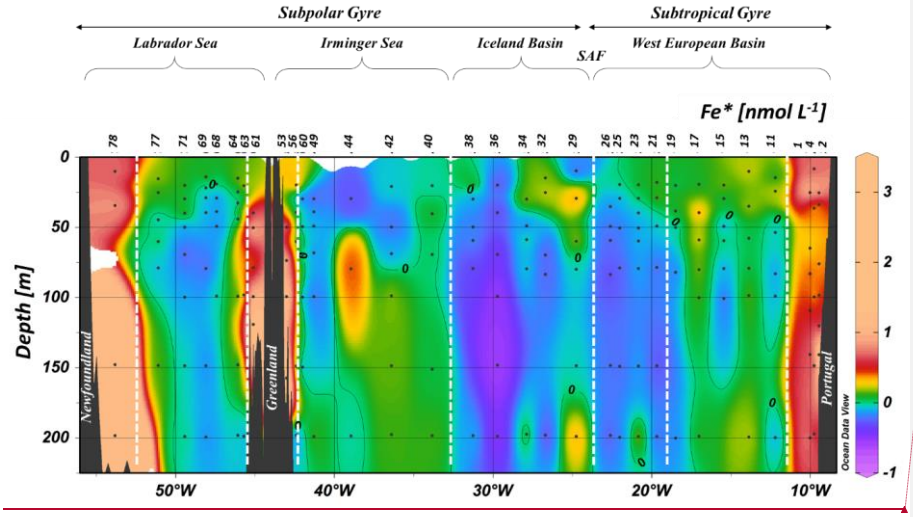
In our study, DFe:NO_3^- ratios displayed a gradient from the West European Basin to Greenland (supplementary material S7 and S8). This trend only reverses when the influence of Greenland was encountered, as also observed by Painter et al. (2014). The remineralisation of organic matter is a major source 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 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) (AOU = 3.88 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 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 (AOU = 22.6 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 NO_3^- (R^2=0.42, n=51, p-value < 0.05). The slope of the relationship, representing the typical remineralisation ratio, was $R_{Fe:N}$ = 0.07 ± 0.01 mmol mol^{-1}. The intercept of the regression line was -0.4 ± 0.2 nmol L^{-1}, reflecting possible excess of preformed NO_3^- compare to DFe in these water masses. These significant correlations allow us to use the 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 PO_4^{3-}, and modified here for NO_3^- as follow:

$$Fe^* = [DFe] - R_{Fe:N} \times [NO_3^-] \quad \text{(eq. 4)}$$

where $R_{Fe:N}$ refers to the average biological uptake ratio Fe over nitrogen, and [NO_3^-] refers to nitrate concentrations in seawater. Although, we imposed a fixed biological $R_{Fe:N}$ of 0.05 mmol mol^{-1}, it is important to note that the biological uptake ratio of DFe:NO_3^- is not likely to be constant. Indeed, this ratio has been found to range from 0.05 to 0.9 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 NO_3^- calculated for this study (i.e. $R_{Fe:N}$ = 0.07 ± 0.01 mmol mol^{-1}, for Subpolar waters). Negative values of 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 NO_3^- ($R_{Fe:N}$ = 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 Fe: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 ($R_{Fe:N}$ = 0.05) within the Iceland Basin, Irminger and Labrador Seas. The Fe deficiency observed in surface waters (> 50 m depth) from the Irminger and Labrador Seas might be explained by low atmospheric deposition for the IcSPMW and the LSW (Shelley et al., 2017). Low atmospheric Fe supply and sub-optimal Fe: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 DFe: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 DFe: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_3^- 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_3^- (from 7 to 8 µmol L^{-1}) concentrations. Similarly, the SAIW exhibited high NO_3^- 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.



Page 22, Line 5: what has the low Fe supply to do with the "inefficient" carbon pump? If you want to talk about the carbon pump, and its inefficiency, you need to support with adequate statements/findings. I do not think this is a finding of your study

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➔ We have removed this part (see above).

Page 22, Line 7: this comes a little out of the blue. Explain a little more the high remineralised carbon fluxes and how they were measured

➔ We replaced in context this sentence and the precision on the measurement of remineralised carbon fluxes is now precised earlier in the MS.

Page 22, Line 12: poor ending. What about other sources? Margins? Rivers?

➔ We have changed the text (see above).

Page 22, Line 15: first sentence superfluous

➔ We have removed this sentence as suggested.

Page 22, Line 23: depletion of nitrate? That doesn't make sense

➔ We have changed the sentence for clarification (see above).

Page 22, Line 27: "entrained it to the deep. . ."

➔ We have changed the text as suggested.

Page 22, Line 29: "in the deep ocean"

➔ We have changed the text as suggested.

Page 22, Line 30: do you mean particles? Sediments are on the seafloor. Same for line 32

➔ Yes you are right, we have changed "sediments" for "particles".

Page 23, Line 3: conclusions need to be rewritten after the discussion is reworked.

➔ 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^{-1} 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 be 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.

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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 that 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.

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