

Dear Dr Schlosser,

We would like to thank you for your very constructive comments. All the issues you raised were carefully considered and addressed.

Below are our detailed answers, including corresponding lines of text in the revised manuscript. We also took into account reviewer 2 comments, and refer to them accordingly in this response. We provide an updated manuscript with on including all modifications in track changes.

We hope that you will find our answers satisfactory and our revised manuscript suitable for publication in this special issue of Biogeosciences

Sincerely yours,

Arthur Gourain, on behalf of all the authors

“This manuscript presents and discusses the distribution of PFe, PAI, PMn and PP in the high latitudinal North Atlantic. The presented water column data is wonderful and I am looking forward to see the data published in the next GEOTRACES intermediate data product. We need more particulate data! And I really like their PMF calculations. However, the discussion is very detailed and long, but I am missing a straight storyline. The authors jump a lot between different topics and even present Ba data at the end of the manuscript, but a discussion is missing. The manuscript needs serious work, and I am suggesting major revision.” [We thank the reviewer for his constructive remarks and his acknowledgment of performed work. Regarding the Ba data, we are not presenting them anymore, as we thought the discussion was easier to follow without remineralisation section.](#)

“My three main points are:

The authors conclude that higher PFe/PAI and PMn/PAI ratios are indicative for biogenic bound particulate Fe. I am missing the discussion of scavenged and authigenic Fe, that could also cause PFe/PAI ratios higher than that of crustal ratios. For my opinion, the authors should include the PFe/PMn ratio, where biogenic ratios (phytoplankton) are available in the literature. [It is true that a discussion on authigenic Fe was clearly missing, and we would like to thank you for this very constructive comment. We, unfortunately, cannot investigate specifically this important fraction of particulate iron. We are considering the PFe/PMn is having more bias than PFe/PAI due to the high kinetic of oxidation of Mn within the ocean. Kinetic wick is different than the kinetic of Fe. We now discuss more carefully on the impact of authigenic particles having on PFe/PAI.](#)

The authors include a PMF model and conclude that variances in PFe are related to changes in the content of lithogenic particles. This is in contradiction to the authors conclusion of biogenic Fe, responsible for changes in PFe/PAI. This needs to be discussed more carefully! [The main lithogenic variability of the PFe is not in contradiction with the biogenic contribution to the PFe/PAI ratio. Indeed with the PMF, we describe the overall variability of PFe. The main variability of the PFe happened with inputs of PFe. These inputs are as described mainly lithogenic and imply the results observed by the PMF. Variation of PFe/PAI ratio between the basins is mainly occurring in open ocean samples where PFe concentrations are around 1nM. Thus, small variation of PFe at a sub-nanomolar level will highly impact the PFe/PAI elemental ratio.](#)

[We add some details at the line 293 to avoid any ambiguity of the PMF interpretation. “The PMF analysis has been realised on the entire dataset, in consequence, the factors described are highly influenced by the major variations of particulate element concentrations \(at the interface, i.e. margin, seafloor, surface,...\).”](#)

There is an entire data set of barium excess concentrations at the end of the manuscript. I am not sure that this data is required for the conclusion of the author. If kept, please discuss the data! [We removed this section following your advice.](#)

Abstract Line 32: What is meant with “At most stations over the Western” and “..relative concentration..”? I cannot see how concentrations show a ubiquitous influence of crustal particles. Ratios maybe! However, be more precise. [We modified the sentence as follow: Within the Iberian Abyssal Plain, ratio of PFe over particulate aluminium \(PAI\) is identical to the continental crust ratio \(0.21\), indicating the important influence of crustal particles in the water column.](#)

Introduction Line 78: Replace to “using the distribution of particulate aluminium, manganese, and phosphorous.” And remove sub-sentence “; to further...” [Done.](#)

Methods Line 90: Sentence too long, please split up. [Done.](#)

Line 91: Missing bracket. [Corrected.](#)

Line 97: Indicate Go-Flo company. “General Oceanics”. [Done.](#)

Line 100: 6mm sounds a bit thin for me. Kable must be wider. [Our mistake, it was 14mm.](#)

Line 111: Replace “litters” by “liters”. [Done.](#)

Line 111ff: Filter cleaning and what kind of filters were applied, should be stated earlier. For instance, before how much volume was passed over them. [This sentence has been moved at the beginning of the paragraph \(line 113\).](#)

Line 113: Remove “-1” from “MΩ cm-1” [Done.](#)

Line 118ff: I cannot follow, is this releant? [It is relevant, as remaining seasalts can seriously reduce the sensitivity of SF-ICP-MS analyses, so they must be reduced as much as possible.](#)

Line 120: Replace “slide” by “dish”. [We did not replace this, as the Millipore company sells these items as « Petrislides » and not « Petri dishes ». They don't have the same design as standard petri dishes.](#)

Line 143: Please provide the values of blanks and limit of detection, maybe in Table 1 (Please also provide the standard deviation of your crm analysis). [These have been added to Table 1.](#)

Results General comment 1: The result section need to be shortened. You mention in line 277 that PAI and PMn and PFe are similar in IrB, IcB , WEB and IAP (in line 270ff that the Reykjanes ridge is similar to IcB). That is the entire stretch between Spain and Greenland! Please combine results! In addition, if I look at Figure 4, the distribution of PFe in LB seems very similar to the concentrations in IcB. I am suggesting to combine the results of open ocean regions and just include separate paragraphs of results from the three margins, Iberian, Greenland and Newfoundland margin. [The results have been re-arranged following your advice, with a first section regrouping all open ocean stations, then a section on margins.](#)

General comment 2: You talk about different surface currents, please include them in Fig. 1. [During the preparation of the manuscript, we tried different possibility to produce a map of our section. Including some currents has been explored. But having the position of every station, their IDs and the different biogeochemical provinces, plus the current make the Figure 1 extremely unclear and difficult to understand. We reference at the beginning of the Hydrography section the paper of Garcia-Ibanez \(Line 182\). In this paper, the current are well explained and a really good map of the current, their figure 1, is produced.](#)

Line 184ff: Could just find ENACW in Figure 2. Please correct text or Figure 2. The text has been corrected; the water masse is called NACW and not ENACW.

Line 198: Remove “really” Done.

Line 210: IB refers to IrB and IcB? Please mention that. The IB refers to the Iceland Basin. This paragraph has been deleted in light of the Reviewer comments.

Line 218ff: There are five concentrations for 4 parameters! The additional concentration referring to another element finally non-discussed has been removed. It was “21.5”nM.

Line 221: Please include the standard deviation of trace metals and PP hosted by small particles Done.

Line 226: Please refer to transmissometry Figure. Done.

Line 228ff: Sentence is hard to follow, please rephrase. The sentence has been change to: “Within the first 50m, PFe concentrations decreased towards the shelf break where PFe dropped down from 2.53 nmol L⁻¹ (station 2) to 0.8 nmol L⁻¹ (Station 1). Line 248.

Line 233ff: Sentence “The highest. . .” does not tell anything new, remove! Since you explain results from the Iberian Margin, later referred as (IM), please include IM in Figure 2. The sentence has been removed. And the figure 2 has been updated, the two other shelves has been also added to keep some consistence.

Line 240ff: There is something wrong with that sentence! The sentence has been completely rewritten in light of your General Comment 1.

Line 242: When it is really the case at “every stations” then there are no exceptions! Please rephrase. Done.

Line 244: I do not understand what is meant here: “Particulate aluminium profiles matched the PFe profiles, with low median concentrations within the first 100m of 1.77 nmol L⁻¹ and 26 pmol L⁻¹ respectively. Then, concentrations increased with depth to reach a maximum close to the oceanic floor.” Did you mean 1.77nM Pal and 26pM PFe? Please provide values for bottom waters. The sentence has been completely rewritten in light of your General Comment 1.

Line 258: Replace “progressive” by “gradual”. Again refer to transmissometry figure. The sentence has been completely rewritten in light of your General Comment 1.

Line 315: You are mentioning lithogenic elements here. How do you now? I know concentrations are high, but before introducing your tool that differentiate between biogenic and lithogenic Fe, I would leave out such terms. The word “lithogenic” elements have been removed.

Discussion Line 323: I would also include run-off, which is probably similar to your “melting ice shelves” but more precise. In addition, why sea ice must have melted recently to be a source for PFe. And what do you mean with biological pool? However, please be careful what you state here as source, for instance, lateral mixing is not per se a source, just when PFe loaded waters are advected offshore. Please be more precise! The sentence has been modified to answer the comment as follows: “Possible candidate sources of PFe include lateral advection offshore from the different margins, atmospheric inputs, continental run-off, melting ice shelves and icebergs, resuspended sediments, hydrothermal inputs and biological uptake.”. Line 271.

Line 350ff: How barite formation refers to remineralistaion of PFe. Expalin! In addition, what inputs and processes are discussed below! A sentence has been added in the previous paragraph at the line 286. The sentence on in the inputs and processes has been removed for more clarity.

Line 365: Equation 2: I am pretty sure that the * should be -. Indeed this was a mistake. It has been corrected.

Line 367: Another possibility might be that lithogenic particles from the Iberian shelf are advected offshore. In addition, the NAC is located further west [D J Reynolds et al., 2016]. Authors need to come up with a better idea, than dust! The sentence has been re-written. The combination of the Iberian Margin input and local circulation is now discussed. Line 317.

Line 375ff: This paragraph needs an overhaul! From fronts, via isobaths and isotherms (not shown) to blooms and LSW. It is really hard to follow this paragraph. In general I would have wished the authors explained differences in PFe/PAI ratio and PFe lith% over the entire transect and not just WEB and IAP. We completely rewrote this paragraph with more explanation on the front and the change of %PFe_{litho} proxy over the section. Line 321.

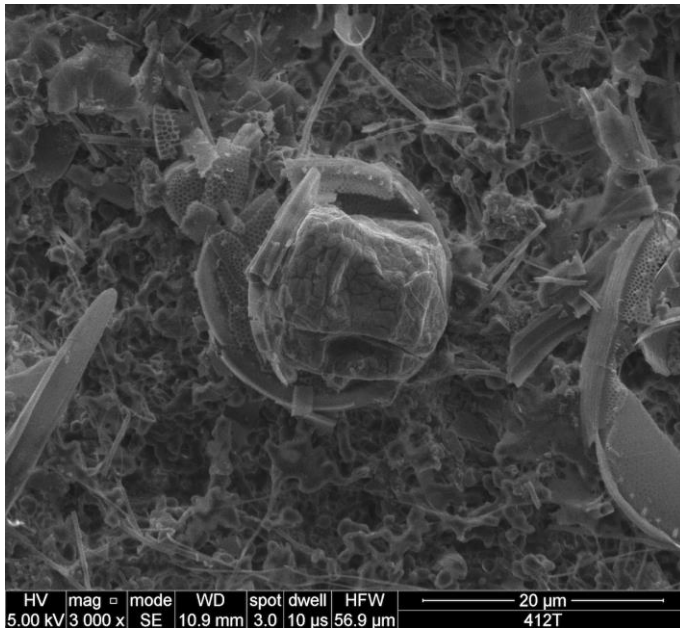
Line 384ff: Figure 8: The approach fingerprinting water masses with trace metals such as Fe and Mn would be nice, if it actually works. Other than NEADW and MW, other water masses have a higher Fe/Al ratio but they are very variable. In this case it is vital to check that the water mass difference is significant. I am suggesting performing a student t-test! The fingerprints analysis does not work using the Fe and Mn concentrations. We added the statistical analysis done on this. We operate a Kruskal Wallis Anova on-ranks test. This analysis demonstrates the significant difference between the different clusters of stations. We added more detail in the text, lines 330.

Line 400-409: You just repeat yourself, please remove! We deleted most of this paragraph except the following sentences: “Inputs from continental shelves and margins have been demonstrated to support high productivity in shallow coastal areas. Inputs of iron from continental margin sediments supporting the high productivity found in shallow coastal regions have been demonstrated in the past (e.g. Cullen et al. (2009), Elrod et al. (2004), Jeandel et al. (2011), Ussher et al. (2007)) and sometimes, were shown to be advected at great distances from the coast (e.g. Lam et al., 2008).”. The importance of the margin inputs for biology has not been discussed previously in the paper and we consider it important to mention it.

Line 414ff: I am not convinced that the different Fe/Al ratio is driven by different sediments. Where are the elemental ratios of the sediments, just because a sediment is muddy does not prove anything. Further Shelley et al. (2017) showed that dust particles along the GA01 section are mainly from the higher latitudes and not from North Africa. Later on you mention biogenic contribution, you have not introduced this term, and now everything higher than the 0.21 is biogenic. This is questionable, what is with scavenging, authigenic FeOOH formation etc. Throughout this paragraph, we are comparing sediment resuspension at the three different margins. The different mineralogical compositions of these sediments highly influence the composition of the resuspended particles measured. The sediment composition is thus, in our opinion, the key factor influencing the elemental ratio of particles sampled under these conditions (sedimentary resuspension). Unfortunately, we do not have sediment cores at these stations to assess by SEM or chemically their exact composition. Concerning the influence of Saharan dust over the Iberian Margin, Shelley et al. (2015) confirm their influence in the WEB. The influence of northern dust is highlighted in the northern part of the section. You are right that we now refer everything higher than 0.21 as “non-lithogenic” and not “biogenic”.

Line 425: I am not convinced that using a Mn/Al ratio from the upper crust, is helpful tracking sediments. Sediments can have a much higher ratio than the upper crust (eg. Sediments on the shelf of South Georgia 0.0066, Schlosser et al 2018). A higher Mn/Al ratio would change your figure 10 entirely. I am suggesting to apply the Mn/Al ratio of sediments from the different regions. In addition, the transmissometry data in figure 10 need to be cited earlier! We completely agree with your comment, the ideal solution will be to have the exact composition of the sediments at our respective stations. Unfortunately as explained in the previous comment. We don't have access to this information. The different study at our margin (Iberian (Blasco et al., 2000; Merinero et al., 2008) or Greenland (Loring et al., 1996) or in the North Atlantic (Menendez et al., 2017) show important variations of ratio on small spatial range. In consequence, we decided to use a uniform reference value, the UCC value, to compare our different samples. We added a sentence to discuss about the caveats of this proxy. “This proxy assumes homogeneity of the sediment PMn/PAI ratio through the section which is maybe not completely the case at every station. In consequence, this proxy is only a tool to identify new benthic resuspension at specific location and inter-comparison between several locations is not possible.” Line 370.

Line 490: I am again not convinced that just biological uptake was responsible for elevated Fe/Al ratios. Scavenging and authigenic precipitation would do the same job. You need SEM data to convince me! [This SEM picture was taken at Station 53, 165m depth:](#)



[On this image, you can see diatom debris and lithogenic particles and no Fe oxides could be detected on these samples.](#)

Line 547ff: What do you mean with “oxido-reductive transformation”. I know this term from microbiology classes, but in sediments? I am agreeing, dead biology sinks and settles on the seafloor. However, organic material is quickly remineralised and released Fe will oxidize quickly forming oxyhydroxides. FeOOH precipitate as single particle or form a coating around sediment particles. This will increase your Fe/Al ratio too, and I think even more pronounced than biogenic Fe, which in comparison to lithogenic particles stores just a small amount of Fe. This small quantity will be strongly obscured by lithogenic Fe. Everything below line 548 is highly speculative. If you would like to track biogenic Fe, you should use the Fe/Mn ratio, ratios are provided by T-Y Ho et al. [2003]. His Fe/Mn ratio for phytoplankton is ~ 1.7, lithogenic particles have a significantly higher ratio (upper crust ~50 and sediments ~ 70), indicative for the formation of authigenic Fe. [By “oxido-reductive transformation”, we mean any reaction of oxidation and reduction of trace element. These reactions are highly important within the sediment link to the oxygen and sulphur concentration. We agree on the plausible transformation of the biogenic PFe after burial in the sediment. But in the case of a quick oxidation of the dissolved iron from reductive sediments, the speciation of PFe is changing but not the molar ratio between PFe/PAI. Concerning the use of PFe/PMn as a proxy for the biological activity, we think using Mn and not Al brings another uncertainties. Indeed, Mn is highly affected by authigenic formation with different kinetic than iron. We agree that PFe/PAI ratio is not an idealistic proxy but we think it’s the best to estimate the contribution of sources over iron cycle. Moreover, results from the PMF demonstrate the weak influence of authigenic particles over the particulate iron cycle.](#)

Line 620ff: Any explanation for Fe depleted particles and aerosols, respectively? Anthropogenic? [Buck et al. \(2010\)](#) are not giving any specific indication about reasons for these particles to be Fe-depleted.

Line 636: I would check the Fe/Mn ratio too! [This paragraph has been deleted. In light of the reviewers’ comments, we decided it was too speculative using the current dataset at our disposition.](#)

Line 650: Now we are back to PFe/PP and not anymore PFeonlith/PP. This is all very confusing. It is an interesting approach using PP, but what are numbers actually tell us. It would be better to show first how much nonlithogenic Fe is in the top 100 m and plot PP as well in a diagram. By looking at picture 3. PP is similarly

high in IrB and LB, changes in PFe/PP are then mainly driven by PFe, but what does it actually mean. Further on, you show nice plots using Ba excess data (Fig. 13 & 14), but there is not a single word towards the end of the discussion. There is more work needed!! ~~This paragraph has been deleted. In light of the reviewers' comments, we decided it was too speculative using the current dataset at our disposition.~~

Dear reviewer,

We would like to thank you for your very constructive comments. All the issues you raised were carefully considered and addressed.

Below are our detailed answers, including corresponding lines of text in the revised manuscript. Note that we also took in account Dr Schlosser's comments when we rewrote the manuscript. We also attach the manuscript in track changes as a supplementary material.

We hope that you will find our answers satisfactory and our revised manuscript suitable for publication in this special issue of Biogeosciences.

Sincerely yours,

Arthur Gourain, on behalf of all the authors

This manuscript presents the vertical distribution of particulate Fe, Al, Mn and P in the North Atlantic along the Geovide section. Particulate trace elements data are still very scarce, and this dataset constitutes a major contribution to our understanding of the biogeochemical cycles of these elements. I am aware that an important work has been done to acquire such a dataset (more than 500 samples!). [We thank the reviewer for this comment.](#)

However, this manuscript is too detailed and the reader can be easily lost. It is difficult to retain clear conclusions from each section. Overall, I think that the discussion section is too ambitious, and the sections about the sources (e.g. dust inputs) and processes (e.g. remineralization) affecting the PFe distribution are sometimes too speculative. [We rewrote the discussion in light of this comment and are more cautious with our conclusions. We removed the remineralisation section which was too speculative.](#)

The discussion could be improved by adding additional information/parameters collected during the cruise (Chl-a, DFe, . . .), and a link between the particulate and dissolved concentrations is missing. [The link between particulate and dissolved is made and discussed thoroughly in Tonnard et al. \(under review for Biogeosciences\), together with Chl-a data ; this is why it is not specifically included in the manuscript. More references to Tonnard et al. are included through the discussion.](#)

The main part of this study used the PFe/PAI ratio to quantify the lithogenic PFe fraction and deduce the non-lithogenic fraction. However, it is likely that this crustal signature is not constant over the Geovide transect. The relevance and limitations of using an unique ratio need to be discussed. [The use of a single PFe/PAI crustal ratio is now discussed line 309.](#)

This work deserves to be published in Biogeosciences, but only after major revisions (see my comments below). Specific comments Overall, the introduction and methods are well written.

Figures and tables are not enough used in the text to discuss the results. [More references to figures and tables are included in the manuscript.](#)

The results section should be shortened – describing the particulate concentrations station by station in is probably not the easiest way to present this dataset. I think the sections 3.2 to 3.10 should be merged and synthetized. [This issue was also raised by Dr Schlosser. The results have been re-arranged following your advice, with a first section regrouping all open ocean stations, then a section on margins.](#)

In addition, the authors try to describe and explain each feature of the transect. It is probably too ambitious and not so useful. Finally, the size fractionation represents an important information. This aspect is not enough discussed in the manuscript. [Regarding the size fractionation, we want to discuss it in a separate paper, which will be focused on the top 100m.](#)

L33 – near-ubiquitous . . . but only in the western part of the transect. The sentence is confusing. The sentence was indeed not clear enough, we changed it by: “Within the Iberian Abyssal Plain, ratio of PFe over particulate aluminium (PAI) is identical to the continental crust ratio (0.21), indicating the important influence of crustal particles in the water column”. Line 32.

L36 – I would prefer to see a flux here instead of a concentration. A flux will be indeed more interesting but we can’t measure a flux over our samples. We’re lacking of a spatial resolution to calculate it.

L61 – The term remineralization usually refers to PFe, not DFe. Indeed the formulation of the sentence wasn’t clear enough, we changed it by: “or produced by remineralisation of particles”. Line 61.

L209-216 –I would remove this section (ms too long), and add one or two sentences with references in the discussion if needed. If this section is conserved, type 6 and 8-haptophytes should be explained. This section has been removed.

Section 3.3 and 3.4 – A figure or table should be cited to help the reader. The Figure 3 is cited at the end of the overview section 3.2 as follow: “Data are shown in Figure 3”. This figure includes all the parameters discussed along the following paragraphs. We are now citing this figure throughout this section

L330-340 – I would transfer this paragraph in the Methods section. Done. It is now located in the section 2.5, line 166.

Section 4.1 – This is an interesting approach. I am not sure if it is possible, but it would be very interesting to do such an analysis for two depth horizons, in surface (eg 0-100 m) and below 100 m. It could enable to highlight the vertical distribution of different processes (eg formation of barite mostly in the mesopelagic?). Indeed this could be interesting to perform and we had a go at it while preparing this manuscript. The main issue we encountered with clustering our dataset by depth range is the loss of positive statistical results. The PMF model needs a lot of data to work properly and by using a small subset of samples, the model is unstable.

L365 – A term is missing in equation 2. We modified it.

L367-373 – I recommend here to indicate that a biogenic pool is likely present but is masked by the huge proportion of lithogenic PFe. Overall, PFe/PAI is a proxy and the interpretation should be done with care. An additional sentence has been added to explain how the proxies need to be used with care and a comment on biogenic influence has been added from line 309 to 315.

L375 – Which feature? The dominance of lithogenic PFe discussed line 369 and 370? The feature described is the dramatic change of regime from station 26. We rearrange this paragraph in light of Dr. Schlosser review. Line 321.

L375-383 – This paragraph is a bit confusing. In addition, why only atmospheric inputs are discussed here? We have reworded this paragraph, and added a discussion on the dispersal of Iberian margin rich particles. A similar comment was raised by Dr Schlosser. From line 316 to line 319.

L414-416 – This sentence, and the whole paragraph seems to say that the Fe/Al ratio from the UCC used to calculate the lithogenic component is not accurate. I am aware that there is no perfect method to discriminate biogenic and lithogenic Fe and PFe/PAI is only a proxy, however this paragraph clearly contradicts the calculation made before. As it is one of the main objective of the paper, this limitation/bias should be discussed. Regarding this paragraph concerning the benthic inputs of particles, we discussed the different composition of sediments along the section. It is important to not consider sediments as a purely lithogenic source. Benthic sediments are the results of sinking of particles from the above water column. And represent in a certain term, a record of the oceanic particles flux. They are a mix of the overall bulk of particles lithogenic, biogenic and autogenic. Differences of ratio in these sediments are not implying in any way a change of ratio in the crust (continental or oceanic).

L416-419 – I may be wrong, but I think that the PFe/PAI signature of the desert dust coming from the Sahara significantly differs from the UCC ratio. See Guieu et al. 2002, Fu et al. 2017, . . . The sentence referring to the aerosol inputs have been removed.

L489 – Replace leaded by led. Done.

L502-507 – Other data collected during the cruise could be used here to illustrate the intensity of the bloom. For example, what was the surface chlorophyll a concentration? I recommend to add this kind of information all along the text, it should help making the manuscript less speculative. We added the Chl-a concentrations corresponding to the bloom and refer to Tonnard et al. (2018) as the Chl-a data are discussed in this paper.

e. L533-535 – What does an important phytoplanktonic community mean? It needs to be more precise. Furthermore, a low PFe concentration is not in contradiction with high Chl-a concentrations as usually most of the PFe concentration is from lithogenic or detrital origin and the biogenic pool is usually minor, and driven by intense cycling in surface. The sentence has been modified in light of this comment “The important phytoplanktonic community present (maximum Chl-a= 4.91 mg m⁻³, Tonnard et al., in prep), is linked to low PFe of 0.79 nmol L⁻¹ at 10 m, but, with a high PFe/PAl ratio, up to 0.4, and PP concentration of 97 nmol L⁻¹, confirming the biologic influence”. Line 472.

L536 – A value / order of magnitude is needed here. Furthermore, it has to be compared with the other areas. The sentence has been modified to: “Concerning this latter process, intense remineralization at station 77 (7 mmol C m⁻² d⁻¹ compared to 4 mmol C m⁻² d⁻¹ in the Western European Basin) has been reported by Lemaitre et al. (2018a)”. Line 475.

L537-539 – This sentence is confusing. We removed this sentence for clarity purposes.

L557-564 – To reduce the length of the manuscript, I would remove this paragraph. We consider that it is important to briefly provide a definition of the benthic nepheloid layers so, to take the reviewer’s point on board, we reduced the length of the paragraph as follows: “Benthic nepheloid layers (BNLs) are important layers where local resuspension of sedimentary particles (Bishop and Biscaye, 1982; Eittrheim et al., 1976; Rutgers Van Der Loeff et al., 2002) occur due to strong hydrographic stresses (i.e. boundary currents, benthic storms and deep eddies) interacting with the ocean floor ((Biscaye and Eittrheim, 1977; Eittrheim et al., 1976; Gardner et al., 2017, 2018). Along the GA01 section, BNLs were observable in each province with different strengths (Figures 3 and 12)”. Line 494.

L552-554 – What did Lam et al. (2017) precisely show? Lam was describing the role of physical characteristic on margin resuspension event. The use wasn’t completely appropriate, we removed the sentence.

Section 4.3.2 – Here, I cannot see a clear conclusion. We added the following sentence: “Along the GEOVIDE section, BNLs are providing high concentrations of particulate trace element in the deep open ocean that can contribute substantially to the pool of particulate trace elements such as as iron.”, Line 516.

L586-601 – This paragraph is probably too long to conclude an absence of hydrothermal inputs. The first part of the paragraph has been removed to shorten the paragraph.

L604 – I can’t see these information on Fig. 7. This paragraph has been removed as explained in the answer of L604-605 comment.

L604-605 – PFe/PAl is higher at station 40 than at station 38. Indeed, in light of it, we decided to remove this paragraph due to the lack of significate proof to support this part of the discussion.

L605 – This a general comment for the whole text: “PMn had a 19% sedimentary origin”. The authors refer to a proxy, and should say “about 20%”. This paragraph has been deleted as explained previously. Moreover we’ve been more careful on the use of proxy over the entire manuscript.

L616-617 – See my previous comment (L416-419). We removed the sentence in question.

Section 4.3.4 – Here, there is no clear conclusion. I would recommend to remove this section. We want to keep this paragraph about atmospheric inputs. Even if the fact we do not observe any atmospheric deposition is not as interesting as huge deposition events. We think it is important to discuss it, even if the conclusion is not as clear as the other sources.

L643 – A range of Fe/P cell quotas has been reported for the North Atlantic (see Twining et al.). It would be interesting here to compare this ratio (assuming 100% of P is from biogenic origin) which gives an estimation of the biogenic PFe in surface with the 100% lithogenic PFe obtained at stations 1-26 using equation 1. This

comparison could help to discuss the limitations of such approach. This paragraph has been deleted. In light of the reviewers' comments, we decided it was too speculative using the current dataset at our disposition.

L638-641 – This sentence needs a reference. This paragraph has been deleted. In light of the reviewers' comments, we decided it was too speculative using the current dataset at our disposition.

L646 – Replace pelagic by mesopelagic. This paragraph has been deleted. In light of the reviewers' comments, we decided it was too speculative using the current dataset at our disposition.

L649 – How is defined the remineralization depth? It needs to be explained. This paragraph has been deleted. In light of the reviewers' comments, we decided it was too speculative using the current dataset at our disposition.

L648-650 – PFe/PAI is probably not the best parameter to discuss remineralization since both elements are mostly lithogenic and the variation of this ratio due to remineralization is likely negligible. This paragraph has been deleted. In light of the reviewers' comments, we decided it was too speculative using the current dataset at our disposition.

L650-651 – I am not convinced by this explanation. PP is much more labile than PFe, whatever the remineralization rate. In addition, Fe scavenging could also contribute to this increase in PFe/PP. This paragraph has been deleted. In light of the reviewers' comments, we decided it was too speculative using the current dataset at our disposition.

L652-653 – The authors should explain why scavenging starts to be important only below 600 m depth. This paragraph has been deleted. In light of the reviewers' comments, we decided it was too speculative using the current dataset at our disposition.

L654-659 – This paragraph is confusing. Figure 13 is not introduced and explained. In addition, how the authors conclude to a stronger scavenging of DFe? This paragraph has been deleted. In light of the reviewers' comments, we decided it was too speculative using the current dataset at our disposition.

L661-664 – It is surprising to see a lower remineralization rate for P compared to Fe. This finding should be discussed. In addition, PFe/PP is not presented in a figure and it is hard for the reader to follow the discussion. This paragraph has been deleted. In light of the reviewers' comments, we decided it was too speculative using the current dataset at our disposition.

Section 4.3.5 – Overall, this section is too speculative. The potential impact of the scavenging process is not really discussed, and I think that the use of the PFe/PAI ratio to discuss the different remineralization patterns is not relevant (eg the evolution of DFe would be more appropriate). Finally, it is not easy to draw any clear conclusions from this section. This paragraph has been deleted. In light of the reviewers' comments, we decided it was too speculative using the current dataset at our disposition.

Figures 13 and 14 – These figures are not introduced and discussed in the manuscript. I would remove them and cite the appropriate study instead. This paragraph has been deleted. In light of the reviewers' comments, we decided it was too speculative using the current dataset at our disposition.

1 **Inputs and processes affecting the distribution of**
2 **particulate iron in the North Atlantic along the GEOVIDE**
3 **(GEOTRACES GA01) section**

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5
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21
22 **Abstract**

23 The GEOVIDE cruise (May-June 2014, R/V Pourquoi Pas?) aimed to provide a better understanding on trace
24 metal biogeochemical cycles in the North Atlantic. As particles play a key role in the global biogeochemical
25 cycle of trace elements in the ocean, we discuss the distribution of particulate iron (PFe), in light of particulate
26 aluminium (PAI), manganese (PMn) and phosphorus (PP) distributions. Overall, 32 full vertical profiles were
27 collected for trace metal analyses, representing more than 500 samples. This resolution provides a solid basis for
28 assessing concentration distributions, elemental ratios, size-fractionation, or adsorptive scavenging processes in
29 key areas of the thermohaline circulation. Total particulate iron (PFe) concentrations ranged from as low as 9
30 pmol L⁻¹ in surface Labrador Sea waters to 304 nmol L⁻¹ near the Iberian margin, while median PFe
31 concentrations of 1.15 nmol L⁻¹ were measured over the sub-euphotic ocean interior.

32 Within the Iberian Abyssal Plain, ratio of PFe over particulate aluminium (PAI) is identical to the continental
33 crust ratio (0.21), indicating the important influence of crustal particles in the water column.~~At most stations~~
34 ~~over the Western, the relative concentrations of total PFe and aluminium (PAI) showed the near-ubiquitous~~
35 ~~influence of crustal particles in the water column.~~ Overall, the lithogenic component explained more than 87%
36 of PFe variance along the section. Within the Irminger and Labrador basins, the formation of biogenic particles
37 led to an increase of the PFe/PAI ratio (up to 0.7 mol mol⁻¹) compared to the continental crust ratio (0.21 mol

38 | mol⁻¹). Margins provide important quantities of particulate trace elements (up to 10 nmol L⁻¹ of PFe) to the open
39 ocean, and in the case of the Iberian margin, advection of PFe was visible more than 250km away from the
40 margin. Additionally, several benthic nepheloid layers spreading over 200m above the seafloor were
41 encountered along the transect, especially in the Icelandic, Irminger and Labrador basins, delivering particles
42 with high PFe content, up to 89 nmol L⁻¹ of PFe. Finally, remineralisation processes are also discussed, and
43 showed different patterns among basins and elements.

44

45 1. Introduction

46 Particles play a key role in the ocean where they drive the residence time of most elements (Jeandel et al., 2015),
47 and strongly influence the global biogeochemistry of macro and micro-nutrients including iron (Milne et al.,
48 2017). In the surface ocean, biological activity produces biogenic suspended matter through planktonic
49 organisms, while atmospheric deposition (Baker et al., 2013; Jickells et al., 2005), riverine discharge (Aguilar-
50 Islas et al., 2013; Berger et al., 2008; Ussher et al., 2004) or ice-melting (Hawkings et al., 2014; Lannuzel et al.,
51 2011, 2014) bring mostly lithogenic derived particles to surface waters. These particulate inputs highly vary,
52 both spatially and seasonally, around the world's oceans. At depth, benthic and shelf sediment resuspension
53 (e.g. Aguilar-Islas et al., 2013; Cullen et al., 2009; Elrod et al., 2004; Fitzwater et al., 2000; Hwang et al., 2010;
54 Lam et al., 2015; Lam and Bishop, 2008; McCave and Hall, 2002), and hydrothermal activity (Elderfield and
55 Schultz, 1996; Lam et al., 2012; Tagliabue et al., 2010, 2017; Trefry et al., 1985), provides important amounts
56 of particles to the water column. Moreover, authigenic particles can be produced *in-situ* by aggregation of
57 colloids (Bergquist et al., 2007) or oxidation processes (Bishop and Fleisher, 1987; Collier and Edmond, 1984).
58 Thus, oceanic particles result from a complex combination of these different sources and processes (Lam et al.,
59 2015).

60 Particles represent the main part of the total iron pool in the upper water column (Radic et al., 2011), and
61 strongly interact with the dissolved pool (e.g. Ellwood et al., 2014). Indeed, dissolved iron can be scavenged
62 onto particles (Gerringa et al., 2015; Rijkenberg et al., 2014), incorporated into biogenic particles (Berger et al.,
63 2008) or [produced by remineralisation of particles](#) (Dehairs et al., 2008; Sarthou et al., 2008). Interestingly,
64 the concept of “reversible scavenging” (i.e. release at depth of dissolved iron previously scavenged onto
65 particles) has been advocated recently (Dutay et al., 2015; Jeandel and Oelkers, 2015; Labatut et al., 2014),
66 while other studies reveal distinct dissolution processes (e.g. Oelkers et al., 2012; Cheize et al., submitted to
67 Chemical Geology). Slow dissolution of particulate iron at margins has also been evoked as a continuous
68 fertilizer of primary production and should be considered as a source of dissolved iron (e.g. Jeandel et al., 2011;
69 Jeandel and Oelkers, 2015; Lam and Bishop, 2008). Within or below the mixed layer, the rates of regeneration
70 processes can also impact the bioavailable pool of iron, among other trace metals (e.g. Ellwood et al., 2014;
71 Nuester et al., 2014). However, the rates of these processes are not yet fully constrained. The study of particulate
72 iron is thus essential to better constrain the global biogeochemical cycle of iron in the ocean. This subject
73 received a growing interest over the last 10 years in particular (e.g. Bishop and Biscaye, 1982; Collier and
74 Edmond, 1984; Frew et al., 2006; Lam et al., 2012; Milne et al., 2017; Planquette et al., 2011, 2013; Sherrell et
75 al., 1998) and, to our knowledge, only two have been performed at an ocean-wide scale and published so far: the
76 GA03 GEOTRACES North Atlantic Zonal Transect (Lam et al., 2015; Ohnemus and Lam, 2015) and the GP16
77 GEOTRACES Pacific Transect (Lam et al., 2017; Lee et al., 2017).

78 In this context, this paper presents the particulate iron distribution in the North Atlantic Ocean, along the
79 GEOTRACES GA01 section (GEOVIDE), and discusses the various sources and processes affecting its
80 distribution, using ~~the distribution of other trace elements, more particularly~~ particulate aluminium, phosphorus
81 or manganese, ~~to further our understanding of this important pool of iron.~~
82

83 2. Methods

84 2.1. Study area

85 Particulate samples were collected at 32 stations during the GEOVIDE (GEOTRACES GA01 section) ~~campaign~~
86 ~~cruise~~ between May and June 2014 aboard the R/V *Pourquoi Pas?* in the North Atlantic. The sampling spanned
87 several biogeochemical provinces (Figure 1): ~~that first comprised~~ the Iberian margin (IM, Stations 2, 1 and 4),
88 the Iberian Abyssal Plain (IAP, Stations 11 to 17), the Western European Basin (WEB, Station 19 to Station 29)
89 ~~and~~ the Icelandic Basin (IcB, Stations 32 to 36). ~~Then, samples were collected,~~ above the Reykjanes Ridge (RR,
90 Station 38), ~~in~~ the Irminger Basin (IrB, Stations 40 to 60), ~~close to~~ the Greenland shelf (GS, Stations 53 and 61),
91 the Labrador Basin (LB, Stations 63 to 77) and finally ~~close~~ the Newfoundland shelf (NS, Station 78) (Figure
92 1).

93 The North Atlantic is characterized by a complex circulation (briefly described in section 2.1 and in detail by
94 Zunino et al. (2017) and García-Ibáñez et al. (2015) and is one of the most productive regions of the global
95 ocean (Martin et al., 1993; Sanders et al., 2014), with a complex phytoplankton community structure composed
96 of diverse taxa (Tonnard et al., in prep.).
97

98 2.2. Sampling

99
100 Samples were collected using the French GEOTRACES clean rosette, equipped with twenty-two 12L GO-FLO
101 bottles (two bottles were leaking and were never deployed during the cruise). GO-FLO bottles (~~General~~
102 ~~Oceanics~~) were initially cleaned in the home laboratory (LEMAR) following the GEOTRACES procedures
103 (Cutter and Bruland, 2012). The rosette was deployed on a ~~146~~mm Kevlar cable with a dedicated, custom-
104 designed clean winch. Immediately after recovery, the GO-FLO bottles were individually covered at each end
105 with plastic bags to minimize contamination. They were then transferred into a clean container (class-100) for
106 sampling, and the filters processed under a laminar flow unit. On each cast, nutrient and/or salinity samples
107 were taken to check potential leakage of the GO-FLO bottles.

108 ~~Prior to filtration, Filters were cleaned following the GEOTRACES protocols~~
109 ~~(<http://www.geotraces.org/images/Cookbook.pdf>) and kept in acid-cleaned 1 L LDPE bottles (Nalgene) filled~~
110 ~~with ultrapure water (Milli-Q, resistivity of 18.2 MΩ cm) until use. All filters were 25 mm diameter in order to~~
111 ~~optimize signal over the filter blank except at the surface depth where 47 mm diameter filters mounted on acid-~~
112 ~~cleaned polysulfone filter holders (Nalgene™) were used. Prior to filtration,~~ the GO-FLO bottles were shaken
113 three times, as recommended in the GEOTRACES cookbook to avoid settling of particles in the lower part of
114 the bottle. GO-FLO bottles were pressurized to <8 psi with 0.2 µm filtered dinitrogen (N₂, Air Liquide).
115 Seawater was then filtered directly through paired filters (Pall Gelman Supor™ 0.45 µm polyetersulfone, and
116 Millipore mixed ester cellulose MF 5 µm) mounted in Swinnex polypropylene filter holders (Millipore),

117 following Planquette and Sherrell (2012) inside the clean container. Filtration was operated until the bottle was
118 empty or until the filter clogged; volume filtered ranged from 2 liters for surface samples to 11L within the
119 water column. ~~Filters were cleaned following the protocol described in Planquette and Sherrell (2012) and kept~~
120 ~~in acid-cleaned 1 L LDPE bottles (Nalgene) filled with ultrapure water (Milli-Q, resistivity of $18.2 \text{ M}\Omega \cdot \text{cm}^{-1}$)~~
121 ~~until use. All filters were 25 mm diameter in order to optimize signal over the filter blank except at the surface~~
122 ~~depth where 47 mm diameter filters mounted on acid-cleaned polysulfone filter holders (NalgeneTM) were used.~~
123 After filtration, filter holders were disconnected from the GO-FLO bottles and a gentle vacuum was applied
124 using a syringe in order to remove any residual water under a laminar flow hood. Filters were then removed
125 from the filter holders with plastic tweezers that were rinsed with Milli-Q between samples. Most of the
126 remaining seawater was ‘sipped’ by capillary action, when placing the non-sampled side of the filter onto a
127 clean 47 mm supor filter. Then, each filter pair was placed in an acid-cleaned polystyrene Petri~~Slides-slide~~
128 (Millipore), double bagged, and finally stored at -20°C until analysis at LEMAR. Between casts, filter holders
129 were thoroughly rinsed with Milli-Q, placed in an acid bath (5% HCl) for 24 hours, then rinsed with Milli-Q.
130 At each station, process blanks were collected as follows: 2L of a deep (1000 m) and a shallow (40 m) seawater
131 samples were first filtered through a $0.2 \mu\text{m}$ pore size capsule filter (Pall Gelman Acropak 200) mounted on the
132 outlet of the GO-FLO bottle before to pass through the particle sampling filter, which was attached directly to
133 the swinnex filter holder.

134

135 2.3. Analytical methods

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

141 Filters were digested following the method described in Planquette and Sherrell (2012). Filter were placed on
142 the inner wall of acid-clean 15mL PFA vials (SavillexTM), and 2 mL of a solution containing 2.9 mol L^{-1}
143 hydrofluoric acid (HF, suprapur grade, Merck) and 8 mol L^{-1} nitric acid (HNO_3 , Ultrapur grade, Merck) was
144 added to each vial. Vials were then closed and refluxed at 130°C on a hot plate for 4 hours. After cooling, the
145 digest solution was evaporated at 110°C until near dryness. Then, $400 \mu\text{L}$ of concentrated HNO_3 (Ultrapur
146 grade, Merck) was added, and the solution was re-evaporated at 110°C . Finally, the obtained residue was
147 dissolved with 3mL of a 0.8 mol L^{-1} HNO_3 (Ultrapure grade, Merck). This archive solution was transferred to an
148 acid cleaned 15 mL polypropylene centrifuge tube (Corning®) and stored at 4°C until analyses.

149 All analyses were performed on a sector field inductively coupled plasma mass spectrometer (SF-ICP-MS
150 Element2, Thermo-Fisher Scientific). Samples were diluted by a factor of 7 on the day of analysis in acid-
151 washed 13 mm (outer diameter) rounded bottom, polypropylene centrifuge tubes (VWR) with 0.8 mol L^{-1} HNO_3
152 (Ultrapur grade, Merck) spiked with $1\mu\text{g L}^{-1}$ of Indium (^{115}In) solution in order to monitor the instrument drift.
153 Samples were introduced with a PFA-ST nebulizer connected to a quartz cyclonic spray chamber (Elemental
154 Scientific Incorporated, Omaha, NE) via a modified SC-Fast introduction system consisting of an SC-2
155 autosampler, a six-port valve and a vacuum-rinsing pump. The autosampler was contained under a HEPA
156 filtered unit (Elemental Scientific). Two 6-points, matrix-matched multi-element standard curves with

157 concentrations bracketing the range of the samples were run at the beginning, the middle and the end of each
158 analytical run. Analytical replicates were made every 10 samples, while accuracy was determined by performing
159 digestions of the certified reference material BCR-414 (plankton, Community Bureau of Reference,
160 Commission of the European Communities), PACS-3 and MESS-4 (marine sediments, National Research
161 Council Canada), following the same protocol as for samples. Recoveries were typically within 10% of the
162 certified values (and within the error of the data, taken from replicate measurements, Table 1). Once all data
163 were normalized to an ¹¹⁵In internal standard and quantified using an external standard curve, the dilution factor
164 of the total digestion was accounted for. Obtained element concentrations per filter (pmol/filter) were then
165 corrected by the process blanks described above. Finally, pmol/filter values were divided by the volume of
166 water filtered through stacked filters.
167 Total concentrations (sum of small size fraction (0.45-5 µm) and large (>5 µm) size fraction) of particulate trace
168 elements are reported in Table S1 (supplementary data).

169
170

171 2.4. Ancillary data:

172 ~~Particulate barium (Ba) concentrations were determined in samples collected using a standard CTD rosette~~
173 ~~equipped with 12 L Niskin bottles. Typically, 18 samples were collected at each station within the first 1000 m.~~
174 ~~Details on analytical procedures are given in Lemaitre et al. (in press, 2018a). Briefly, particulate biogenic~~
175 ~~Barium, or excess Barium (Ba_{ex}), were calculated by subtracting the particulate lithogenic barium (PBa_{litho})~~
176 ~~from the total particulate barium (PBa). The PBa_{litho} was determined by multiplying the particulate aluminium~~
177 ~~(PAI) concentration by the upper continental crust (UCC) Ba: Al molar ratio (0.00135 mol mol⁻¹; Taylor and~~
178 ~~Melelland, 1985). Potential temperature (θ), salinity (S), and transmissometry data were retrieved from the CTD~~
179 ~~sensors (CTD SBE911 equipped with a SBE43).~~

180
181

181 2.5. Positive matrix factorisation

182 Positive Matrix Factorisation (PMF) was run to characterise the main factors influencing the particulate trace
183 elements variances along the GEOVIDE section. In addition to PFe, PAI, PMn, and PP, nine additionnal
184 elements were included in the PMF: Y, Ba, Pb, Th, Ti, V, Co, Cu and Zn. The analysis has been conducted on
185 samples where all the 13 elements previously cited were above the detection limits; after selection, 445 of the
186 549 existing data points were used. Analyses were performed using the PMF software, EPA PMF 5.0,
187 developed by the USA Environmental Protection Agency (EPA). Models have been tested with several factors
188 number (from 3 to 6), after full error estimation of each model, we decide to use the configuration providing the
189 lowest errors estimations and in consequence the most reliable.
190 In consequence, models were set up with four factors and were run 100 times to observe the stability of the
191 obtained results. After displacement, error estimations and bootstraps error estimations, the model was
192 recognised as stable.

193
194

194 **3. Results**

195

3.1. Hydrography and biological setting

196 Here, we briefly describe the hydrography encountered during the GEOVIDE section (Figure 2), as a thorough
197 description is available in García-Ibáñez et al. (2015). The warm and salty Mediterranean Water (MW, $S=36.50$,
198 $\theta^{\circ}=11.7^{\circ}\text{C}$) was sampled between 600 and 1700 m in the Iberian Abyssal Plain (IAP). MW resulted from the
199 mixing between the Mediterranean Overflow Water plume coming from the Mediterranean Sea and local
200 waters. Surface water above the Iberian Shelf was characterised by low salinity ($S=34.95$) at station 2 and 4
201 compared to surrounding water masses. Close to the floor of the Iberian Abyssal Basin, the North East Atlantic
202 Deep Water (NEADW, $S=34.89$, $\theta^{\circ}=2.0^{\circ}\text{C}$) spread southward. The ~~East~~ North Atlantic Central Water
203 (~~E~~NACW, $S>35.60$, $\theta^{\circ}>12.3^{\circ}\text{C}$) was the warmest water mass of the transect and was observed in the subsurface
204 layer of the Western European Basin and Iberian Abyssal Plain. An old Labrador Sea Water (LSW, $S=34.87$,
205 $\theta^{\circ}=3.0^{\circ}\text{C}$) flowed inside the Western European and Icelandic Basins, between 1000 and 2500m depth. In the
206 Icelandic Basin, below the old LSW, the Iceland-Scotland Overflow Water (ISOW, $S=34.98$, $\theta^{\circ}=2.6^{\circ}\text{C}$) spread
207 along the Reykjanes Ridge slope. This cold water, originating from the Arctic, led to the formation of NEADW
208 after mixing with surrounding waters. North Atlantic hydrography was impacted by the northward flowing of
209 the North Atlantic Current (NAC), which carried up warm and salty waters from the subtropical area. When
210 NAC crossed the Mid-Atlantic ridge through the Charlie-Gibbs Fracture Zone (CGFZ), it created the Subpolar
211 Mode Water (SPMW). The recirculation of SPMW inside the Icelandic and Irminger Basins led to the formation
212 of regional modal waters: the Iceland Subpolar Mode Water (IcSPMW, $S=35.2$, $\theta^{\circ}=8.0^{\circ}\text{C}$) and the Irminger
213 Subpolar Mode Water (IrSPMW, $S=35.01$, $\theta^{\circ}=5.0^{\circ}\text{C}$) respectively. IcSPMW was a relatively warm water mass
214 with potential temperature up to 7°C (García-Ibáñez et al., 2015). Another branch of the NAC mixed with
215 Labrador Current waters to form the relatively fresh SubArctic Intermediate Water (SAIW, $S<34.8$,
216 $4.5^{\circ}\text{C}<\theta^{\circ}<6^{\circ}\text{C}$). The Irminger Basin is a ~~really~~ complex area with a multitude of water masses. In the middle of
217 the basin, an old LSW, formed one year before (Straneo et al., 2003), spread between 500 and 1200 m depth.
218 Close to the bottom, the Denmark Strait Overflow Water (DSOW, $S=34.91$) flowed across the basin. Greenland
219 coastal waters were characterised by low salinity values, down to $S=33$. The strong East Greenland Current
220 (EGC) flowed southward along the Greenland shelf in the Irminger Basin. When reaching the southern tip of
221 Greenland, this current entered the Labrador Basin along the west coast of Greenland and followed the outskirts
222 of the basin until the Newfoundland shelf. In the Labrador Basin, the deep convection of SPMW at 2000 m was
223 involved in the formation of the LSW ($S=34.9$, $\theta^{\circ}\text{C}=3.0^{\circ}\text{C}$) (García-Ibáñez et al., 2015; Yashayaev and Loder,
224 2009). Above the Newfoundland Shelf, surface waters were affected by discharge from rivers and ice-melting
225 and characterised by extreme low salinity for open ocean waters, below 32 in the first 15 meters.

226

~~227 During GEOVIDE, diatoms and type 6 haptophytes dominated the bloom close to the IM, while type 6~~
~~228 haptophytes and dinophytes were dominant in the WEB province (Tonnard et al., in prep.). The IB bloom was~~
~~229 dominated by type 6 haptophytes and the IrB was dominated by diatoms. GS and NS coastal stations were~~
~~230 almost exclusively composed of large diatoms. Finally, the LB was dominated by diatoms and type 6 and 8~~
~~231 haptophytes. The NS, LB, GS and IrB provinces (stations 44 to 77) were sampled just after the bloom peak. The~~
~~232 LB was characterized by an intense particulate organic carbon (POC) export and high remineralization activity~~

233 ~~(Lemaitre et al., 2018a). In contrast, low remineralization fluxes and high POC exports were determined within~~
234 ~~the IB and WEB provinces, where the bloom was still active (Lemaitre et al., 2018a, b).~~

235 3.2. Section overview

236 Total particulate iron (PFe), aluminium (PAI), manganese (PMn) and phosphorus (PP) concentrations spanned a
237 large range of concentrations from below detection to 304, 1544, ~~21.5~~, 3.5 and 402 nmol L⁻¹ respectively.

238 PFe, PAI, and PMn were predominantly found (>90%) in particles larger than 5 µm, except in surface waters,
239 where ~~209% ± 8.6 %~~ of PFe, ~~38.8 ± 8.6 %~~ of PP, ~~3510.9 ± 15.4 %~~ of PAI and ~~32.8 ± 16.6 % up to 60%~~ of PMn
240 were hosted by smaller particles (0.45-5 µm). The ranges of concentrations are comparable to other studies
241 recently published (Table 2). Data are shown in Figure 3.

242

243 3.3. Open Ocean stations :Iberian Abyssal Plain (stations 11 to 17), Western European Basin 244 (stations 19 to 29), Icelandic Basin (stations 32 to 36), Reykjanes Ridge (station 38), Irminger Basin 245 (stations 40 to 60; except Stations 53 and 56) and Labrador Basin (stations 63 to 77)

246 ~~In the open ocean, p~~Particulate iron concentration vertical profiles presented identical patterns at open ocean
247 ~~stations sampled in every~~each oceanic basins encountered along the ~~GEOVIDE~~ section. Median PFe were low
248 at 0.25 nmol L⁻¹ (~~n=???~~) within the first 100 m and steadily increased with depth. However, at two stations,
249 elevated concentrations were determined in the upper 100m, up to 4.4 nmol L⁻¹ at station 77 at 40 m and 7 nmol
250 L⁻¹ at station 63 between 70 and 100 m depth. PFe concentrations ~~were increasing~~ gradually increased with
251 depth, with a median PFe of 1.74 nmol L⁻¹ below 1000m (~~n=???~~), ~~to reach a maximum close to the bottom.~~
252 ~~Close to the seafloor of some stations (26, 29, 32, 34, 49, 60, and 71), high concentrations of PFe were~~
253 observed, up to 88 nmol L⁻¹ (station 71 at 3736 m). These high PFe values were associated with low beam
254 transmissometry value inferior or equal to 97 % (Figure 9b and supplementary table S2).

255 Particulate aluminium (PAI) and manganese (PMn) profiles were similar to PFe profiles, with low median
256 ~~value~~concentrations were ~~observ~~measured in the first 100 m (1.88 nmol L⁻¹ (~~n=??~~) and 55 pmol L⁻¹ (~~n=??~~),
257 respectively) and increased towards the seafloor. Close to the seafloor, high concentrations were determined at
258 the same stations cited above for PFe, with a ~~where high value were observed at certain stations (same than~~
259 ~~PFe) with~~ maximum of 264 nmol L⁻¹ and 3.5 nmol L⁻¹ for PAI and PMn, respectively at station 71 ~~;~~
260 (supplementary Table S1). Highest particulate phosphorus (PP) concentrations were ~~maximum observed in the~~
261 first 50m, with a median value of 66 nmol L⁻¹ (~~n=???~~). Deeper in the water column, below 200m, PP
262 concentrations decreased to values ~~under~~below 10 nmol L⁻¹ ~~below 200 meters depth~~. Inter-basins differences
263 were observed within the surface samples, with Irminger basin had the higher median PP concentration being
264 higher in the Irminger Basin (127 nmol L⁻¹), ~~n=???~~ of the section while ~~thethan in the Iberian Abyssal Plain~~
265 ~~had the lower median (28 nmol L⁻¹), n=???~~.

266 Finally, above the Reykjanes Ridge, PP, PMn, PAI and PFe concentrations were in the same range than the
267 surrounding open ocean stations. However, close to the seafloor, high concentrations were measured, with PFe,
268 PAI, and PMn reaching 16.2 nmol L⁻¹, 28.8 nmol L⁻¹, and 0.51 nmol L⁻¹ at 1354 m, respectively (Figure 3 and
269 ~~supplementary material Table S1~~Table 1, ~~supplementary material and Figure xxx~~).

270
271 3.4. Margins and Shelves: Iberian Margin (stations 1 to 4), Greenland coast (stations 53, 56
272 and 61) and Newfoundland Shelf (station 78)

273
274 The Iberian margin was characterised by low beam transmissometry values at station 2 (88% at 140 m, Figure
275 9b) suggesting significant particle concentrations. Particulate iron concentrations varied between 0.02 nmol L⁻¹,
276 station 2 to 304 nmol L⁻¹, Station 2. In Within the first 50m, PFe concentrations decreased from the Iberian
277 Shelf (Station 2, 2.53 nmol L⁻¹) towards the shelf break where PFe dropped down from 2.53 nmol L⁻¹ to
278 (station 2, depth) to 0.8 nmol L⁻¹ (Station 1). PFe concentrations increased with depth at all three stations and
279 reached a maximum at the bottom of station 2 (138.5 m) with more than 300 nmol L⁻¹ of PFe. Lithogenic
280 tracers, such as PAI or PMn, presented similar profiles to PFe with concentrations ranging between 0.11 and
281 1544 nmol L⁻¹, and from below detection limit to 2.51 nmol L⁻¹ respectively (station, depth) (Figure 3, Table 1,
282 supplementary material Table S1, Figure xxxxx). Total particulate phosphorus (PP) concentrations were
283 relatively low in surface ranging from undetectable values to 38 nmol L⁻¹ (station xxxxx) (Table 1, supplementary material, Figure
284 xxxxx) in surface; then Maximum PP was measured in surface at Station 1 (20 m depth), then concentrations
285 decreased with depth and were less than 0.7 nmol L⁻¹ below 1000 m depth.
286 Particulate Fe concentrations in In the vicinity of the Greenland shelf, PFe concentrations had a high median
287 concentration value of 10.8 nmol L⁻¹ (n=???) while and were associated with high median PAI and PMn also had
288 high median concentrations of 32.3 nmol L⁻¹ (n=???) and 0.44 nmol L⁻¹ (n=???) , respectively. Concentrations of
289 PP were high at the surface with a value of 197 nmol L⁻¹ at 25 m of station 61. Then, PP concentrations
290 decreased strongly, less than 30 nmol L⁻¹, below 100 meters depth. Furthermore, beam transmissometry values
291 in surface waters at these three stations, were the lowest of the entire section, with values below 85 %.
292 Close to the Newfoundland margin, surface waters displayed a small load of particulate trace metals as PFe,
293 PAI, and PMn were below 0.8 nmol L⁻¹, 2 nmol L⁻¹, and 0.15 nmol L⁻¹ respectively. Then close to the bottom of
294 station 78, at 371 m, beam transmissometry values dropped to 94% and were associated with extremely high
295 concentrations of PFe=168 nmol L⁻¹, PAI=559 nmol L⁻¹, and PMn=2 nmol L⁻¹. Total PP concentrations in the
296 first 50 m ranged from 35 to 97 nmol L⁻¹. Below the surface, PP remained relatively high with values up to 16
297 nmol L⁻¹ throughout the water column. (Table 1, supplementary material and Figure XXXX3 and supplementary
298 material Table S1).

299 3.5. Reykjanes Ridge (station 38)

300 Above the Reykjanes Ridge, the upper portion of the Mid Atlantic Ridge, particulate trace
301 elements concentrations were in the same range than the surrounding open ocean stations. However,
302 close to the seafloor, high concentrations were measured on the ridge, with PFe, PAI, and PMn
303 reaching 16.2 nmol L⁻¹, 28.8 nmol L⁻¹, and 0.51 nmol L⁻¹ at 1354 m, respectively. Low concentrations
304 of PP were measured in surface waters, with a median value of 24.8 nmol L⁻¹ in the top 100 m and a
305 maximum of only 72.6 nmol L⁻¹ at 20 m. 3.3. Iberian Margin (stations 1 to 4)

306 The Iberian margin was characterised by low beam transmissometry values at station 2 (88% at 140 m)
307 suggesting significant particle concentrations. Particulate iron concentrations varied between 0.02 nmol L⁻¹ (20
308 m) to 304 nmol L⁻¹ (138 m) in this area. Above the Iberian Shelf, high PFe concentrations were measured in

309 surface (Station 2, 2.53 nmol L⁻¹); then, on the shelf break, surface concentrations dropped down to 0.8 nmol L⁻¹
310 (Station 1 at 20 m depth). PFe concentrations increased with depth at all three stations and reached a maximum
311 at the bottom of station 2 (138.5 m) with more than 300 nmol L⁻¹ of PFe. Lithogenic tracers, such as PAI or
312 PMn, presented similar profiles to PFe with concentrations ranging between 0.11 and 1544 nmol L⁻¹, and from
313 below detection to 2.51 nmol L⁻¹ respectively. The highest concentrations were also measured at the bottom of
314 station 2 (138.5 m). Total particulate phosphorus (PP) concentrations were relatively low in this area ranging
315 from undetectable values to 38 nmol L⁻¹. Maximum PP was measured in surface at Station 1 (20 m depth), then
316 concentrations decreased with depth and were less than 0.7 nmol L⁻¹ below 1000 m depth.

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3.4. Iberian Abyssal Plain (stations 11 to 17) and Western European Basin (stations 19 to 29)

320 In the Iberian Abyssal Plain (IAP) and the Western European Basin (WEB), particulate iron concentration
321 vertical profiles were similar (Figure 4); median PFe concentrations were 0.18 nmol L⁻¹ in the first 100 m and
322 steadily increased with depth. Close to the seafloor, concentrations of PFe were up to 1.4 nmol L⁻¹ at every
323 station and reached values superior to 8 nmol L⁻¹ at stations 26 and 29, with low beam transmissometry
324 (<98%). Particulate aluminium profiles matched the PFe profiles, with low median concentrations within the
325 first 100m of 1.77 nmol L⁻¹ and 26 pmol L⁻¹ respectively. Then, concentrations increased with depth to reach a
326 maximum close to the oceanic floor. At stations 26 and 29, total PAI concentrations reached high values, up to
327 42 nmol L⁻¹. In the Western European Basin, PMn concentrations ranged from below detection to 0.36 nmol L⁻¹;
328 except close to the bottom of stations 26 and 29, where high concentrations of 0.91 and 1.31 nmol L⁻¹ were
329 measured, respectively. Particulate phosphorus profiles, while similar between stations of this basin, differed a
330 lot from the other element profiles. In the WEB, surface median PP concentration was two times higher than in
331 the Iberian margin (60 nmol L⁻¹ against 28 nmol L⁻¹ in the first 50 m with a maximum of 162 nmol L⁻¹ (station
332 21). Concentrations dropped drastically with depth and remained under 10 pmol L⁻¹ below 100 m.

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3.5. Icelandic Basin (stations 32 to 36)

336 Concentrations of PFe were in a similar range and displayed analogous profiles to the ones collected in the
337 Western European Basin (figure 4), from below detection to 40.6 nmol L⁻¹; with low values at the surface (<1
338 nmol L⁻¹) and a progressive increase with depth. Close to the basin seafloor, low beam transmissometry (97.4%)
339 measurements were associated with high PFe concentrations of 40.6 nmol L⁻¹ at 3271 m of station 32.
340 Particulate aluminium vertical profiles were similar to those in the WEB but with extremely low surface
341 concentrations below 0.6 nmol L⁻¹; PAI then increased steadily with depth, reaching values up to 2 nmol L⁻¹
342 below 500 m. As previously observed for PFe, PAI concentrations were higher close to the seafloor, from 29
343 nmol L⁻¹ at station 34 to 101 nmol L⁻¹ at station 32. PMn also presented similar distributions than PFe and PAI.
344 Median surface concentrations were low within the first 100 m, 31 pmol L⁻¹ and 35 pmol L⁻¹, respectively, and
345 increased in the deep ocean to reach a maximum of 2.98 nmol L⁻¹ close to the seafloor. The Icelandic Basin had
346 a typical vertical profile for PP, with high concentrations at the surface, reaching 129 nmol L⁻¹ at station 32 and
347 really low concentrations below 150 m, inferior to 20 nmol L⁻¹.

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3.6. Reykjanes Ridge (station 38)

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Surface concentrations of particulate Fe, Al, and Mn above the Reykjanes Ridge (RR) were similar to the Icelandic Basin (Figure 3). However, close to the seafloor, high concentrations were measured, with PFe, PAI, and PMn reaching 16.2 nmol L^{-1} , 28.8 nmol L^{-1} , and 0.51 nmol L^{-1} at 1354 m, respectively. Low concentrations of PP were measured in surface waters, with a median value of 24.8 nmol L^{-1} in the top 100 m and a maximum of only 72.6 nmol L^{-1} at 20 m.

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3.7. Irminger Basin (stations 40 to 60; except Stations 53 and 56)

Particulate Iron, Aluminium and Manganese distributions were similar to stations sampled in the WEB, IeB and IAP provinces (Figure 3). Surface concentrations of these elements were lower than 1.1 nmol L^{-1} , 3.4 nmol L^{-1} , and 0.4 nmol L^{-1} , respectively. Then, below 50 m depth, concentrations of PFe, PAI, and PMn increased and reached high values close to the seafloor, especially at stations 42 and 44; reaching up to 40 nmol L^{-1} , 90 nmol L^{-1} , and 1.5 nmol L^{-1} respectively. Close to the Greenland margin, at the bottom of stations 49 and 60, concentrations of particulate trace metals were also elevated with PFe greater than 10 nmol L^{-1} . Particulate phosphorus concentrations were relatively high in surface waters, of the Irminger Basin, with a median value of 127 nmol L^{-1} within the first 50 m. Particulate phosphorus decreased with depth and remained constant below 500 m with concentration below 10 nmol L^{-1} .

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3.8. Greenland coast (stations 53, 56 and 61)

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Particulate Fe concentrations in the vicinity of the Greenland shelf had a high median concentration of 10.8 nmol L^{-1} , while PAI and PMn also had high median concentration of 32.3 nmol L^{-1} and 0.44 nmol L^{-1} , respectively. Concentrations of PP were maximum at the surface with a value of 197 nmol L^{-1} at 25 m of station 61. Then, PP concentrations decreased strongly with depth with values below 30 nmol L^{-1} below 100 meters depth. Furthermore, beam transmissometry values in surface waters at these two stations, were the lowest of the entire section, with values below 85 %.

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3.9. Labrador Basin (stations 63 to 77)

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In the Labrador Basin, median concentrations of PFe within the first 100 m were low, with a median value of 0.9 nmol L^{-1} (n=30). However, at two stations, elevated concentrations were determined, up to 4.4 nmol L^{-1} at station 77 at 40 m and 7 nmol L^{-1} at station 63 between 70 and 100 m depth. Below the surface waters, PFe remained constant with depth until in proximity of the seafloor (Fig. 3). Between stations 64 and 71, the median concentration between 100 m and 200 m above the seafloor was 2.0 nmol L^{-1} (n=39). Particulate Fe concentration at station 63, close to the Greenland margin, remained constant below 100 m depth, with a high median value of 5.7 nmol L^{-1} . On the other side of the Labrador Basin, station 77, close to the Newfoundland margin, constant PFe values of 3 nmol L^{-1} between the surface and 200 m above bottom depth were observed.

384 ~~As previously described, PFe concentration increased close to the seafloor to 88 nmol L⁻¹ at station 71 at 3736~~
385 ~~m. Particulate Al and Mn displayed similar characteristics to PFe, with low median concentrations at the surface~~
386 ~~of 3.37 nmol L⁻¹ and 90 pmol L⁻¹ respectively. Close to the seafloor of Station 71, at 3736 m depth, PAl, and~~
387 ~~PMn reached high concentrations of 264 and 3.5 nmol L⁻¹. Particulate Phosphorus distribution was no different~~
388 ~~than in the eastern basins, with 71 nmol L⁻¹ median PP within the first 50 m. Then below 50 m, the~~
389 ~~concentration dropped off quickly to a median PP of 3 nmol L⁻¹.~~

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391 3.10. Newfoundland Shelf (station 78)

392 ~~Close to the Newfoundland margin, surface waters displayed a small load of particulate trace metals as PFe,~~
393 ~~PAl, and PMn were below 0.8 nmol L⁻¹, 2 nmol L⁻¹, and 0.15 nmol L⁻¹ respectively. Then close to the bottom, at~~
394 ~~371 m, beam transmissometry values dropped to 94% and were associated with extremely high~~
395 ~~concentrations of lithogenic elements: PFe=168 nmol L⁻¹, PAl=559 nmol L⁻¹, and PMn=2 nmol L⁻¹. Total PP~~
396 ~~concentrations in the first 50 m ranged from 35 to 97 nmol L⁻¹. Below the surface, PP remained relatively high~~
397 ~~with values up to 16 nmol L⁻¹ throughout the water column.~~

398

399 **4. Discussion**

400 Our goal in this work was to investigate mechanisms that drive the distribution of PFe in the North Atlantic, in
401 particular the different routes of supply and removal.

402 Possible candidate sources of PFe include lateral ~~mixing advection offshore~~ from ~~the different~~ margins,
403 atmospheric inputs, ~~recently melted sea ice continental run-off~~, melting ice shelves and icebergs, resuspended
404 sediments, ~~or~~ hydrothermal inputs and biological ~~pooluptake~~. Removal processes include remineralization and
405 dissolution processes.

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

409 4.1. Analysis of the principal factors controlling variance: near-ubiquitous influence of 410 crustal particles in the water column

411 ~~The positive matrix factorisation~~ Positive Matrix Factorisation (PMF) was run to characterise the main factors
412 ~~influencing the particulate trace elements variances along the GEOVIDE section. In addition to PFe, PAl, PMn,~~
413 ~~and PP, nine additional elements were included in the PMF: Y, Ba, Pb, Th, Ti, V, Co, Cu and Zn. The analysis~~
414 ~~has been conducted on samples where all the 13 elements previously cited are above the detection limits; after~~
415 ~~selection, 445 of the 549 existing data points were used. Analyses were performed using the PMF software,~~
416 ~~EPA PMF 5.0, developed by the USA Environmental Protection Agency (EPA). Models have been tested with~~
417 ~~several factors number (from 3 to 6), after full error estimation of each model, we decide to use the~~
418 ~~configuration providing the lowest errors estimations and in consequence the most reliable. results~~
419 ~~In consequence, models were set up with four factors and were run 100 times to observe the stability of the~~
420 ~~obtained results. After displacement, error estimations and bootstraps error estimations, the model was~~

421 ~~recognised as stable.~~ Results, ~~are~~ shown in Figure 5, indicate the overall variances explained by each of the 5
422 factors.

423 The first factor is characterised by lithogenic elements, representing 86.8% of the variance of PFe, 75.8% of
424 PAI and 90.5% of PTi. The second factor is correlated with both Mn and Pb and explains no less than 76.5%
425 and 77.0% of their respective variances. Ohnemus and Lam (2015) observed this co-relation between
426 manganese and lead particles and explained it by the co-transport on Mn-oxides (Boyle et al., 2005). The
427 formation of barite is causing the third factor constraining 87.7% of the Ba variance in the studied regions.
428 Biogenic barite accumulation within the mesopelagic layer is related to bacterial activity and organic
429 remineralisation (Lemaitre et al., 2018a). –A biogenic component is the fourth factor and explained most of
430 particulate phosphorus variance, 83.7%. The micronutrient trace metals, copper, cobalt and zinc, had more than
431 a quarter of their variances influenced by this factor.

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435 These results indicate that a Along the GA01 section, PFe distributions were predominantly controlled by
436 lithogenic ~~material-material~~ and to a smaller extent by remineralisation processes (as seen by a Factor 3
437 contribution of 4.1%). This does not rule out some biogenic influence on PFe distribution, especially in surface,
438 but this contribution is veiled by the high lithogenic contribution. The PMF analysis has been realised on the
439 entire dataset, in consequence, the factors described are highly influenced by the major variations of particulate
440 element concentrations (usually at the interface, i.e. margin, seafloor, surface layer....).

441 ~~–These inputs and processes are discussed below.~~

442 To further investigate the influence of crustal material on the distribution of PFe, it is instructive to examine the
443 distribution of the molar ratio of PFe/PAI along the section as a way to assess the lithogenic inputs (Lannuzel et
444 al., 2014; Ohnemus and Lam, 2015; Planquette et al., 2009) (Figure 6) along the section.

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445
446 The PFe/PAI ratio can be used to estimate the proportion of lithogenic particles within the bulk particulate
447 material. A comparison with the Upper Continental Crust (UCC) ratio of Taylor and McLennan (1995), 0.21,
448 was used to calculate the lithogenic components of particles (PFe_{litho}) following Eq. (1):

449
450
$$\%PFe_{litho} = 100 * \left(\frac{PAI}{PFe}\right)_{sample} * \left(\frac{PFe}{PAI}\right)_{UCC\ ratio} \quad (1)$$

451
452 Then the non-lithogenic PFe is simply obtained using Eq. (2):

453
454
$$\%PFe_{non_litho} = 100 - \%PFe_{litho} \quad (2)$$

455
456 Both the lithogenic and non-lithogenic fraction of PFe are estimated using ~~in fine~~ the UCC ratio. Spatial and
457 temporal variation of the lithogenic components ratio may falsely influence the estimated fraction value. –The
458 PFe_{litho} and $PFe_{excessnon_litho}$ ~~non-litho~~

459 ~~proxies are interesting tools to evaluate the importance of lithogenic and authigenic formation of Fe~~
460 ~~oxyhydroxides non-lithogenic (either biogenic or authigenic). Biogenic contributions over the section~~
461 ~~to be used with consideration.~~

462 ~~Overall, the estimated lithogenic contribution to PFe varies from 25% (station 60, 950 m) to 100% at stations~~
463 ~~located within the Western European Basin. 100% of estimated lithogenic PFe doesn't necessary mean that~~
464 ~~biogenic particles are absent; they may just be masked by the important load of lithogenic particles. Important~~
465 ~~inter-basins variations are observed along the section (Figure 6). The IAP and WEB basins are linked with high~~
466 ~~median value of the proxy %Fe_{litho}, 90%. This could be linked to a lateral advection of iron rich lithogenic~~
467 ~~particles sourced from the Iberian margin and to atmospheric particles (Shelley et al., 2017). This point is~~
468 ~~discussed with more detail in section 4.3.1. While the Iceland, Irminger and the Labrador basins are~~
469 ~~characterised with median %PFe value under 55%. An interesting feature observable was the dramatic decrease~~
470 ~~of the %PFe proxy values happening at the station 26 (Figure 6). This feature is likely be associated to the~~
471 ~~presence of the Sub-Arctic Front, located between 49.5 and 51°N latitude and 23.5 and 22°W longitude (Zunino~~
472 ~~et al., 2017). Indeed, this front which separates cold and fresh water of subpolar origin from warm and salty~~
473 ~~water of subtropical origin was clearly identifiable at station 26 by the steep gradient of the isotherms and~~
474 ~~isohalines (Figure 2).~~

475 ~~Overall, the lithogenic contribution to PFe varies from 24% (station 60, 950 m) to 100% at stations located~~
476 ~~within the Western European Basin. This could be linked to a lateral advection of iron rich lithogenic particles~~
477 ~~sourced from the Iberian margin and to a less extent atmospheric particles with the North Atlantic Central Water~~
478 ~~flowing northward (Shelley et al., 2017; Garcia Ibanez et al. (2015). This point is discussed with more detail in~~
479 ~~section 4.3.1.~~

480 ~~The most striking feature is the almost exclusive lithogenic nature of PFe from stations 1 to 26 throughout the~~
481 ~~water column, except between 1000 and 3000 m at stations 21 to 26 (Figure 6 and 7). This feature could be~~
482 ~~linked to the fact that atmospheric inputs generally dominate the supply of PFe—deposited from Saharan dust~~
483 ~~and transported via the Gulf Stream and North Atlantic Current to the WEB (Shelley et al., 2017; Garcia Ibanez~~
484 ~~et al. (2015)), even if low atmospheric fluxes were reported during our cruise.~~

486 ~~This feature at 1000 and 2500 m between stations 21 and 26 is likely be associated to the presence of the Sub-~~
487 ~~Arctic Front, located between 49.5 and 51°N latitude and 23.5 and 22°W longitude (Zunino et al., 2017).~~
488 ~~Indeed, this front which separates cold and fresh water of subpolar origin from warm and salty water of~~
489 ~~subtropical origin was clearly identifiable at station 26 by the steep gradient of the isotherms and isohalines. The~~
490 ~~fact that the WEB was sampled close to but just after the bloom maximum is limiting any higher PFe/PAI~~
491 ~~signatures (see also section 3.3.4). The intrusion of an old LSW (Garcia Ibanez et al., this issue) at stations 21 to~~
492 ~~26 between 1000 and 2500 m with a different PFe/PAI signature could explain the smaller contribution of~~
493 ~~lithogenic PFe in this depth range as atmospheric inputs to the Labrador Sea region are relatively small (Shelley~~
494 ~~et al., 2017). This is discussed with more detail in section 4.2.~~

495 4.2. Fingerprinting watermasses

496 | The GEOVIDE section crossed several distinct water masses along the North Atlantic, each of them being
497 | distinguishable by their salinity and potential temperature signatures (García-Ibáñez et al., 2015; Figure 2).
498 | Based on this study, we applied a Kruskal-Wallis test on molar PFe/PAI ratios of nine water masses (Figure 87)
499 | in order to test the presence of significant differences. Water masses for which we had less than 5 data points for
500 | PFe/PAI were excluded from this test. As the differences in the median values among the treatment groups were
501 | greater than would be expected by chance; the difference in PFe/PAI between water masses is statistically
502 | significant ($P = <0.001$).

503 | As previously seen, the lithogenic imprint is dominant in the WEB, with MW and NEADW showing PFe/PAI
504 | values close to the UCC value of $0.21 \text{ mol mol}^{-1}$. Interestingly, the PFe/PAI signature of $0.36 \text{ mol mol}^{-1}$ within
505 | the old LSW_{WEB} is probably due to the effect of biogenic inputs associated with the strong bloom encountered in
506 | the Irminger Sea than in the WEB (see section 4.3.5).

507 | While it appears that lithogenic particles are dominating the water column in the ~~WEB,WEB~~ and that some
508 | water masses have a clear PFe/PAI fingerprint, it is important to discuss the origin of these signatures, which is
509 | the purpose of the following sections.

510

511 | 4.3. Tracking the different inputs of particulate iron

512 | 4.3.1. Inputs at margins: Iberian, Greenland and Newfoundland

513 | Inputs from continental shelves and margins have been demonstrated to support high productivity in shallow
514 | coastal areas. Inputs of iron from continental margin sediments supporting the high productivity found in
515 | shallow coastal regions have been demonstrated in the past (e.g. Cullen et al. (2009), Elrod et al. (2004), Jeandel
516 | et al. (2011), Ussher et al. (2007)) and sometimes, were shown to be advected at great distances from the coast
517 | (e.g. Lam et al., 2008). ~~Moreover, freshwater inputs that are usually present in these regions can also play a key~~
518 | ~~role in the global biogeochemical cycling of trace metals (Blain and Tagliabue, 2016; Guieu et al., 1991; Martin~~
519 | ~~and Meybeck, 1979). Rivers, runoff and continental glacial melt and/or sea ice melt can also supply dissolved~~
520 | ~~and particulate iron to coastal waters, thus sustaining important phytoplankton production (Fung, 2000).~~

521 | In the following section, we will investigate these possible candidate sources in proximity of the different
522 | margins encountered. Along the GEOVIDE section, sediments at margins were of various compositions
523 | (Dutkiewicz et al., 2015). Sediments originating from the Iberian margin were mainly constituted of silts and
524 | clays (Cacador et al., 1996; Duarte et al., 2014). East Greenland margin sediments were a mixture of sands and
525 | grey/green muds, while, sediments from the West Greenland margin were mainly composed of grey/green muds
526 | (Loring and Asmund, 1996). At the western end of the section, sediments from the Newfoundland margin were
527 | composed of gravelly and sandy muds (Mudie et al., 1984). The different sediment compositions of the three
528 | margins sampled during GEOVIDE have different mineralogy/composition, which are reflected in their
529 | different PFe/PAI ratios (Figure 89). While the Iberian Margin had a PFe/PAI close to UCC ratio, ~~mainly due to~~
530 | ~~seasonal dust inputs from North Africa (Shelley et al., 2017),~~ the highest biogenic contribution ratio could be
531 | seen at the East Greenland (stations 53 and 56) and West Greenland (station 61) Margins, with median PFe/PAI

532 reaching 0.45 mol mol⁻¹. The Newfoundland margin displayed an intermediate behaviour, with Fe/Al ratios of
533 0.35 mol mol⁻¹.

534
535
536 In addition to PAI, PMn can be used as a tracer of inputs from shelf resuspension (Lam and Bishop, 2008).
537 Indeed, Mn is really sensitive to oxidation mediated by bacteria (Tebo et al., 1984; Tebo and Emerson, 1985)
538 and forms manganese oxides (MnO₂). These authigenic particles lead to an enrichment of Mn in particle
539 compositions. In order to track the influence of shelf resuspension, a percentage of sedimentary inputs “%bulk
540 sediment inputs” can be ~~calculated~~estimated using PMn/PAI ratio from GEOVIDE samples and the PMn/PAI
541 UCC value (0.0034; Taylor and McLennan, 1995) according to the following equation:

$$542 \quad \% \text{bulk sediment PMn} = 100 * \left(\frac{\text{PAI}}{\text{PMn}} \right)_{\text{sample}} * \left(\frac{\text{PMn}}{\text{PAI}} \right)_{\text{UCC ratio}} \quad (3)$$

543 This proxy is a good indicator of direct and recent sediment resuspension. We assume that particles newly
544 resuspended in water column will have the same PMn/PAI ratio than the UCC ratio leading to a “%bulk
545 sediment Mn” of 100%. ~~This value will decrease by authigenic formation of Mn oxides. This proxy assumes~~
546 ~~homogeneity of the sediment PMn/PAI ratio through the section which is maybe not completely the case at~~
547 ~~every station. In consequence, this proxy is only a tool to identify new benthic resuspension at specific location~~
548 ~~and inter-comparison between several locations is not possible.~~—When a sample presents a “%bulk sediment
549 Mn” greater than 100%, we assign a value of 100% to simplify the following discussion. —As the Mn cycle can
550 also be affected by biologic uptake (e.g. Peers and Price, 2004; Sunda and Huntsman, 1983), this proxy is only
551 used at depths where biologic activity is negligible (i.e. below 150m depth).

552 553 *The Iberian margin*

554 Coastal waters of the Iberian Shelf are impacted by the runoff for the Tagus River, which is characterised by
555 high suspended matter discharges, ranging between 0.4 to 1 × 10⁶ tons yr⁻¹, and with a high anthropogenic
556 signature (Jouanneau et al., 1998). During the GEOVIDE section, the freshwater input was observable at
557 stations 1, 2 and 4 in the first 20 m; salinity was below 35.2 psu while surrounding waters masses had salinity
558 up to 35.7 psu. Within the freshwater plume, particulate concentrations were important at station 2; ~~with at 20m,~~
559 PFe ~~was of~~ 1.83 nmol L⁻¹. Further away from the coast, the particulate concentrations remained low at 20m
560 depth, with PFe, PAI, and PMn concentrations of 0.77 nmol L⁻¹, 3.5 nmol L⁻¹, and 0.04 nmol L⁻¹, respectively at
561 station 1. The low expansion of the Tagus plume is likely due to the rapid settling of suspended matter. Indeed,
562 our coastal station 2 was already located at around 50 km of the Iberian coast and according to Jouanneau et al.
563 (1998), the surface particle load can be observable at a maximum 30km of the Tagus estuary.
564 Besides, ADCP data acquired during GEOVIDE (Zunino et al., 2017) and several studies have reported an
565 intense current spreading northward coming from Strait of Gibraltar and Mediterranean Sea, leading to a strong
566 resuspension of benthic sediments above the Iberian Shelf; ~~e.g. Biscaye and Eittrheim (1977), Eittrheim et al.~~
567 (1976), McCave and Hall (2002), Spinrad et al. (1983). The importance of the sediment resuspension was
568 observable by low beam transmissometry value (87.6%) at the bottom of station 2. This important sediment
569 resuspension led to an extensive input of lithogenic particles within the water column associated with high

570 concentrations of PFe (304 nmol L⁻¹), PAI (1500 nmol L⁻¹), and PMn (2.5 nmol L⁻¹) (Figure 3, Table S1).
571 Moreover, one hundred percent of PFe is estimated to have a lithogenic origin (Figure 140) while 100% of the
572 PMn was estimated to be the result of a recent sediment resuspension according to the %Fe_{litho} and “%bulk
573 sediment Mn” proxies (supplementary material, Table S4S2), confirming the resuspended particle input.
574 At distance from the shelf, within the Iberian Abyssal Plain, an important lateral advection of PFe from the
575 margin was observable (Figure 4410). These lateral inputs occurred at two depth ranges: between 400 and 1000
576 m ~~as~~ seen at stations 4 and 1, with PFe concentrations reaching 4 nmol L⁻¹, and between 2500 m and the bottom
577 (3575 m) of station 1, with PFe concentrations reaching 3.5 nmol L⁻¹. While 100% of PFe had a lithogenic
578 ~~origin~~signature, the sedimentary source input estimation decreased, between 40% and ~~85~~90% of the PMn
579 (Figure 4410). Transport of lithogenic particles was observable until station 11 (12.2°W) at 2500 m where PFe
580 concentration was 7.74 nmol L⁻¹ and 60% of PMn had a sedimentary origin (Figure 949). Noteworthy, no
581 particular increase in PFe, PMn or PAI was seen between 500 and 2000 m depth, where the MOW spreads,
582 which is consistent with that was observed DFe concentrations (Tonnard et al., this issue); ~~l-y-y~~ yet in contrast with
583 the dissolved aluminium values (Menzel Barraqueta et al, subm., this issue) which were high in the MOW and
584 with the study of Ohnemus and Lam (2015) that reported a maximum PFe concentration at 695 m depth
585 associated with the particle-rich Mediterranean Overflow Water (Eittrheim et al., 1976) in the IAP. However,
586 their station was located further south of our station 1. The shallower inputs observed at stations 1 and 4 could
587 therefore be attributed to sediment resuspension from the Iberian margin and nepheloid layer at depth for station
588 1.
589 Therefore, the Iberian margin appears to be an important source of lithogenic-derived iron-rich particles in the
590 Atlantic Ocean; shelf resuspension impact was perceptible until 280 km away from the margin (Station 11) in
591 the Iberian Abyssal Plain.

592

593 *South Greenland*

594 Several studies already demonstrated the importance of icebergs and sea ice as source of dissolved and
595 particulate iron (e.g. van der Merwe et al., 2011a, 2011b; Planquette et al., 2011; Raiswell et al., 2008). The
596 Greenland shelf is highly affected by external fresh water inputs as ice-melting or riverine runoff (Fragoso et al.,
597 2016), that are important sources of iron to the Greenland Shelf (Bhatia et al., 2013; Hawkings et al., 2014;
598 Statham et al., 2008).

599 Both East and West Greenland shelves (stations 53 and 60) had high concentration of particles (beam
600 transmissometry of 83%) and particulate trace elements, reaching 22.1 nmol L⁻¹ and 18.7 nmol L⁻¹ of PFe,
601 respectively (station 53 at 100m and station 61 at 136 m). During the cruise, the relative freshwater observed
602 (S<33 psu) within the first 25 meters of stations 53 and 61 were associated with high PFe (19 nmol L⁻¹), PAI (61
603 nmol L⁻¹), PMn (0.6 nmol L⁻¹) and a low beam transmissometry (≤ 85%) (Figure 409 and Table S1). Particles
604 associated were enriched in iron compared to aluminium, as PFe/PAI ratio was 0.3 within the meteoric water
605 plume. High biological production, in agreement with PP concentrations reaching 197 nmol L⁻¹ induced by the
606 supply of bioavailable dissolved iron from meteoric water (Raiswell et al., 2008; Statham et al., 2008; Tonnard
607 et al., submitted, this issue), ~~leaded~~ led to a transfer of DFe to the particulate phase. This is in line with the fact that
608 around 30% ~~of the~~ PFe had a non-lithogenic ~~and likely biogenic~~ origin. In addition, only ~~35~~40% ~~of the~~ PMn
609 originated from resuspended sediments. Interestingly, these two proxies remained constant from the seafloor to

610 | the surface (Station 49, Figure 4+10), with around 25% of the PMn of sedimentary origin, which could be due to
611 | an important mixing happening on the shelf. The lithogenic PFe could result from the release of PFe from
612 | Greenland bedrock captured during the ice sheet formation on land.

613 | The spatial extent of the off-shelf lateral transport of particles was not important on the east Greenland coast.
614 | Indeed, no visible increase of particulate trace metal concentrations was visible at the first station off-shelf,
615 | station 60 (Figure 4+10), except at 1000 m depth, where a strong increase (up to 8990%) of sedimentary PMn
616 | was seen. This is probably due to the East Greenland Coastal Current (EGCC) that was located at station 53
617 | constrained these inputs while stations 56 and 60 were under the influence of another strong current, the East
618 | Greenland-Irminger current (EGIC) (Zunino et al., 2017).

619 |
620 | To the west of the Greenland margin, lateral transport of particles was slightly more important. Noticeable
621 | concentrations of particulate lithogenic elements were observable until station 64 located 125 km away from
622 | shoreline. These particles had decreasing PFe lithogenic contribution (520%) with a similar (257%) sedimentary
623 | PMn content than closer to the margin. The increasing nature of non-lithogenic PFe is linked to the bloom in
624 | surface (associated with a PFe/PAI ratio of 0.30 mol mol⁻¹, and a PP of 197 nmol L⁻¹ at station 61 and a Chl-a
625 | concentrations of 6.21 mg m⁻³), with the biogenic PFe settling down along the transport of particles.

626 |
627 | Therefore, particles newly resuspended from Greenland sediments are an important source, representing around
628 | a third of the pMn pool, combined with surface inputs such as riverine runoff and/or ice-melting that are
629 | delivering particles on the shelf and biological production. Unlike the Iberian shelf, Greenland margin was not
630 | an important provider of particulate metals inside the Irminger and Labrador Basin, due to the circulation that
631 | constrained the extent of the margin plume.

632 | 633 | *The Newfoundland Shelf*

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

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

643 | The proportion of lithogenic PFe was relatively high and constant in the entire water column, with a median
644 | value of 67-70%. At station 78, 95-100% of the PMn had a sedimentary origin close to the seafloor (371 m). The
645 | spreading of the recent sediment resuspension was observable until 140 m depth where the contribution of
646 | sedimentary Mn was still 51% (Figure 4+10, Table S2). This could correspond to an intense nepheloid layer as
647 | previously reported by Biscaye and Eitrem (1977) (see also section 3.3.2). The high PFe concentration (184
648 | nmol L⁻¹, station 78, 371 m) associated with a high percentage of sedimentary PMn (95%) observed at the

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649 bottom of this station, was therefore the result of an important resuspension of shelf sediments. This was
650 confirmed with low transmissometry values of 95%.

651 ~~Despite t~~The important phytoplanktonic community present (~~maximum Chl-a= 4.91 mg m⁻³, Tonnard et al., in~~
652 ~~prep), is linked to the low~~ PFe ~~remained low of~~at 0.79 nmol L⁻¹ at 10 m, but, ~~with the~~ high PFe/PAI ratio, up to
653 0.4, and ~~the~~PP concentration of 97 nmol L⁻¹, ~~confirming~~ the biologic influence. Either the biogenic particles
654 settled quickly, and/or they were quickly remineralized. Concerning this latter process, intense remineralization
655 ~~at station 77 (7 mmol C m⁻² d⁻¹ compared to 4 mmol C m⁻² d⁻¹ in the Western European Basin) at station 77~~ has
656 been reported by Lemaitre et al. (2018a ~~and 2018b~~), which could explain the low PFe values throughout the
657 water column.

658 ~~That said, resuspended particles are were still laterally transported off shelf until station 71 (Figure 6) where~~
659 ~~PFe concentrations were higher than the background value, up to 2 nmol L⁻¹ at depths greater than 100 m.~~

661 Along the GEOVIDE section, continental shelves provided an important load of particles within the surrounding
662 water column. The three margins sampled during GEOVIDE behaved differently; the Iberian margin discharged
663 high quantities of lithogenic particles far away from the coast while the Greenland and Newfoundland margins
664 did not reveal important PFe concentrations. Spreading of particles is tightly linked to hydrodynamic conditions,
665 which in the case of the Greenland margin, prevented long distance seeding of PFe. Moreover, each margin
666 showed a specific PFe/PAI ratio (Figure ~~89~~) indicating different composition of the resuspended particles.
667 Resuspended particles represent the composition of sediment at the margin if oxido-reductive transformation of
668 iron and aluminium are considered negligible under these circumstances. Differences between margins were due
669 to the presence of non-crustal particles, ~~either biogenic or authigenic~~. Biological production in surface waters
670 ~~and authigenic formation of iron hydroxide~~ produces particles with a higher PFe/PAI content and their export
671 through the water column to the sediment increased the PFe/PAI ratio at depth. Regions where biological
672 production is intense such as in the vicinity of Newfoundland presented higher PFe/PAI ratios of resuspended
673 benthic particles. ~~These results are in agreement with the study of Lam et al. (2017), which showed the different~~
674 ~~behaviour between margins are a function of several parameters such as boundary currents, internal waves and~~
675 ~~margin sediment composition.~~

677 4.3.2 Benthic resuspended sediments

678 Benthic nepheloid layers (~~BNLs~~) (~~BNLs~~) can play a significant role in trace element distributions at depth as
679 ~~previously described (Dutay et al., 2015; Lam et al., 2015; Ohnemus et al., 2015; Revels et al., 2015).~~ BNLs are
680 important layers where local resuspension of sedimentary particles (Bishop and Biscaye, 1982; Eittrheim et al.,
681 1976; Rutgers Van Der Loeff et al., 2002) occur due to strong hydrographic stresses (~~i.e. boundary currents,~~
682 ~~benthic storms and deep eddies~~) interacting with the ocean floor (~~Biscaye and Eittrheim, 1977; Eittrheim et al.,~~
683 ~~1976; Gardner et al., 2017, 2018).~~ ~~In the North Atlantic, boundary currents were suspected to be the origin of~~
684 ~~theses stresses (Biscaye and Eittrheim, 1977; Eittrheim et al., 1976) but more recent studies demonstrate the~~
685 ~~essential role of benthic storms and deep eddies (Gardner et al., 2018).~~ Along the GA01 section, BNLs were
686 observable in each province with different strengths (Figures 3 and 12).

687

688 In BNLs located within the WEB, PFe concentrations reached up to 10 nmol L⁻¹ (stations 26 and 29, [Table S1,](#)
689 [supplementary material](#)). These concentrations were smaller than PFe concentrations encountered in BNL from
690 the Icelandic, Irminger and Labrador Basins, where benthic resuspension led to PFe concentrations higher than
691 40 nmol L⁻¹, even reaching 89 nmol L⁻¹ at the bottom of station 71 (3736 m). Moreover, in the Irminger and
692 Labrador Basins, PFe/PAI molar ratios within BNLs were higher than the ones measured within the WEB at
693 station 26 and 29. In the Irminger Basin, PFe/PAI reached 0.4 mol mol⁻¹, which could reveal a mixture of
694 lithogenic and biogenic matter previously exported. This feature was also observed in the Labrador Basin, with
695 PFe/PAI ratio ranging between 0.34 and 0.44 mol mol⁻¹. In contrast, BNLs sampled in the WEB have clearly a
696 lithogenic imprint, with PFe/PAI molar ratios close to the crustal one. Resuspended sediments with a non-crustal
697 contribution seem to hold a higher PFe content than sediments with a lithogenic characteristic. Nevertheless,
698 interestingly all BNLs present during GEOVIDE were spreading identically, with impacts observable up to 200
699 meters above the oceanic seafloor (Figure 112), as reflected in beam transmissometry values, and PFe
700 concentrations, that returned to a background level at 200 m above the seafloor. The presence of these BNLs has
701 also been reported by Le Roy et al. (submitted, this issue). [Important differences of PFe intensities could also be](#)
702 [due to different hydrographic components and topographic characteristics. As previously explained, two main](#)
703 [triggers of BNLs are benthic storms and deep eddies; by definition these processes are highly variable](#)
704 [geographically and temporally, but no physical data could allow us to investigate further this hypothesis.](#)
705 [Along the GEOVIDE section, BNLs are providing high concentrations of particulate trace element in the deep](#)
706 [open ocean, contributing highly to the total trace elements budget as iron.](#) ~~Important differences of PFe~~
707 ~~intensities could also be due to different hydrographic components and topographic characteristics. As~~
708 ~~previously explained, two main triggers of BNLs are benthic storms and deep eddies; by definition these~~
709 ~~processes are highly variable geographically and temporally, but no physical data could allow us to investigate~~
710 ~~further this hypothesis.~~

711
712

4.3.3. Reykjanes Ridge inputs

713 ~~Recently, hydrothermal inputs of iron in the open ocean have been re-evaluated by (Fitzsimmons et al., 2017;~~
714 ~~Resing et al., 2015; Tagliabue et al., 2014). These studies demonstrated the importance of hydrothermal~~
715 ~~activities on the global iron biogeochemical cycle through particulate and dissolved iron fluxes. During the~~
716 ~~cruise, samples of station 38 have been collected above the Reykjanes Ridge, the upper section of the Mid-~~
717 ~~Atlantic Ridge in the North Atlantic, which has inferred hydrothermal sites from several studies conducted in~~
718 ~~the area (Baker and German, 2004). Above the ridge, high PFe concentrations were measured, reaching 16 nmol~~
719 ~~L⁻¹ just above the seafloor, while increased DFe concentrations were reported to the East of the ridge (Tonnard~~
720 ~~et al., this issue). The exact sources of iron-rich particles cannot be well constrained, as they could come from~~
721 ~~active hydrothermal vents or resuspension of particulate matter from new crustal matter produced at the ridge.~~
722 ~~According to the oceanic circulation (Zunino et al., 2017; Garcia-Ibanez et al., 2017), hydrothermal particles~~
723 ~~could have been seen in the ISOW within the Icelandic Basin. Nevertheless, at the vicinity of the ridge, scanning~~
724 ~~electron microscope (SEM) analyses of our samples did reveal a number of biological debris and clays but not~~
725 ~~the presence of iron (oxy-)hydroxide particles, which are known to be highly produced close to hydrothermal~~
726 ~~vents (Elderfield and Schultz, 1996). Their absence could thus indicate an absence of vents. However, other~~

727 proxies, such as helium-3, are necessary to claim with more accuracy the presence or absence of an
728 hydrothermal source close to station 38.

729
730 ~~Alternatively, resuspended sediments transported with ISOW flowing across the Reykjanes Ridge could explain~~
731 ~~the high PFe concentrations below 1000 m depth at station 38. This feature was associated with lower median~~
732 ~~PFe concentrations and PFe/PAI ratios (Figure 7) at station 40 (2.2 nmol L^{-1} , and $0.58 \text{ mol mol}^{-1}$ respectively)~~
733 ~~than at station 38 (6.8 nmol L^{-1} , $0.48 \text{ mol mol}^{-1}$ respectively). Moreover, PMn had a 19% sedimentary origin,~~
734 ~~constant from the bottom to 1163 m depth, a contribution that is very low for the shallower water depths.~~
735 ~~Consequently, the increase in PFe within the ISOW_{west} more likely came from sediment resuspension as the~~
736 ~~ISOW_{east} flows through the Charlie Gibbs Bight Fracture Zones.~~

737

738 4.3.4. Atmospheric inputs

739 Atmospheric deposition is an important input of trace elements in surface of the open ocean (e.g. (Jickells et al.,
740 2005). Atmospheric inputs, both wet and dry, were reported to be low during the GEOVIDE cruise (Menzel-
741 barraqueta et al., 2018, this issue; Shelley et al., 2017; 2018). In fact, oceanic particles measurements in surface
742 waters along the section did not reveal high PFe or PAI concentrations, therefore, the surface composition of
743 particles did not seem to be highly affected by atmospheric deposition at the time of the cruise. ~~However,~~
744 ~~PFe/PAI ratio was closed to the UCC one, probably due to the overall influence of atmospheric deposition in~~
745 ~~this area.~~ One pattern is also interesting to note: the surface waters of the Iberian Abyssal Plain and Western
746 European Basin, between stations 11 and 23 presented a characteristic feature with really low PFe/PAI
747 elemental ratios, of 0.11, smaller than the UCC ratio of 0.21 (Figure 6). Such low ratios have been reported in
748 the same region by Barrett et al. (2012). One possible explanation is given by Buck et al. (2010) who described
749 Fe-depleted aerosols in this area of the North Atlantic with PFe/PAI ratio below UCC ratio. However, Shelley et
750 al. (2017) found a higher PFe/PAI ratio around 0.25 in this area (their sample geo5-6). This result, highlights
751 some of the difficulties that link atmospheric inputs to water column data (Baker et al., 2016), and implies a
752 probable fractionation after aerosol deposition. In addition, there is high spatial and temporal variability of
753 atmospheric deposition (Mahowald et al., 2005) and a certain degree of uncertainty about the dissolution
754 processes of atmospherically-transported particles (Bonnet and Guieu, 2004).

755

756 ~~4.3.5. Influence of phytoplankton assemblages, remineralisation and scavenging in~~ 757 ~~the upper water column~~

758 ~~Biological activity in surface waters impacts the particle composition in the upper water column. In bulk particle~~
759 ~~samples, direct measurement of the biogenic metal fraction is not possible due to the heterogeneity of particles,~~
760 ~~and in particular, the presence of lithogenic particles (Collier and Edmond, 1984). It is however possible to~~
761 ~~estimate the $\text{PFe}_{\text{nonlithic}}/\text{PFe}_{\text{excess}}/\text{PP}$ molar ratios, based on Eq. (1) and (23), and assuming that most of the PP is of~~
762 ~~biogenic origin. As 100% of the PFe was estimated to be of lithogenic origin, stations 1 to 26 are excluded from~~
763 ~~the discussion below. Overall, the median $\text{PFe}_{\text{excess}}/\text{PP}$ molar ratios varied from 1.0 (Irminger Basin) to 38.7~~
764 ~~mmol mol^{-1} (Greenland margin) in the upper 50 m. These ratios are consistent with the few available bulk~~

765 PFe/PP ratios available in the literature (Twining and Baines, 2013 and references therein), ranging from 1 to 31
766 mmol mol⁻¹ and the phytoplankton assemblages encountered during GEOVIDE (Tonnard et al., in prep.). This
767 corresponds to a % PFe_{bio} of x to x% in the upper 50m. Interestingly, the biogenic contribution is higher in the
768 basin than in the basin.

769 Overall, the median PFe_{nonlitho}/PP molar ratios varied from 1.0 (Irminger Basin) to 38.7 mmol mol⁻¹ (Greenland
770 margin) in the upper 50 m. These ratios are consistent with the few available bulk PFe/PP ratios available in the
771 literature (Twining and Baines, 2013 and references therein), ranging from 1 to 31 mmol mol⁻¹ and the
772 phytoplankton assemblages encountered during GEOVIDE (Tonnard et al., in prep.). Indeed, the highest
773 PFe_{nonlitho}/PP molar ratio determined at stations 53 and 56 close to the South Greenland margin coincide
774 with a bloom mostly composed of large diatoms, whereas, the smallest ratios were associated with a bloom
775 mainly composed of cyanobacteria and haptophytes (Tonnard et al., in prep.). The effect of biological uptake is
776 also clearly visible when looking at PFe/PAI vertical variation, which increases from the surface to
777 approximately 100m depth (Figure 13), except in the Iberian Margin, which is under the strong influence of
778 lithogenic inputs.

781 At deeper depths, pelagic remineralisation processes influence the composition of particles (Barbeau et al.,
782 1996, 2001; Boyd et al., 2010; Strzpek et al., 2005). Taking in account remineralization depths that are derived
783 from Baxs proxy which is described and discussed in great detail in Lemaitre et al. (this issue), it is possible to
784 look at the vertical variation of PFe_{bio}/PP along the section (Figure 13).

785 Close to the IM and within the IAP, no PFe/PAI decrease that could point to a preferential remineralisation of
786 PFe over PAI could be observed within the remineralisation depth (200 to 400 m depth, Figure 13), whereas
787 preferential remineralisation of PP over PFe occurs, as reflected by increasing PFe/PP ratios (Figure 14). This is
788 probably due to the fact that remineralisation rates were low (Lemaitre et al., 2018a), and that PFe was mostly of
789 lithogenic origin, more difficult and slow to remineralize (Boyd et al., 2010). Below 600 m depth, scavenging
790 processes could explain the increasing PFe:PP ratios, from 0.30 to 0.80 mol mol⁻¹ at station 13, which is
791 consistent with decreasing dFe concentrations within this depth range reported in Tonnard et al. (this issue).

792 Within the WEB, between 200 and 500 m depth, remineralisation of PFe over PAI occurs, although reported to
793 be small (Lemaitre et al., 2018a) as reflected by decreasing PFe:PAI ratios (Figure 13), while PFe:PP ratios
794 remained constant, pointing out to similar remineralisation rates of PFe and PP. Below 600 m depth, a stronger
795 scavenging of DFe onto particles formation of Fe oxyhydroxydes (si tu peux calculer et reporter les % à ces
796 intervalles de profondeur, ça aiderait) than in IM and IAP is likely to explain the increasing ratios of PFe:PAI
797 from 0.18 to 0.30 mol mol⁻¹ and PFe:PP from 0.047 to 0.367 (Station 21), and from 0.16 to 1.05 (Station 26) mol
798 mol⁻¹. Similar patterns occur in IeB (station 32), as dFe⁻ concentrations increased (Tonnard et al., this issue)
799 therefore ruling out the possibility of PFe enrichment from scavenging.

801 Above the RR, and in the IrB, at station 38, PFe_P is remineralized preferentially over PAI and PFe_o, with
802 decreasing increasing PFe:PAI ratios from 0.46 to 0.19 mol mol⁻¹ and decreasing PFe:PP ratios from 0.24 to
803 0.04 mol mol⁻¹. This interesting ese features is are associated with moderate and high POC remineralisation

804 ~~fluxes (Lemaitre et al., 2018a) at stations 38 and 44 respectively (Figure 13), and the fact that a stronger fraction~~
805 ~~of PFe was associated with biogenic lithogenic material ($PFe_{bio} = \%$, $PFe_{litho} = \%$), easier to recycle.~~

806 ~~In the IrB, PP is preferentially remineralized over PFe and PAI, as reflected by increasing PFe:PP ratios and~~
807 ~~constant PFe:PAI ratios within the remineralisation depth. This is associated with high POC remineralisation~~
808 ~~fluxes (Lemaitre et al., 2018a) and a high proportion of lithogenic PFe.~~

809 ~~Finally, within the LB, PFe:PAI and PFe:PP remained constant within the deep remineralisation depth,~~
810 ~~extending from 200 to 1000 m depth due to the deep convection of the LSW (Lemaitre et al., 2018a). In this~~
811 ~~basin, either PP and PFe are remineralized at a constant rates, but the fact that dFe is depleted (Tonnard et al.,~~
812 ~~this issue) in this depth range points out to the influence of scavenging. Below 1000 m, PFe:PP ratios increase~~
813 ~~from 0.29 to 0.85 mol mol⁻¹, while PFe:PAI ratios still remain constant. This could be explained by the fact that~~
814 ~~most PP has been recycled due to the strongest remineralisation fluxes reported in this area (Lemaitre et al.,~~
815 ~~2018a).~~

816

817 **5. Conclusions**

818 This investigation of the PFe compositions of suspended particulate matter in the North Atlantic indicates the
819 pervasive influence of crustal particles, augmented by sedimentary inputs at margins, and at depths, within
820 benthic nepehloid layers.

821 Indeed, along the GEOVIDE section, continental shelves provided an important load of particles within the
822 surrounding water column, with PFe mostly residing in non-biogenic particulate form. The Iberian margin
823 discharged high quantities of lithogenic particles originating from riverine inputs far away from the coast while
824 the Greenland margin did not reveal a long distance seeding of PFe, due to hydrodynamic conditions. Both
825 Greenland and Newfoundland margins PFe resuspended particles were under a strong biogenic influence that
826 were exported at depth. This resulted in different remineralisation fluxes among the different provinces.

827 Scavenging processes could also be visible at depths greater than 1000 m, these effects being the most
828 pronounced within the [WEBLabrador Basin](#).

829 Finally, resuspended sediments above the Reykjanes Ridge increased the PFe composition of the Iceland
830 Scottish Overflow Water. A similar feature occurs for the Labrador Sea Water, as it flows from the Irminger
831 Basin to the Western European Basin.

832

833

834

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844

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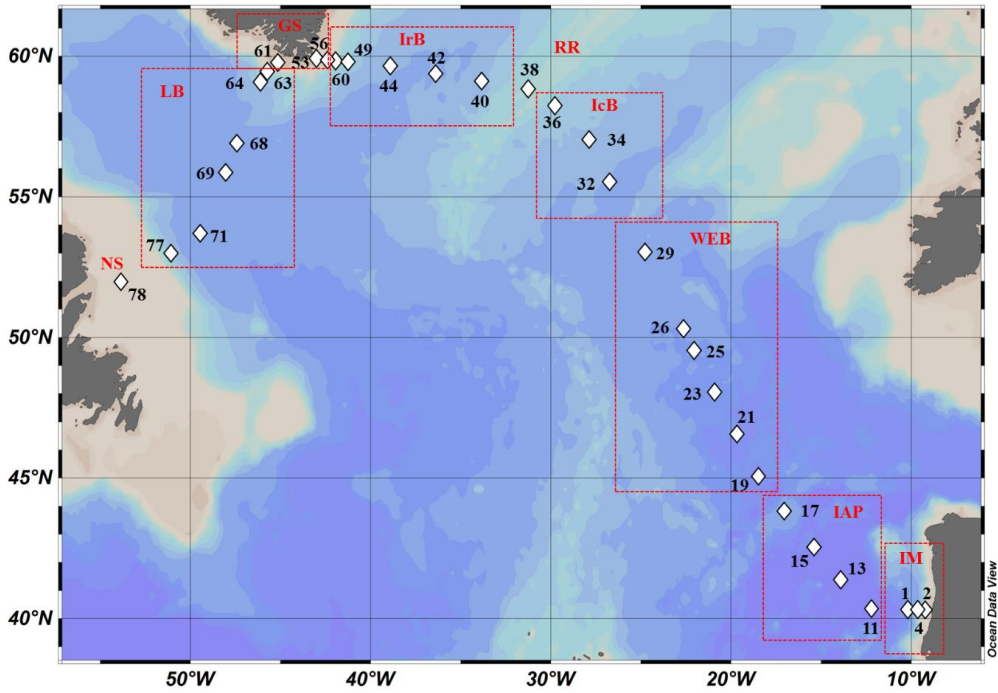
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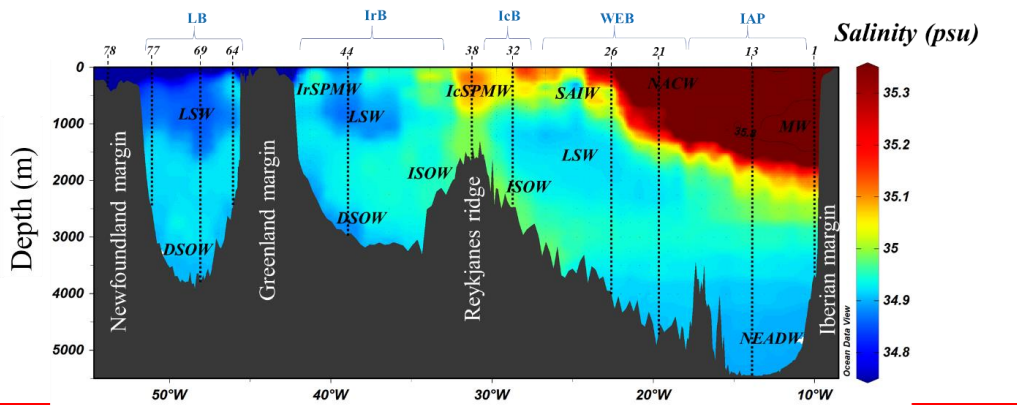
1183 **Figure 1: Map of stations where suspended particle samples were collected with GO-FLO bottles during the**
1184 **GEOVIDE cruise (GA01). Biogeochemical provinces are indicated by red squares, IM: Iberian Margin, IAP: Iberian**
1185 **Abyssal Plain, WEB: Western European Basin, IcB: Iceland Basin, RR: Reykjanes Ridge, IrB: Irminger Basin, GS:**
1186 **Greenland Shelf, LB: Labrador Basin, NS: Newfoundland Shelf. This figure was generated by Ocean Data View**
1187 **(Schlitzer, R., Ocean Data View, odv.awi.de, 2017).**



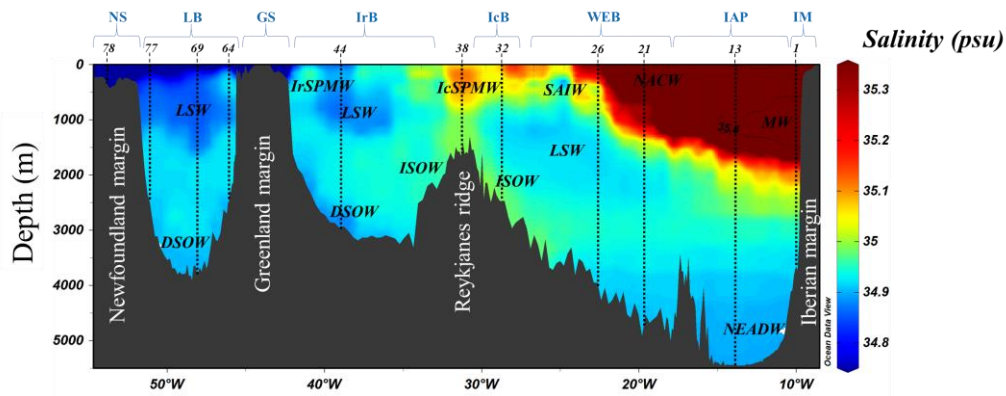
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Figure 2: Salinity section during the GEOVIDE cruise. Water masses are indicated in black, MW: Mediterranean Water; NACW: North Atlantic Central Water; NEADW: North East Atlantic Deep Water; LSW: Labrador Sea Water; ISOW: Iceland-Scotland Overflow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

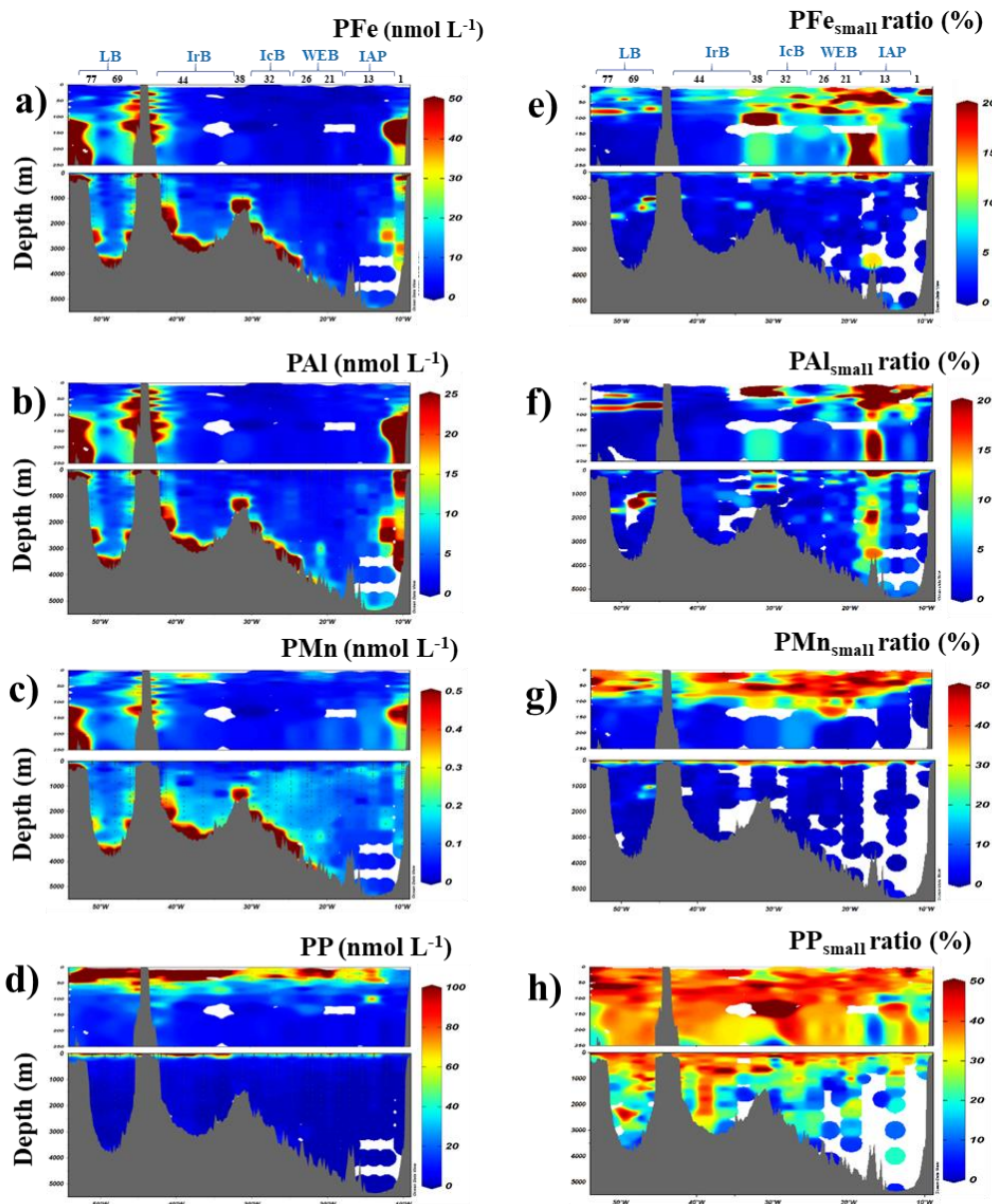


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1233 **Figure 3:** Left) Distribution of total particulate iron (a, PFe), aluminium (b, PAI), manganese (c, PMn) and
 1234 phosphorus (d, PP) concentrations (in nmol L^{-1}) along the GEOVIDE section. Right) Contribution of small size
 1235 fraction ($0.45\text{-}5\ \mu\text{m}$) expressed as percentage (%) of the total concentration of PFe (e), PAI (f), PMn (g) and PP (h).
 1236 Station IDs and biogeochemical region are indicated on top of section a. This figure was generated by Ocean Data
 1237 View (Schlitzer, R., Ocean Data View, odv.awi.de, 2017).



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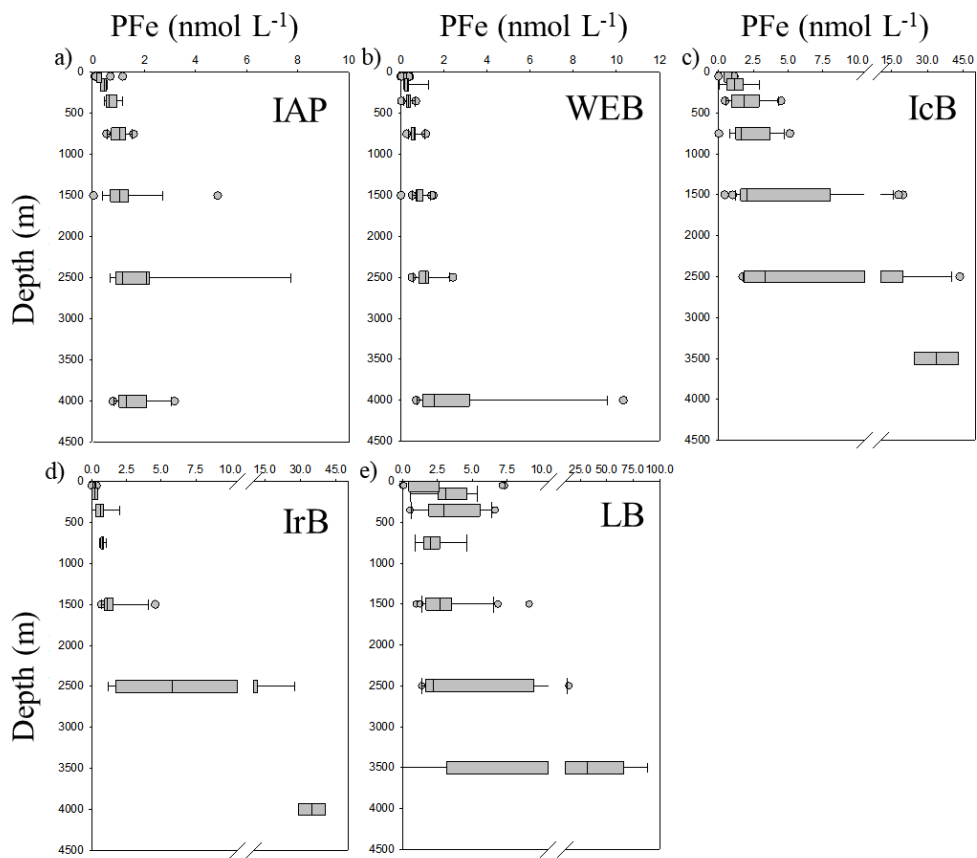
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Figure 4: Boxplot figure of the particulate iron vertical profile (in nmol L^{-1}) in the a) Iberian abyssal plain (IAP), b) Western European basin (WEB), c) Icelandic basin (IcB), d) Irminger basin (IrB) and e) Labrador basins (LB). The left boundary of the box represents the 25th percentile while the right boundary represents the 75th percentile, the line within the box marks the median value. Whiskers represent the 90th and 10th percentiles and dots are the outlying data. Seven depth boxes have been used (0-100m, 100-200m, 200-500m, 500-1000m, 1000-2000m, 2000-3000m and 3000m-bottom depth).



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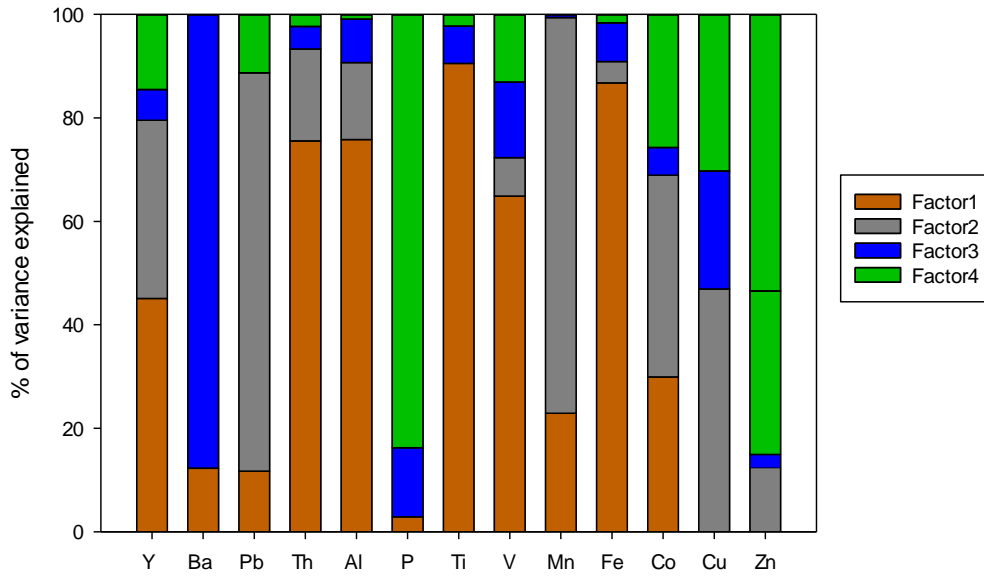
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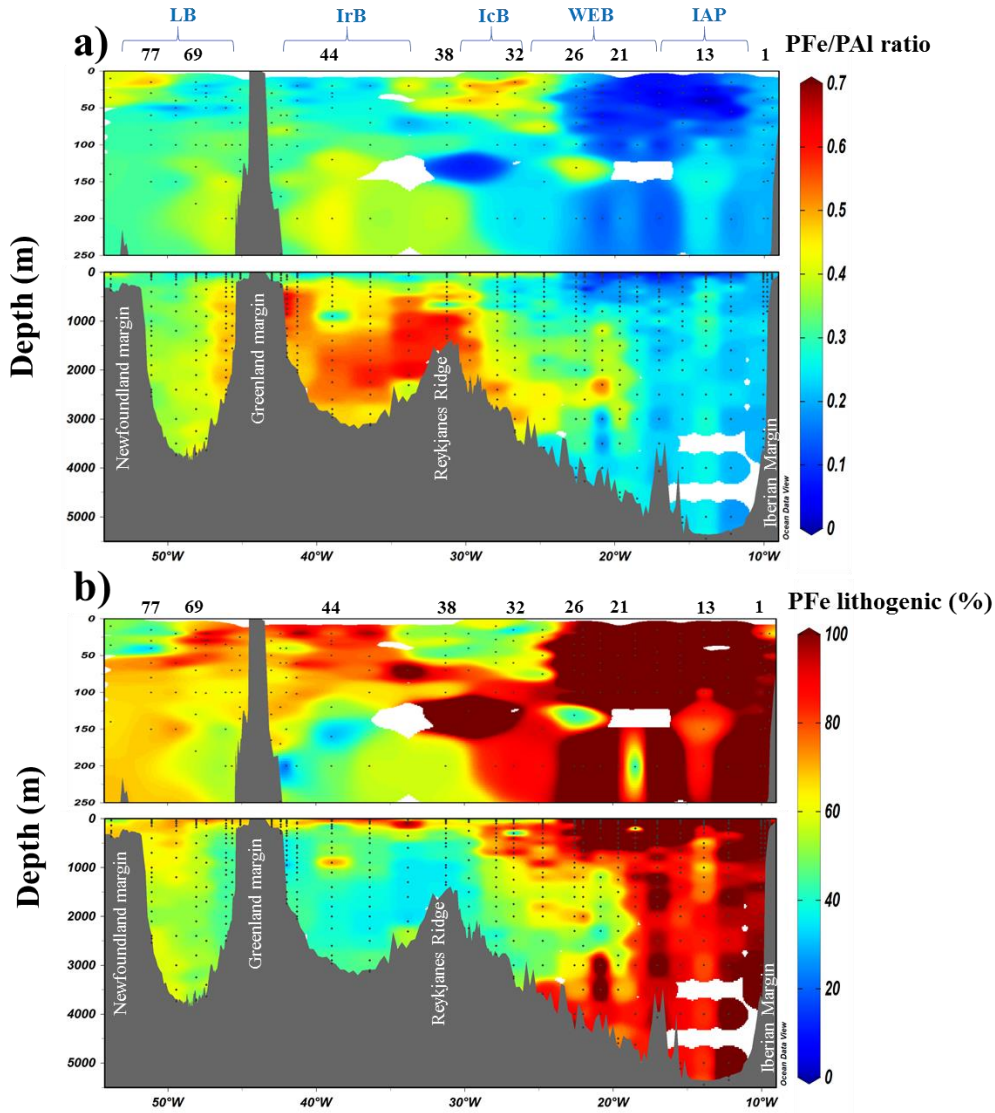
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1261 Figure 5: Factor fingerprint of the positive matrix factorisation. The four factors are represented in a stacked bar
 1262 chart of the percentage of variance explained per element.



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1282 Figure 6: a) Section of the PFe to PAI molar ratio (mol mol^{-1}); (b) contribution of lithogenic PFe (%) based on Eq. (1).
 1283 Station IDs and biogeochemical provinces are indicated above each section. This figure was generated by Ocean Data
 1284 View (Schlitzer, R., Ocean Data View, odv.awi.de, 2017).



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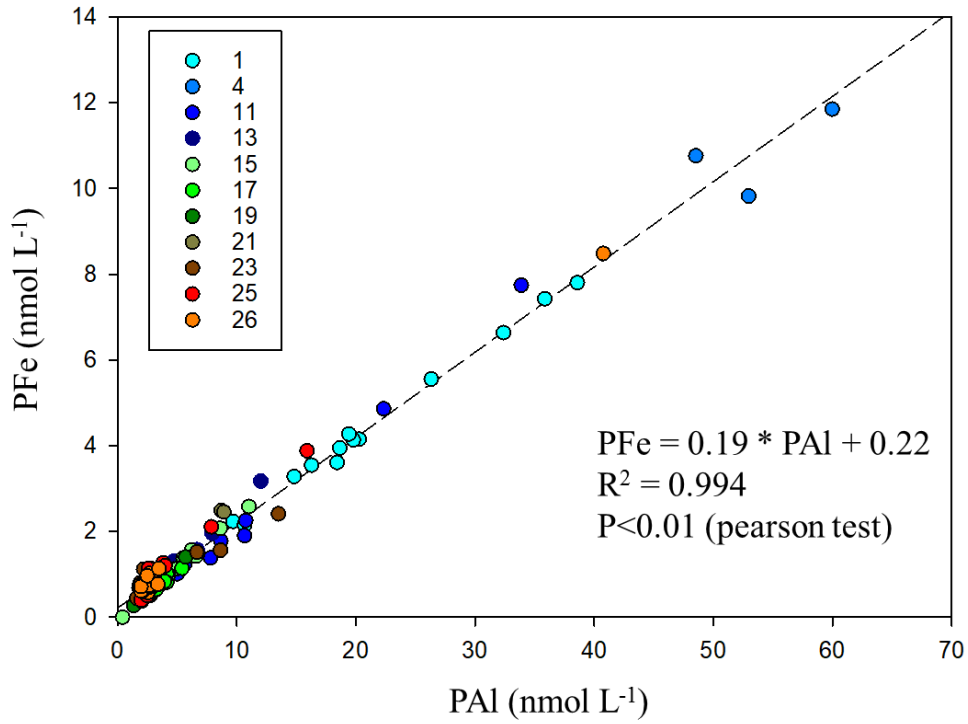
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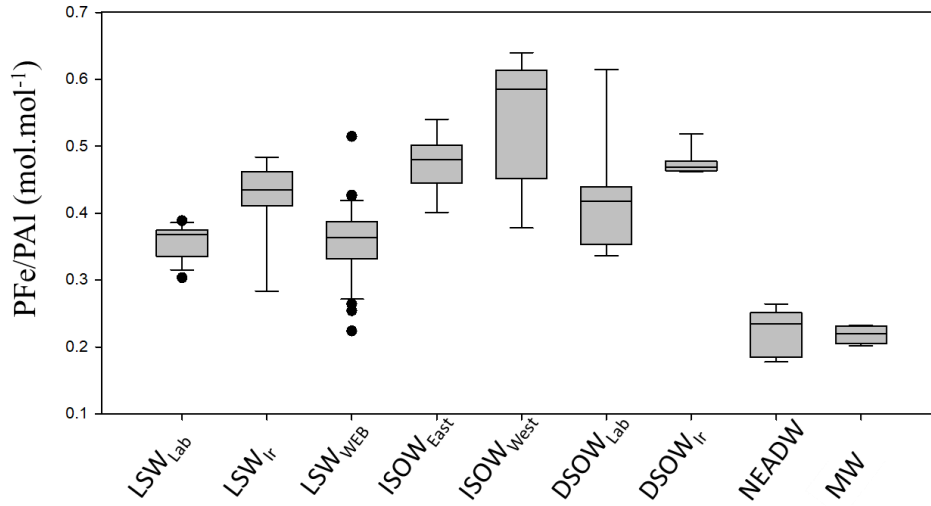
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Figure 7: PFe over PAI concentrations (nmol L^{-1}) for all stations located in the Iberian Abyssal Plain and Western European Basin. Note that the total concentrations of the two elements covaried strongly.



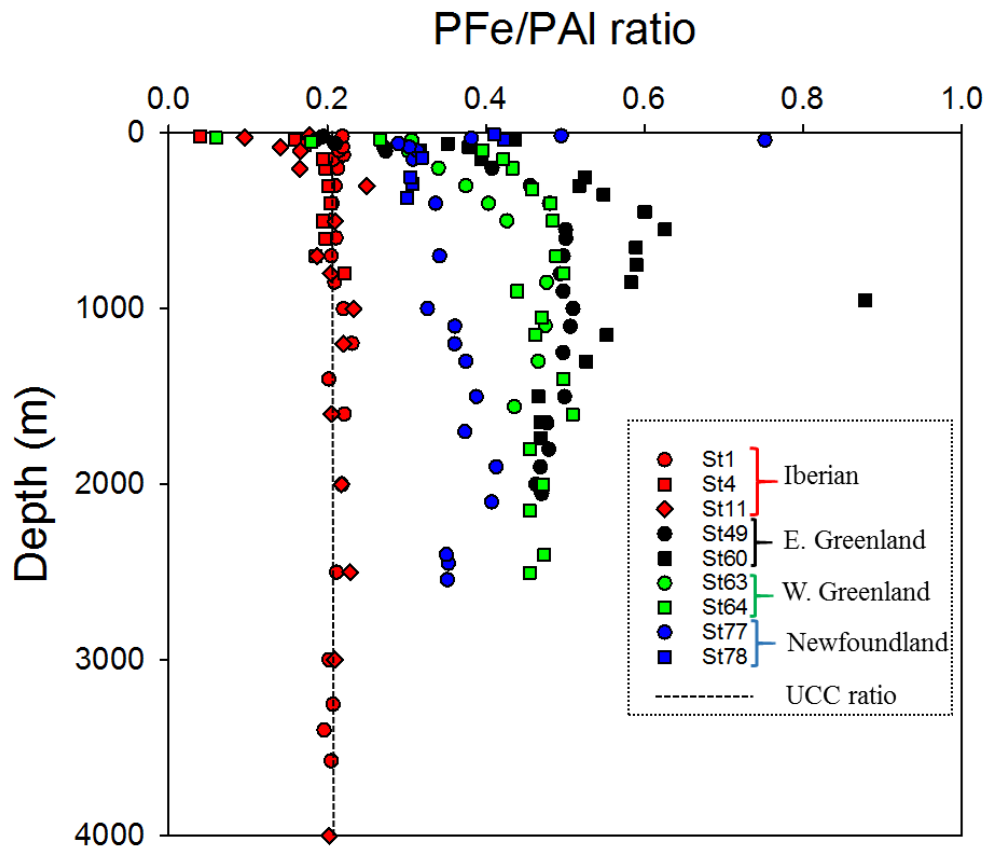
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1307 | Figure 87: Whisker diagram of PFe/PAI molar ratio (mol mol^{-1}) in the different water masses sampled along the
 1308 GA01 line. Median values for the water masses were as follows: $\text{LSW}_{\text{lab}}=0.37$; $\text{LSW}_{\text{lr}}=0.44$; $\text{LSW}_{\text{WEB}}=0.36$;
 1309 $\text{ISOW}_{\text{east}}=0.48$; $\text{ISOW}_{\text{west}}=0.58$; $\text{DSOW}_{\text{lab}}=0.42$; $\text{DSOW}_{\text{lr}}=0.47$; $\text{NEADW}=0.23$; $\text{MW}=0.22 \text{ mol mol}^{-1}$.



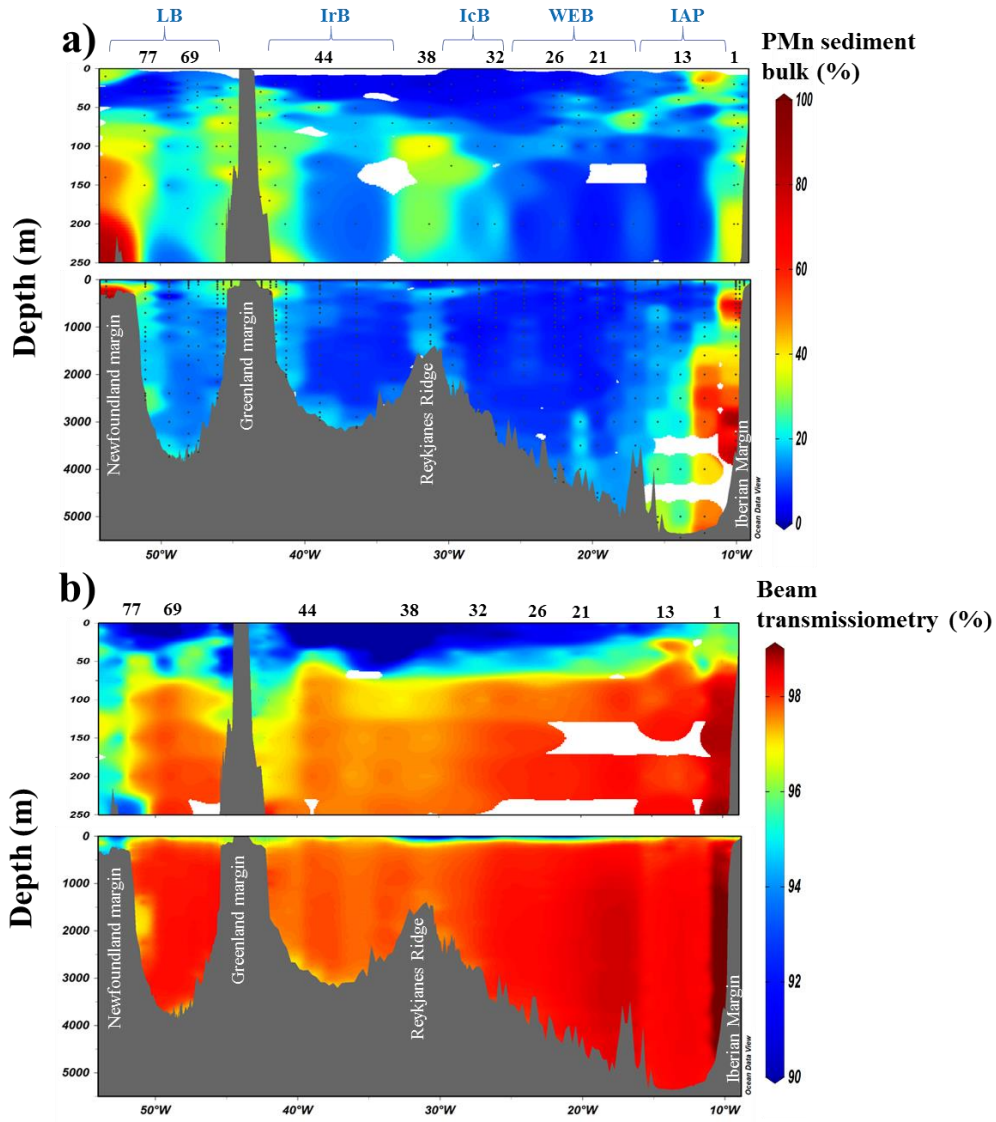
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1330 | Figure 98: Scatter of the PFe/PAI ratio at the Iberian (red dots), East Greenland (black dots), West Greenland (green
 1331 dots) and Newfoundland margins (blue dots). Dashed line indicate the UCC ratio (Taylor and McLennan, 1995).



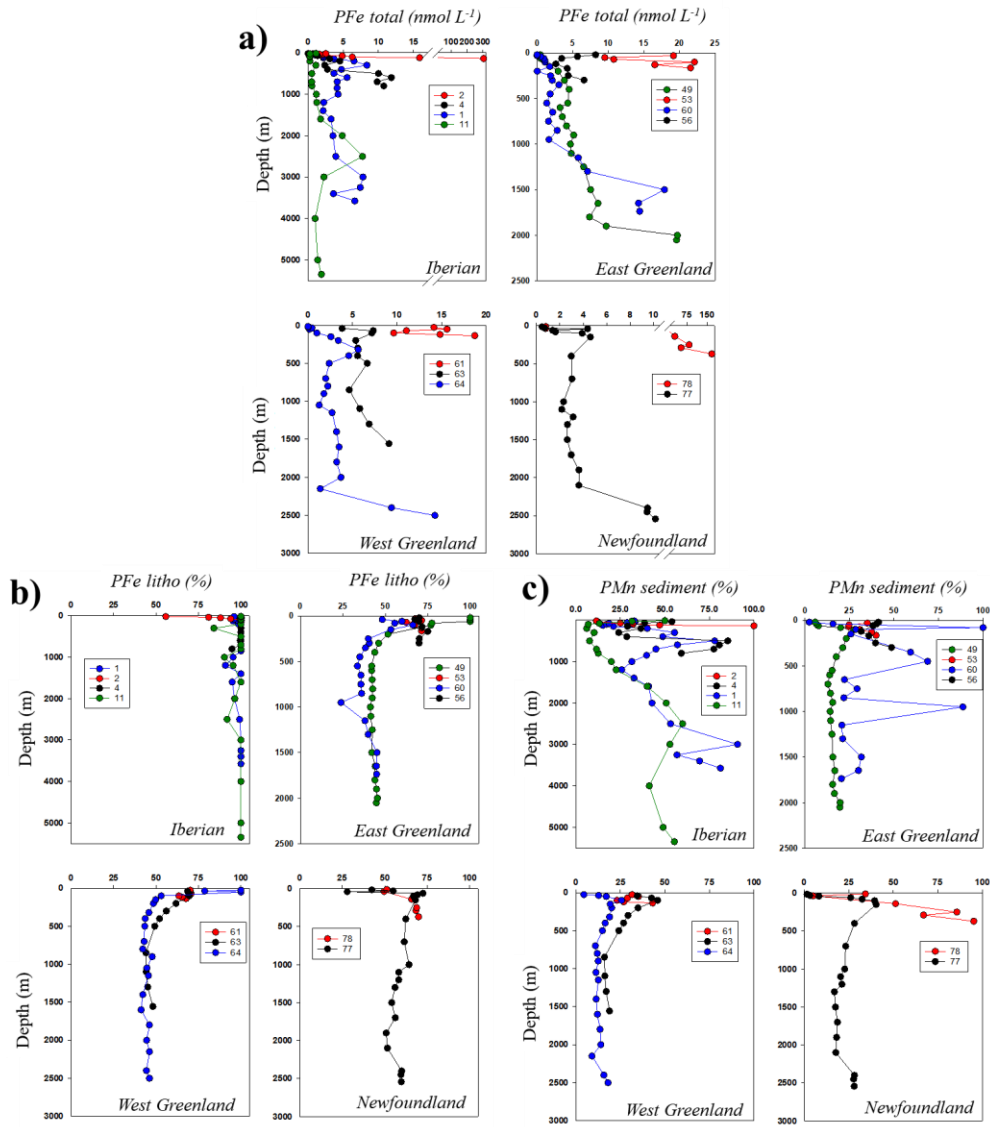
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1343 | Figure 109: Section of derived contribution of sedimentary inputs manganese bulk sediment proxy (a) and
 1344 transmissometry (b) along the GA01 section. Station IDs and biogeochemical region are indicated above the section
 1345 (a). This figure was generated by Ocean Data View (Schlitzer, R., Ocean Data View, odv.awi.de, 2017).



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1352 | Figure 140: Vertical profiles of PFe (nmol L⁻¹), lithogenic proportion of particulate iron (%), and sedimentary
 1353 proportion of particulate manganese (%) at the Iberian, East-West Greenland and Newfoundland margins.



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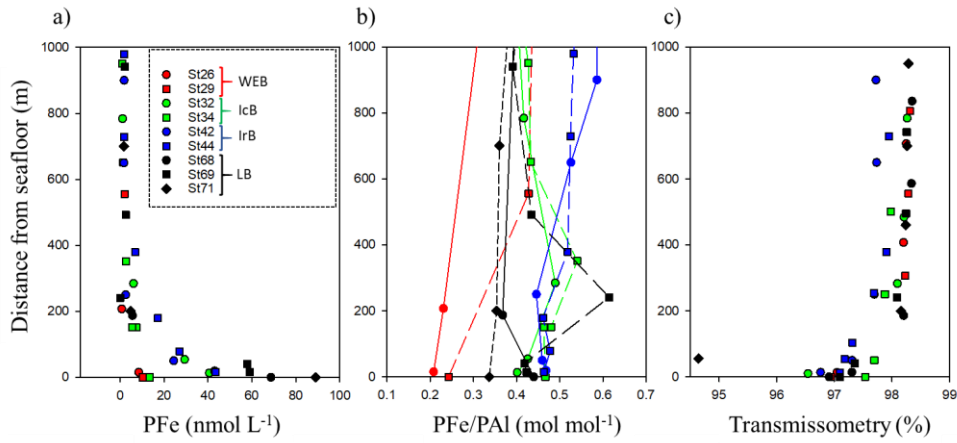
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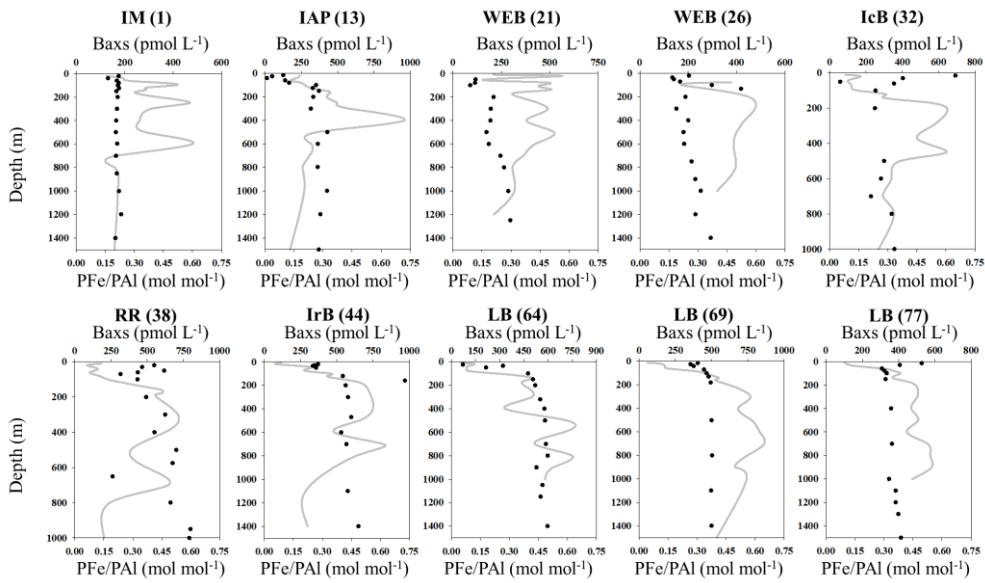
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1361 | Figure 112: PFe total (a); PFe/PAI ratio (b) and beam transmissometry (%) as a function of depth above the seafloor
 1362 (m) at selected stations where a decrease in transmissometry was recorded.



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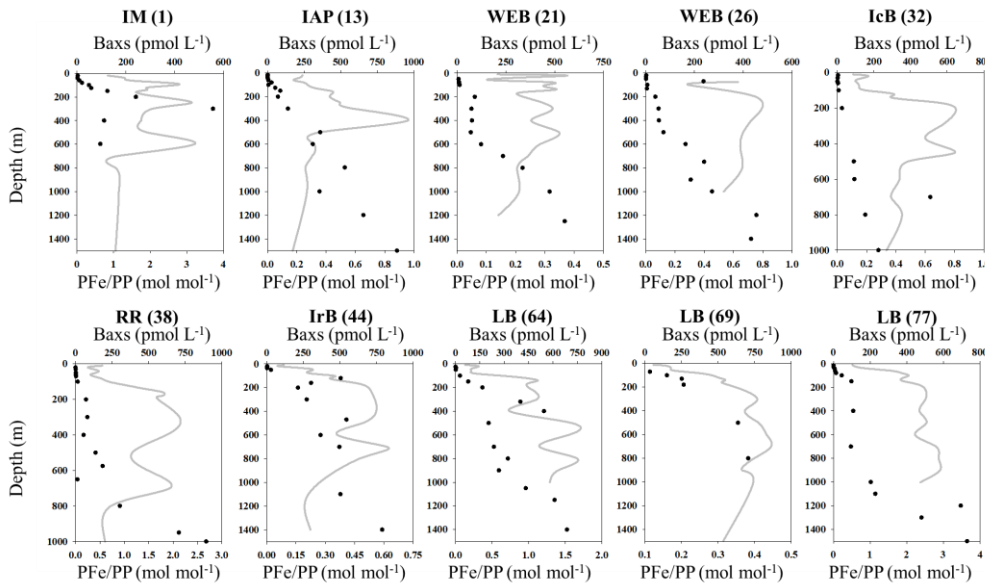
1377 | **Figure 13: Vertical profiles of B_{act} (grey line, data from Lemaitre et al., 2018a) superimposed with PFe/PAI molar**
 1378 **ratios (black dots) at stations sampled in the Iberian Margin (IM), Iberian Abyssal Plain (IAP), Western European**
 1379 **Basin (WEB), Iceland Basin (IcB), above the Reykjanes Ridge (RR), Irminger Basin (IrB), and Labrador Basin (LB).**
 1380 **Note that B_{act} concentrations over the background level of 180 pmol L^{-1} are indicative of remineralisation processes**
 1381 **(Lemaitre et al., 2018a).**



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Figure 14: Vertical profiles of Baxs (grey line, data from Lemaitre et al., 2018a) superimposed with PFe_{bio}/PP molar ratios (black dots) at stations sampled in the Iberian Margin (IM), Iberian Abyssal Plain (IAP), Western European Basin (WEB), Iceland Basin (IcB), above the Reykjanes Ridge (RR), Irminger Basin (IrB), and Labrador Basin (LB). Note that $Baxs$ concentrations over the background level of 180 pmol L^{-1} are indicative of remineralisation processes (Lemaitre et al., 2018a).



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| | | Fe | Al | P | Mn |
|--|----------------|---------|---------|---------|----------|
| Blank (nmol L ⁻¹) | 5µm filter | 0.072 | 0.100 | 0.511 | 0.003 |
| | 0.45µm filter | 0.132 | 0.164 | 1.454 | 0.005 |
| Limit of detection (nmol L ⁻¹) | 5µm filter | 0.011 | 0.030 | 0.365 | 0.001 |
| | 0.45µm filter | 0.026 | 0.046 | 1.190 | 0.001 |
| Recovery CRM (%) | BCR-414 (n=10) | 88 ± 7 | | | 94 ± 7 |
| | MESS-4 (n=5) | 98 ± 14 | 97 ± 14 | 80 ± 30 | 110 ± 18 |
| | PACS-3 (n=8) | 101 ± 9 | 99 ± 14 | 91 ± 34 | 112 ± 11 |

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| % recovery | Al | P | Mn | Fe |
|-----------------------|---------|---------|----------|---------|
| BCR-414 (n=10) | - | - | 94 ± 7 | 88 ± 7 |
| MESS-4 (n=5) | 97 ± 14 | 80 ± 30 | 110 ± 18 | 98 ± 14 |
| PACS-3 (n=8) | 99 ± 14 | 91 ± 34 | 112 ± 11 | 101 ± 9 |

Table 1: Blank and limit of detection (nmol L⁻¹) of the two filters and Certified reference material (CRM) recoveries during GEOVIDE suspended particle digestion.

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

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

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1 **Inputs and processes affecting the distribution of**
2 **particulate iron in the North Atlantic along the GEOVIDE**
3 **(GEOTRACES GA01) section**

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5
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19
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21
22 **Abstract**

23 The GEOVIDE cruise (May-June 2014, R/V Pourquoi Pas?) aimed to provide a better understanding on trace
24 metal biogeochemical cycles in the North Atlantic. As particles play a key role in the global biogeochemical
25 cycle of trace elements in the ocean, we discuss the distribution of particulate iron (PFe), in light of particulate
26 aluminium (PAI), manganese (PMn) and phosphorus (PP) distributions. Overall, 32 full vertical profiles were
27 collected for trace metal analyses, representing more than 500 samples. This resolution provides a solid basis for
28 assessing concentration distributions, elemental ratios, size-fractionation, or adsorptive scavenging processes in
29 key areas of the thermohaline circulation. Total particulate iron (PFe) concentrations ranged from as low as 9
30 pmol L⁻¹ in surface Labrador Sea waters to 304 nmol L⁻¹ near the Iberian margin, while median PFe
31 concentrations of 1.15 nmol L⁻¹ were measured over the sub-euphotic ocean interior.

32 Within the Iberian Abyssal Plain, ratio of PFe over particulate aluminium (PAI) is identical to the continental
33 crust ratio (0.21), indicating the important influence of crustal particles in the water column.~~At most stations~~
34 ~~over the Western, the relative concentrations of total PFe and aluminium (PAI) showed the near-ubiquitous~~
35 ~~influence of crustal particles in the water column.~~ Overall, the lithogenic component explained more than 87%
36 of PFe variance along the section. Within the Irminger and Labrador basins, the formation of biogenic particles
37 led to an increase of the PFe/PAI ratio (up to 0.7 mol mol⁻¹) compared to the continental crust ratio (0.21 mol

38 | mol⁻¹). Margins provide important quantities of particulate trace elements (up to 10 nmol L⁻¹ of PFe) to the open
39 ocean, and in the case of the Iberian margin, advection of PFe was visible more than 250km away from the
40 margin. Additionally, several benthic nepheloid layers spreading over 200m above the seafloor were
41 encountered along the transect, especially in the Icelandic, Irminger and Labrador basins, delivering particles
42 with high PFe content, up to 89 nmol L⁻¹ of PFe. Finally, remineralisation processes are also discussed, and
43 showed different patterns among basins and elements.

44

45 1. Introduction

46 Particles play a key role in the ocean where they drive the residence time of most elements (Jeandel et al., 2015),
47 and strongly influence the global biogeochemistry of macro and micro-nutrients including iron (Milne et al.,
48 2017). In the surface ocean, biological activity produces biogenic suspended matter through planktonic
49 organisms, while atmospheric deposition (Baker et al., 2013; Jickells et al., 2005), riverine discharge (Aguilar-
50 Islas et al., 2013; Berger et al., 2008; Ussher et al., 2004) or ice-melting (Hawkings et al., 2014; Lannuzel et al.,
51 2011, 2014) bring mostly lithogenic derived particles to surface waters. These particulate inputs highly vary,
52 both spatially and seasonally, around the world's oceans. At depth, benthic and shelf sediment resuspension
53 (e.g. Aguilar-Islas et al., 2013; Cullen et al., 2009; Elrod et al., 2004; Fitzwater et al., 2000; Hwang et al., 2010;
54 Lam et al., 2015; Lam and Bishop, 2008; McCave and Hall, 2002), and hydrothermal activity (Elderfield and
55 Schultz, 1996; Lam et al., 2012; Tagliabue et al., 2010, 2017; Trefry et al., 1985), provides important amounts
56 of particles to the water column. Moreover, authigenic particles can be produced *in-situ* by aggregation of
57 colloids (Bergquist et al., 2007) or oxidation processes (Bishop and Fleisher, 1987; Collier and Edmond, 1984).
58 Thus, oceanic particles result from a complex combination of these different sources and processes (Lam et al.,
59 2015).

60 Particles represent the main part of the total iron pool in the upper water column (Radic et al., 2011), and
61 strongly interact with the dissolved pool (e.g. Ellwood et al., 2014). Indeed, dissolved iron can be scavenged
62 onto particles (Gerringa et al., 2015; Rijkenberg et al., 2014), incorporated into biogenic particles (Berger et al.,
63 2008) or [produced by remineralisation of particles](#) (Dehairs et al., 2008; Sarthou et al., 2008). Interestingly,
64 the concept of “reversible scavenging” (i.e. release at depth of dissolved iron previously scavenged onto
65 particles) has been advocated recently (Dutay et al., 2015; Jeandel and Oelkers, 2015; Labatut et al., 2014),
66 while other studies reveal distinct dissolution processes (e.g. Oelkers et al., 2012; Cheize et al., submitted to
67 Chemical Geology). Slow dissolution of particulate iron at margins has also been evoked as a continuous
68 fertilizer of primary production and should be considered as a source of dissolved iron (e.g. Jeandel et al., 2011;
69 Jeandel and Oelkers, 2015; Lam and Bishop, 2008). Within or below the mixed layer, the rates of regeneration
70 processes can also impact the bioavailable pool of iron, among other trace metals (e.g. Ellwood et al., 2014;
71 Nuester et al., 2014). However, the rates of these processes are not yet fully constrained. The study of particulate
72 iron is thus essential to better constrain the global biogeochemical cycle of iron in the ocean. This subject
73 received a growing interest over the last 10 years in particular (e.g. Bishop and Biscaye, 1982; Collier and
74 Edmond, 1984; Frew et al., 2006; Lam et al., 2012; Milne et al., 2017; Planquette et al., 2011, 2013; Sherrell et
75 al., 1998) and, to our knowledge, only two have been performed at an ocean-wide scale and published so far: the
76 GA03 GEOTRACES North Atlantic Zonal Transect (Lam et al., 2015; Ohnemus and Lam, 2015) and the GP16
77 GEOTRACES Pacific Transect (Lam et al., 2017; Lee et al., 2017).

78 In this context, this paper presents the particulate iron distribution in the North Atlantic Ocean, along the
79 GEOTRACES GA01 section (GEOVIDE), and discusses the various sources and processes affecting its
80 distribution, using ~~the distribution of other trace elements, more particularly~~ particulate aluminium, phosphorus
81 or manganese, ~~to further our understanding of this important pool of iron.~~
82

83 2. Methods

84 2.1. Study area

85 Particulate samples were collected at 32 stations during the GEOVIDE (GEOTRACES GA01 section) ~~campaign~~
86 ~~cruise~~ between May and June 2014 aboard the R/V *Pourquoi Pas?* in the North Atlantic. The sampling spanned
87 several biogeochemical provinces (Figure 1): ~~that first comprised~~ the Iberian margin (IM, Stations 2, 1 and 4),
88 the Iberian Abyssal Plain (IAP, Stations 11 to 17), the Western European Basin (WEB, Station 19 to Station 29)
89 ~~and~~ the Icelandic Basin (IcB, Stations 32 to 36). ~~Then, samples were collected,~~ above the Reykjanes Ridge (RR,
90 Station 38), ~~in~~ the Irminger Basin (IrB, Stations 40 to 60), ~~close to~~ the Greenland shelf (GS, Stations 53 and 61),
91 the Labrador Basin (LB, Stations 63 to 77) and finally ~~close~~ the Newfoundland shelf (NS, Station 78) (Figure
92 1).

93 The North Atlantic is characterized by a complex circulation (briefly described in section 2.1 and in detail by
94 Zunino et al. (2017) and García-Ibáñez et al. (2015) and is one of the most productive regions of the global
95 ocean (Martin et al., 1993; Sanders et al., 2014), with a complex phytoplankton community structure composed
96 of diverse taxa (Tonnard et al., in prep.).
97

98 2.2. Sampling

99
100 Samples were collected using the French GEOTRACES clean rosette, equipped with twenty-two 12L GO-FLO
101 bottles (two bottles were leaking and were never deployed during the cruise). GO-FLO bottles (General
102 Oceanics) were initially cleaned in the home laboratory (LEMAR) following the GEOTRACES procedures
103 (Cutter and Bruland, 2012). The rosette was deployed on a ~~146~~mm Kevlar cable with a dedicated, custom-
104 designed clean winch. Immediately after recovery, the GO-FLO bottles were individually covered at each end
105 with plastic bags to minimize contamination. They were then transferred into a clean container (class-100) for
106 sampling, and the filters processed under a laminar flow unit. On each cast, nutrient and/or salinity samples
107 were taken to check potential leakage of the GO-FLO bottles.

108 ~~Prior to filtration, Filters were cleaned following the GEOTRACES protocols~~
109 ~~(<http://www.geotraces.org/images/Cookbook.pdf>) and kept in acid-cleaned 1 L LDPE bottles (Nalgene) filled~~
110 ~~with ultrapure water (Milli-Q, resistivity of 18.2 MΩ cm) until use. All filters were 25 mm diameter in order to~~
111 ~~optimize signal over the filter blank except at the surface depth where 47 mm diameter filters mounted on acid-~~
112 ~~cleaned polysulfone filter holders (Nalgene™) were used. Prior to filtration,~~ the GO-FLO bottles were shaken
113 three times, as recommended in the GEOTRACES cookbook to avoid settling of particles in the lower part of
114 the bottle. GO-FLO bottles were pressurized to <8 psi with 0.2 µm filtered dinitrogen (N₂, Air Liquide).
115 Seawater was then filtered directly through paired filters (Pall Gelman Supor™ 0.45 µm polyetersulfone, and
116 Millipore mixed ester cellulose MF 5 µm) mounted in Swinnex polypropylene filter holders (Millipore),

117 following Planquette and Sherrell (2012) inside the clean container. Filtration was operated until the bottle was
118 empty or until the filter clogged; volume filtered ranged from 2 liters for surface samples to 11L within the
119 water column. ~~Filters were cleaned following the protocol described in Planquette and Sherrell (2012) and kept~~
120 ~~in acid-cleaned 1 L LDPE bottles (Nalgene) filled with ultrapure water (Milli-Q, resistivity of $18.2 \text{ M}\Omega \cdot \text{cm}^{-1}$)~~
121 ~~until use. All filters were 25 mm diameter in order to optimize signal over the filter blank except at the surface~~
122 ~~depth where 47 mm diameter filters mounted on acid-cleaned polysulfone filter holders (NalgeneTM) were used.~~
123 After filtration, filter holders were disconnected from the GO-FLO bottles and a gentle vacuum was applied
124 using a syringe in order to remove any residual water under a laminar flow hood. Filters were then removed
125 from the filter holders with plastic tweezers that were rinsed with Milli-Q between samples. Most of the
126 remaining seawater was ‘sipped’ by capillary action, when placing the non-sampled side of the filter onto a
127 clean 47 mm supor filter. Then, each filter pair was placed in an acid-cleaned polystyrene Petri~~Slides-slide~~
128 (Millipore), double bagged, and finally stored at -20°C until analysis at LEMAR. Between casts, filter holders
129 were thoroughly rinsed with Milli-Q, placed in an acid bath (5% HCl) for 24 hours, then rinsed with Milli-Q.
130 At each station, process blanks were collected as follows: 2L of a deep (1000 m) and a shallow (40 m) seawater
131 samples were first filtered through a $0.2 \mu\text{m}$ pore size capsule filter (Pall Gelman Acropak 200) mounted on the
132 outlet of the GO-FLO bottle before to pass through the particle sampling filter, which was attached directly to
133 the swinnex filter holder.

134

135 2.3. Analytical methods

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

141 Filters were digested following the method described in Planquette and Sherrell (2012). Filter were placed on
142 the inner wall of acid-clean 15mL PFA vials (SavillexTM), and 2 mL of a solution containing 2.9 mol L^{-1}
143 hydrofluoric acid (HF, suprapur grade, Merck) and 8 mol L^{-1} nitric acid (HNO_3 , Ultrapur grade, Merck) was
144 added to each vial. Vials were then closed and refluxed at 130°C on a hot plate for 4 hours. After cooling, the
145 digest solution was evaporated at 110°C until near dryness. Then, $400 \mu\text{L}$ of concentrated HNO_3 (Ultrapur
146 grade, Merck) was added, and the solution was re-evaporated at 110°C . Finally, the obtained residue was
147 dissolved with 3mL of a 0.8 mol L^{-1} HNO_3 (Ultrapure grade, Merck). This archive solution was transferred to an
148 acid cleaned 15 mL polypropylene centrifuge tube (Corning®) and stored at 4°C until analyses.

149 All analyses were performed on a sector field inductively coupled plasma mass spectrometer (SF-ICP-MS
150 Element2, Thermo-Fisher Scientific). Samples were diluted by a factor of 7 on the day of analysis in acid-
151 washed 13 mm (outer diameter) rounded bottom, polypropylene centrifuge tubes (VWR) with 0.8 mol L^{-1} HNO_3
152 (Ultrapur grade, Merck) spiked with $1\mu\text{g L}^{-1}$ of Indium (^{115}In) solution in order to monitor the instrument drift.
153 Samples were introduced with a PFA-ST nebulizer connected to a quartz cyclonic spray chamber (Elemental
154 Scientific Incorporated, Omaha, NE) via a modified SC-Fast introduction system consisting of an SC-2
155 autosampler, a six-port valve and a vacuum-rinsing pump. The autosampler was contained under a HEPA
156 filtered unit (Elemental Scientific). Two 6-points, matrix-matched multi-element standard curves with

157 concentrations bracketing the range of the samples were run at the beginning, the middle and the end of each
158 analytical run. Analytical replicates were made every 10 samples, while accuracy was determined by performing
159 digestions of the certified reference material BCR-414 (plankton, Community Bureau of Reference,
160 Commission of the European Communities), PACS-3 and MESS-4 (marine sediments, National Research
161 Council Canada), following the same protocol as for samples. Recoveries were typically within 10% of the
162 certified values (and within the error of the data, taken from replicate measurements, Table 1). Once all data
163 were normalized to an ¹¹⁵In internal standard and quantified using an external standard curve, the dilution factor
164 of the total digestion was accounted for. Obtained element concentrations per filter (pmol/filter) were then
165 corrected by the process blanks described above. Finally, pmol/filter values were divided by the volume of
166 water filtered through stacked filters.
167 Total concentrations (sum of small size fraction (0.45-5 µm) and large (>5 µm) size fraction) of particulate trace
168 elements are reported in Table S1 (supplementary data).

169
170

171 2.4. Ancillary data:

172 ~~Particulate barium (Ba) concentrations were determined in samples collected using a standard CTD rosette~~
173 ~~equipped with 12 L Niskin bottles. Typically, 18 samples were collected at each station within the first 1000 m.~~
174 ~~Details on analytical procedures are given in Lemaitre et al. (in press, 2018a). Briefly, particulate biogenic~~
175 ~~Barium, or excess Barium (Ba_{ex}), were calculated by subtracting the particulate lithogenic barium (PBa_{litho})~~
176 ~~from the total particulate barium (PBa). The PBa_{litho} was determined by multiplying the particulate aluminium~~
177 ~~(PAI) concentration by the upper continental crust (UCC) Ba: Al molar ratio (0.00135 mol mol⁻¹; Taylor and~~
178 ~~Melelland, 1985). Potential temperature (θ), salinity (S), and transmissometry data were retrieved from the CTD~~
179 ~~sensors (CTD SBE911 equipped with a SBE43).~~

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181

181 2.5. Positive matrix factorisation

182 Positive Matrix Factorisation (PMF) was run to characterise the main factors influencing the particulate trace
183 elements variances along the GEOVIDE section. In addition to PFe, PAI, PMn, and PP, nine additionnal
184 elements were included in the PMF: Y, Ba, Pb, Th, Ti, V, Co, Cu and Zn. The analysis has been conducted on
185 samples where all the 13 elements previously cited were above the detection limits; after selection, 445 of the
186 549 existing data points were used. Analyses were performed using the PMF software, EPA PMF 5.0,
187 developed by the USA Environmental Protection Agency (EPA). Models have been tested with several factors
188 number (from 3 to 6), after full error estimation of each model, we decide to use the configuration providing the
189 lowest errors estimations and in consequence the most reliable.
190 In consequence, models were set up with four factors and were run 100 times to observe the stability of the
191 obtained results. After displacement, error estimations and bootstraps error estimations, the model was
192 recognised as stable.

193
194

194 **3. Results**

195

3.1. Hydrography and biological setting

196 Here, we briefly describe the hydrography encountered during the GEOVIDE section (Figure 2), as a thorough
197 description is available in García-Ibáñez et al. (2015). The warm and salty Mediterranean Water (MW, $S=36.50$,
198 $\theta^{\circ}=11.7^{\circ}\text{C}$) was sampled between 600 and 1700 m in the Iberian Abyssal Plain (IAP). MW resulted from the
199 mixing between the Mediterranean Overflow Water plume coming from the Mediterranean Sea and local
200 waters. Surface water above the Iberian Shelf was characterised by low salinity ($S=34.95$) at station 2 and 4
201 compared to surrounding water masses. Close to the floor of the Iberian Abyssal Basin, the North East Atlantic
202 Deep Water (NEADW, $S=34.89$, $\theta^{\circ}=2.0^{\circ}\text{C}$) spread southward. The ~~East~~ North Atlantic Central Water
203 (~~E~~NACW, $S>35.60$, $\theta^{\circ}>12.3^{\circ}\text{C}$) was the warmest water mass of the transect and was observed in the subsurface
204 layer of the Western European Basin and Iberian Abyssal Plain. An old Labrador Sea Water (LSW, $S=34.87$,
205 $\theta^{\circ}=3.0^{\circ}\text{C}$) flowed inside the Western European and Icelandic Basins, between 1000 and 2500m depth. In the
206 Icelandic Basin, below the old LSW, the Iceland-Scotland Overflow Water (ISOW, $S=34.98$, $\theta^{\circ}=2.6^{\circ}\text{C}$) spread
207 along the Reykjanes Ridge slope. This cold water, originating from the Arctic, led to the formation of NEADW
208 after mixing with surrounding waters. North Atlantic hydrography was impacted by the northward flowing of
209 the North Atlantic Current (NAC), which carried up warm and salty waters from the subtropical area. When
210 NAC crossed the Mid-Atlantic ridge through the Charlie-Gibbs Fracture Zone (CGFZ), it created the Subpolar
211 Mode Water (SPMW). The recirculation of SPMW inside the Icelandic and Irminger Basins led to the formation
212 of regional modal waters: the Iceland Subpolar Mode Water (IcSPMW, $S=35.2$, $\theta^{\circ}=8.0^{\circ}\text{C}$) and the Irminger
213 Subpolar Mode Water (IrSPMW, $S=35.01$, $\theta^{\circ}=5.0^{\circ}\text{C}$) respectively. IcSPMW was a relatively warm water mass
214 with potential temperature up to 7°C (García-Ibáñez et al., 2015). Another branch of the NAC mixed with
215 Labrador Current waters to form the relatively fresh SubArctic Intermediate Water (SAIW, $S<34.8$,
216 $4.5^{\circ}\text{C}<\theta^{\circ}<6^{\circ}\text{C}$). The Irminger Basin is a ~~really~~ complex area with a multitude of water masses. In the middle of
217 the basin, an old LSW, formed one year before (Straneo et al., 2003), spread between 500 and 1200 m depth.
218 Close to the bottom, the Denmark Strait Overflow Water (DSOW, $S=34.91$) flowed across the basin. Greenland
219 coastal waters were characterised by low salinity values, down to $S=33$. The strong East Greenland Current
220 (EGC) flowed southward along the Greenland shelf in the Irminger Basin. When reaching the southern tip of
221 Greenland, this current entered the Labrador Basin along the west coast of Greenland and followed the outskirts
222 of the basin until the Newfoundland shelf. In the Labrador Basin, the deep convection of SPMW at 2000 m was
223 involved in the formation of the LSW ($S=34.9$, $\theta^{\circ}\text{C}=3.0^{\circ}\text{C}$) (García-Ibáñez et al., 2015; Yashayaev and Loder,
224 2009). Above the Newfoundland Shelf, surface waters were affected by discharge from rivers and ice-melting
225 and characterised by extreme low salinity for open ocean waters, below 32 in the first 15 meters.

226

~~227 During GEOVIDE, diatoms and type 6 haptophytes dominated the bloom close to the IM, while type 6~~
~~228 haptophytes and dinophytes were dominant in the WEB province (Tonnard et al., in prep.). The IB bloom was~~
~~229 dominated by type 6 haptophytes and the IrB was dominated by diatoms. GS and NS coastal stations were~~
~~230 almost exclusively composed of large diatoms. Finally, the LB was dominated by diatoms and type 6 and 8~~
~~231 haptophytes. The NS, LB, GS and IrB provinces (stations 44 to 77) were sampled just after the bloom peak. The~~
~~232 LB was characterized by an intense particulate organic carbon (POC) export and high remineralization activity~~

233 ~~(Lemaitre et al., 2018a). In contrast, low remineralization fluxes and high POC exports were determined within~~
234 ~~the IB and WEB provinces, where the bloom was still active (Lemaitre et al., 2018a, b).~~

235 3.2. Section overview

236 Total particulate iron (PFe), aluminium (PAI), manganese (PMn) and phosphorus (PP) concentrations spanned a
237 large range of concentrations from below detection to 304, 1544, ~~21.5~~, 3.5 and 402 nmol L⁻¹ respectively.

238 PFe, PAI, and PMn were predominantly found (>90%) in particles larger than 5 µm, except in surface waters,
239 where ~~209% ± 8.6 %~~ of PFe, ~~38.8 ± 8.6 %~~ of PP, ~~3510.9 ± 15.4 %~~ of PAI and ~~32.8 ± 16.6 % up to 60%~~ of PMn
240 were hosted by smaller particles (0.45-5 µm). The ranges of concentrations are comparable to other studies
241 recently published (Table 2). Data are shown in Figure 3.

242

243 3.3. Open Ocean stations :Iberian Abyssal Plain (stations 11 to 17), Western European Basin 244 (stations 19 to 29), Icelandic Basin (stations 32 to 36), Reykjanes Ridge (station 38), Irminger Basin 245 (stations 40 to 60; except Stations 53 and 56) and Labrador Basin (stations 63 to 77)

246 ~~In the open ocean, p~~Particulate iron concentration vertical profiles presented identical patterns at open ocean
247 ~~stations sampled in every each~~ oceanic basins encountered along the ~~GEOVIDE~~ section. Median PFe were low
248 at 0.25 nmol L⁻¹ (~~n=???~~) within the first 100 m and steadily increased with depth. However, at two stations,
249 elevated concentrations were determined in the upper 100m, up to 4.4 nmol L⁻¹ at station 77 at 40 m and 7 nmol
250 L⁻¹ at station 63 between 70 and 100 m depth. PFe concentrations ~~were increasing gradually~~ increased with
251 depth, with a median PFe of 1.74 nmol L⁻¹ below 1000m (~~n=???~~), ~~to reach a maximum close to the bottom.~~
252 ~~Close to the seafloor of some stations (26, 29, 32, 34, 49, 60, and 71), high concentrations of PFe were~~
253 ~~observed, up to 88 nmol L⁻¹ (station 71 at 3736 m). These high PFe values were associated with low beam~~
254 ~~transmissometry value inferior or equal to 97 % (Figure 9b and supplementary table S2).~~

255 ~~Particulate aluminium (PAI) and manganese (PMn) profiles were similar to PFe profiles, with low median~~
256 ~~value~~ concentrations were ~~observ~~ measured in the first 100 m (1.88 nmol L⁻¹ (~~n=??~~) and 55 pmol L⁻¹ (~~n=??~~),
257 ~~respectively) and increased towards the seafloor. Close to the seafloor, high concentrations were determined at~~
258 ~~the same stations cited above for PFe, with a~~ ~~where high value were observed at certain stations (same than~~
259 ~~PFe) with~~ maximum of 264 nmol L⁻¹ and 3.5 nmol L⁻¹ for PAI and PMn, respectively at station 71 ~~;~~
260 ~~(supplementary Table S1). Highest particulate phosphorus (PP) concentrations were maximum observed in the~~
261 ~~first 50m, with a median value of 66 nmol L⁻¹ (~~n=???~~). Deeper in the water column, below 200m, PP~~
262 ~~concentrations decreased to values~~ ~~under~~ below 10 nmol L⁻¹ ~~below 200 meters depth. Inter-basins differences~~
263 ~~were observed within the surface samples, with Irminger basin had the higher median PP concentration being~~
264 ~~higher in the Irminger Basin (127 nmol L⁻¹), ~~n=???~~ of the section while the than in the Iberian Abyssal Plain~~
265 ~~had the lower median (28 nmol L⁻¹), ~~n=???~~).~~

266 ~~Finally, above the Reykjanes Ridge, PP, PMn, PAI and PFe concentrations were in the same range than the~~
267 ~~surrounding open ocean stations. However, close to the seafloor, high concentrations were measured, with PFe,~~
268 ~~PAI, and PMn reaching 16.2 nmol L⁻¹, 28.8 nmol L⁻¹, and 0.51 nmol L⁻¹ at 1354 m, respectively (Figure 3 and~~
269 ~~supplementary material Table S1 Table 1, supplementary material and Figure xxx).~~

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271 3.4. Margins and Shelves: Iberian Margin (stations 1 to 4), Greenland coast (stations 53, 56
272 and 61) and Newfoundland Shelf (station 78)

273
274 The Iberian margin was characterised by low beam transmissometry values at station 2 (88% at 140 m, Figure
275 9b) suggesting significant particle concentrations. Particulate iron concentrations varied between 0.02 nmol L⁻¹,
276 station 2 to 304 nmol L⁻¹, Station 2. In Within the first 50m, PFe concentrations decreased from the Iberian
277 Shelf (Station 2, 2.53 nmol L⁻¹) towards the shelf break where PFe dropped down from 2.53 nmol L⁻¹ to
278 (station 2, depth) to 0.8 nmol L⁻¹ (Station 1). PFe concentrations increased with depth at all three stations and
279 reached a maximum at the bottom of station 2 (138.5 m) with more than 300 nmol L⁻¹ of PFe. Lithogenic
280 tracers, such as PAI or PMn, presented similar profiles to PFe with concentrations ranging between 0.11 and
281 1544 nmol L⁻¹, and from below detection limit to 2.51 nmol L⁻¹ respectively (station, depth) (Figure 3, Table 1,
282 supplementary material Table S1, Figure xxxxx). Total particulate phosphorus (PP) concentrations were
283 relatively low in surface ranging from undetectable values to 38 nmol L⁻¹ (station xxxxx) (Table 1, supplementary material, Figure
284 xxxxx) in surface; then Maximum PP was measured in surface at Station 1 (20 m depth), then concentrations
285 decreased with depth and were less than 0.7 nmol L⁻¹ below 1000 m depth.
286 Particulate Fe concentrations in In the vicinity of the Greenland shelf, PFe concentrations had a high median
287 concentration value of 10.8 nmol L⁻¹ (n=???) while and were associated with high median PAI and PMn also had
288 high median concentrations of 32.3 nmol L⁻¹ (n=???) and 0.44 nmol L⁻¹ (n=???) , respectively. Concentrations of
289 PP were high at the surface with a value of 197 nmol L⁻¹ at 25 m of station 61. Then, PP concentrations
290 decreased strongly, less than 30 nmol L⁻¹, below 100 meters depth. Furthermore, beam transmissometry values
291 in surface waters at these three stations, were the lowest of the entire section, with values below 85 %.
292 Close to the Newfoundland margin, surface waters displayed a small load of particulate trace metals as PFe,
293 PAI, and PMn were below 0.8 nmol L⁻¹, 2 nmol L⁻¹, and 0.15 nmol L⁻¹ respectively. Then close to the bottom of
294 station 78, at 371 m, beam transmissometry values dropped to 94% and were associated with extremely high
295 concentrations of PFe=168 nmol L⁻¹, PAI=559 nmol L⁻¹, and PMn=2 nmol L⁻¹. Total PP concentrations in the
296 first 50 m ranged from 35 to 97 nmol L⁻¹. Below the surface, PP remained relatively high with values up to 16
297 nmol L⁻¹ throughout the water column. (Table 1, supplementary material and Figure XXXX3 and supplementary
298 material Table S1).

299 3.5. Reykjanes Ridge (station 38)

300 Above the Reykjanes Ridge, the upper portion of the Mid Atlantic Ridge, particulate trace
301 elements concentrations were in the same range than the surrounding open ocean stations. However,
302 close to the seafloor, high concentrations were measured on the ridge, with PFe, PAI, and PMn
303 reaching 16.2 nmol L⁻¹, 28.8 nmol L⁻¹, and 0.51 nmol L⁻¹ at 1354 m, respectively. Low concentrations
304 of PP were measured in surface waters, with a median value of 24.8 nmol L⁻¹ in the top 100 m and a
305 maximum of only 72.6 nmol L⁻¹ at 20 m. 3.3. Iberian Margin (stations 1 to 4)

306 The Iberian margin was characterised by low beam transmissometry values at station 2 (88% at 140 m)
307 suggesting significant particle concentrations. Particulate iron concentrations varied between 0.02 nmol L⁻¹ (20
308 m) to 304 nmol L⁻¹ (138 m) in this area. Above the Iberian Shelf, high PFe concentrations were measured in

309 surface (Station 2, 2.53 nmol L⁻¹); then, on the shelf break, surface concentrations dropped down to 0.8 nmol L⁻¹
310 (Station 1 at 20 m depth). PFe concentrations increased with depth at all three stations and reached a maximum
311 at the bottom of station 2 (138.5 m) with more than 300 nmol L⁻¹ of PFe. Lithogenic tracers, such as PAI or
312 PMn, presented similar profiles to PFe with concentrations ranging between 0.11 and 1544 nmol L⁻¹, and from
313 below detection to 2.51 nmol L⁻¹ respectively. The highest concentrations were also measured at the bottom of
314 station 2 (138.5 m). Total particulate phosphorus (PP) concentrations were relatively low in this area ranging
315 from undetectable values to 38 nmol L⁻¹. Maximum PP was measured in surface at Station 1 (20 m depth), then
316 concentrations decreased with depth and were less than 0.7 nmol L⁻¹ below 1000 m depth.

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3.4. Iberian Abyssal Plain (stations 11 to 17) and Western European Basin (stations 19 to 29)

320 In the Iberian Abyssal Plain (IAP) and the Western European Basin (WEB), particulate iron concentration
321 vertical profiles were similar (Figure 4); median PFe concentrations were 0.18 nmol L⁻¹ in the first 100 m and
322 steadily increased with depth. Close to the seafloor, concentrations of PFe were up to 1.4 nmol L⁻¹ at every
323 station and reached values superior to 8 nmol L⁻¹ at stations 26 and 29, with low beam transmissometry
324 (<98%). Particulate aluminium profiles matched the PFe profiles, with low median concentrations within the
325 first 100m of 1.77 nmol L⁻¹ and 26 pmol L⁻¹ respectively. Then, concentrations increased with depth to reach a
326 maximum close to the oceanic floor. At stations 26 and 29, total PAI concentrations reached high values, up to
327 42 nmol L⁻¹. In the Western European Basin, PMn concentrations ranged from below detection to 0.36 nmol L⁻¹;
328 except close to the bottom of stations 26 and 29, where high concentrations of 0.91 and 1.31 nmol L⁻¹ were
329 measured, respectively. Particulate phosphorus profiles, while similar between stations of this basin, differed a
330 lot from the other element profiles. In the WEB, surface median PP concentration was two times higher than in
331 the Iberian margin (60 nmol L⁻¹ against 28 nmol L⁻¹ in the first 50 m with a maximum of 162 nmol L⁻¹ (station
332 21). Concentrations dropped drastically with depth and remained under 10 pmol L⁻¹ below 100 m.

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3.5. Icelandic Basin (stations 32 to 36)

336 Concentrations of PFe were in a similar range and displayed analogous profiles to the ones collected in the
337 Western European Basin (figure 4), from below detection to 40.6 nmol L⁻¹; with low values at the surface (<1
338 nmol L⁻¹) and a progressive increase with depth. Close to the basin seafloor, low beam transmissometry (97.4%)
339 measurements were associated with high PFe concentrations of 40.6 nmol L⁻¹ at 3271 m of station 32.
340 Particulate aluminium vertical profiles were similar to those in the WEB but with extremely low surface
341 concentrations below 0.6 nmol L⁻¹; PAI then increased steadily with depth, reaching values up to 2 nmol L⁻¹
342 below 500 m. As previously observed for PFe, PAI concentrations were higher close to the seafloor, from 29
343 nmol L⁻¹ at station 34 to 101 nmol L⁻¹ at station 32. PMn also presented similar distributions than PFe and PAI.
344 Median surface concentrations were low within the first 100 m, 31 pmol L⁻¹ and 35 pmol L⁻¹, respectively, and
345 increased in the deep ocean to reach a maximum of 2.98 nmol L⁻¹ close to the seafloor. The Icelandic Basin had
346 a typical vertical profile for PP, with high concentrations at the surface, reaching 129 nmol L⁻¹ at station 32 and
347 really low concentrations below 150 m, inferior to 20 nmol L⁻¹.

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3.6. Reykjanes Ridge (station 38)

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Surface concentrations of particulate Fe, Al, and Mn above the Reykjanes Ridge (RR) were similar to the Icelandic Basin (Figure 3). However, close to the seafloor, high concentrations were measured, with PFe, PAI, and PMn reaching 16.2 nmol L^{-1} , 28.8 nmol L^{-1} , and 0.51 nmol L^{-1} at 1354 m, respectively. Low concentrations of PP were measured in surface waters, with a median value of 24.8 nmol L^{-1} in the top 100 m and a maximum of only 72.6 nmol L^{-1} at 20 m.

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3.7. Irminger Basin (stations 40 to 60; except Stations 53 and 56)

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Particulate Iron, Aluminium and Manganese distributions were similar to stations sampled in the WEB, IeB and IAP provinces (Figure 3). Surface concentrations of these elements were lower than 1.1 nmol L^{-1} , 3.4 nmol L^{-1} , and 0.4 nmol L^{-1} , respectively. Then, below 50 m depth, concentrations of PFe, PAI, and PMn increased and reached high values close to the seafloor, especially at stations 42 and 44; reaching up to 40 nmol L^{-1} , 90 nmol L^{-1} , and 1.5 nmol L^{-1} respectively. Close to the Greenland margin, at the bottom of stations 49 and 60, concentrations of particulate trace metals were also elevated with PFe greater than 10 nmol L^{-1} . Particulate phosphorus concentrations were relatively high in surface waters, of the Irminger Basin, with a median value of 127 nmol L^{-1} within the first 50 m. Particulate phosphorus decreased with depth and remained constant below 500 m with concentration below 10 nmol L^{-1} .

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3.8. Greenland coast (stations 53, 56 and 61)

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Particulate Fe concentrations in the vicinity of the Greenland shelf had a high median concentration of 10.8 nmol L^{-1} , while PAI and PMn also had high median concentration of 32.3 nmol L^{-1} and 0.44 nmol L^{-1} , respectively. Concentrations of PP were maximum at the surface with a value of 197 nmol L^{-1} at 25 m of station 61. Then, PP concentrations decreased strongly with depth with values below 30 nmol L^{-1} below 100 meters depth. Furthermore, beam transmissometry values in surface waters at these two stations, were the lowest of the entire section, with values below 85 %.

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3.9. Labrador Basin (stations 63 to 77)

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In the Labrador Basin, median concentrations of PFe within the first 100 m were low, with a median value of 0.9 nmol L^{-1} (n=30). However, at two stations, elevated concentrations were determined, up to 4.4 nmol L^{-1} at station 77 at 40 m and 7 nmol L^{-1} at station 63 between 70 and 100 m depth. Below the surface waters, PFe remained constant with depth until in proximity of the seafloor (Fig. 3). Between stations 64 and 71, the median concentration between 100 m and 200 m above the seafloor was 2.0 nmol L^{-1} (n=39). Particulate Fe concentration at station 63, close to the Greenland margin, remained constant below 100 m depth, with a high median value of 5.7 nmol L^{-1} . On the other side of the Labrador Basin, station 77, close to the Newfoundland margin, constant PFe values of 3 nmol L^{-1} between the surface and 200 m above bottom depth were observed.

384 ~~As previously described, PFe concentration increased close to the seafloor to 88 nmol L⁻¹ at station 71 at 3736~~
385 ~~m. Particulate Al and Mn displayed similar characteristics to PFe, with low median concentrations at the surface~~
386 ~~of 3.37 nmol L⁻¹ and 90 pmol L⁻¹ respectively. Close to the seafloor of Station 71, at 3736 m depth, PAl, and~~
387 ~~PMn reached high concentrations of 264 and 3.5 nmol L⁻¹. Particulate Phosphorus distribution was no different~~
388 ~~than in the eastern basins, with 71 nmol L⁻¹ median PP within the first 50 m. Then below 50 m, the~~
389 ~~concentration dropped off quickly to a median PP of 3 nmol L⁻¹.~~

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391 3.10. Newfoundland Shelf (station 78)

392 ~~Close to the Newfoundland margin, surface waters displayed a small load of particulate trace metals as PFe,~~
393 ~~PAl, and PMn were below 0.8 nmol L⁻¹, 2 nmol L⁻¹, and 0.15 nmol L⁻¹ respectively. Then close to the bottom, at~~
394 ~~371 m, beam transmissometry values dropped to 94% and were associated with extremely high~~
395 ~~concentrations of lithogenic elements: PFe=168 nmol L⁻¹, PAl=559 nmol L⁻¹, and PMn=2 nmol L⁻¹. Total PP~~
396 ~~concentrations in the first 50 m ranged from 35 to 97 nmol L⁻¹. Below the surface, PP remained relatively high~~
397 ~~with values up to 16 nmol L⁻¹ throughout the water column.~~

398

399 **4. Discussion**

400 Our goal in this work was to investigate mechanisms that drive the distribution of PFe in the North Atlantic, in
401 particular the different routes of supply and removal.

402 Possible candidate sources of PFe include lateral ~~mixing advection offshore~~ from ~~the different~~ margins,
403 atmospheric inputs, ~~recently melted sea ice continental run-off~~, melting ice shelves and icebergs, resuspended
404 sediments, ~~or~~ hydrothermal inputs and biological ~~pooluptake~~. Removal processes include remineralization and
405 dissolution processes.

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

409 4.1. Analysis of the principal factors controlling variance: near-ubiquitous influence of 410 crustal particles in the water column

411 ~~The positive matrix factorisation~~ Positive Matrix Factorisation (PMF) was run to characterise the main factors
412 ~~influencing the particulate trace elements variances along the GEOVIDE section. In addition to PFe, PAl, PMn,~~
413 ~~and PP, nine additional elements were included in the PMF: Y, Ba, Pb, Th, Ti, V, Co, Cu and Zn. The analysis~~
414 ~~has been conducted on samples where all the 13 elements previously cited are above the detection limits; after~~
415 ~~selection, 445 of the 549 existing data points were used. Analyses were performed using the PMF software,~~
416 ~~EPA PMF 5.0, developed by the USA Environmental Protection Agency (EPA). Models have been tested with~~
417 ~~several factors number (from 3 to 6), after full error estimation of each model, we decide to use the~~
418 ~~configuration providing the lowest errors estimations and in consequence the most reliable. results~~
419 ~~In consequence, models were set up with four factors and were run 100 times to observe the stability of the~~
420 ~~obtained results. After displacement, error estimations and bootstraps error estimations, the model was~~

421 ~~recognised as stable.~~ Results, ~~are~~ shown in Figure 5, indicate the overall variances explained by each of the 5
422 factors.

423 The first factor is characterised by lithogenic elements, representing 86.8% of the variance of PFe, 75.8% of
424 PAI and 90.5% of PTi. The second factor is correlated with both Mn and Pb and explains no less than 76.5%
425 and 77.0% of their respective variances. Ohnemus and Lam (2015) observed this co-relation between
426 manganese and lead particles and explained it by the co-transport on Mn-oxides (Boyle et al., 2005). The
427 formation of barite is causing the third factor constraining 87.7% of the Ba variance in the studied regions.
428 Biogenic barite accumulation within the mesopelagic layer is related to bacterial activity and organic
429 remineralisation (Lemaitre et al., 2018a). –A biogenic component is the fourth factor and explained most of
430 particulate phosphorus variance, 83.7%. The micronutrient trace metals, copper, cobalt and zinc, had more than
431 a quarter of their variances influenced by this factor.

432
433
434
435 These results indicate that a Along the GA01 section, PFe distributions were predominantly controlled by
436 lithogenic ~~material-material~~ and to a smaller extent by remineralisation processes (as seen by a Factor 3
437 contribution of 4.1%). This does not rule out some biogenic influence on PFe distribution, especially in surface,
438 but this contribution is veiled by the high lithogenic contribution. The PMF analysis has been realised on the
439 entire dataset, in consequence, the factors described are highly influenced by the major variations of particulate
440 element concentrations (usually at the interface, i.e. margin, seafloor, surface layer....).

441 ~~–These inputs and processes are discussed below.~~

442 To further investigate the influence of crustal material on the distribution of PFe, it is instructive to examine the
443 distribution of the molar ratio of PFe/PAI along the section as a way to assess the lithogenic inputs (Lannuzel et
444 al., 2014; Ohnemus and Lam, 2015; Planquette et al., 2009) (Figure 6) along the section.

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445
446 The PFe/PAI ratio can be used to estimate the proportion of lithogenic particles within the bulk particulate
447 material. A comparison with the Upper Continental Crust (UCC) ratio of Taylor and McLennan (1995), 0.21,
448 was used to calculate the lithogenic components of particles (PFe_{litho}) following Eq. (1):

449
450
$$\%PFe_{litho} = 100 * \left(\frac{PAI}{PFe}\right)_{sample} * \left(\frac{PFe}{PAI}\right)_{UCC\ ratio} \quad (1)$$

451
452 Then the non-lithogenic PFe is simply obtained using Eq. (2):

453
454
$$\%PFe_{non_litho} = 100 - \%PFe_{litho} \quad (2)$$

455
456 Both the lithogenic and non-lithogenic fraction of PFe are estimated using ~~in fine~~ the UCC ratio. Spatial and
457 temporal variation of the lithogenic components ratio may falsely influence the estimated fraction value. –The
458 PFe_{litho} and $PFe_{excessnon_litho}$ non-litho

459 ~~proxies are interesting tools to evaluate the importance of lithogenic and authigenic formation of Fe~~
460 ~~oxyhydroxides non-lithogenic (either biogenic or authigenic). Biogenic contributions over the section~~
461 ~~to be used with consideration.~~

462 ~~Overall, the estimated lithogenic contribution to PFe varies from 25% (station 60, 950 m) to 100% at stations~~
463 ~~located within the Western European Basin. 100% of estimated lithogenic PFe doesn't necessary mean that~~
464 ~~biogenic particles are absent; they may just be masked by the important load of lithogenic particles. Important~~
465 ~~inter-basins variations are observed along the section (Figure 6). The IAP and WEB basins are linked with high~~
466 ~~median value of the proxy %Fe_{litho}, 90%. This could be linked to a lateral advection of iron rich lithogenic~~
467 ~~particles sourced from the Iberian margin and to atmospheric particles (Shelley et al., 2017). This point is~~
468 ~~discussed with more detail in section 4.3.1. While the Iceland, Irminger and the Labrador basins are~~
469 ~~characterised with median %PFe value under 55%. An interesting feature observable was the dramatic decrease~~
470 ~~of the %PFe proxy values happening at the station 26 (Figure 6). This feature is likely be associated to the~~
471 ~~presence of the Sub-Arctic Front, located between 49.5 and 51°N latitude and 23.5 and 22°W longitude (Zunino~~
472 ~~et al., 2017). Indeed, this front which separates cold and fresh water of subpolar origin from warm and salty~~
473 ~~water of subtropical origin was clearly identifiable at station 26 by the steep gradient of the isotherms and~~
474 ~~isohalines (Figure 2).~~

475 ~~Overall, the lithogenic contribution to PFe varies from 24% (station 60, 950 m) to 100% at stations located~~
476 ~~within the Western European Basin. This could be linked to a lateral advection of iron rich lithogenic particles~~
477 ~~sourced from the Iberian margin and to a less extent atmospheric particles with the North Atlantic Central Water~~
478 ~~flowing northward (Shelley et al., 2017; Garcia Ibanez et al. (2015). This point is discussed with more detail in~~
479 ~~section 4.3.1.~~

480 ~~The most striking feature is the almost exclusive lithogenic nature of PFe from stations 1 to 26 throughout the~~
481 ~~water column, except between 1000 and 3000 m at stations 21 to 26 (Figure 6 and 7). This feature could be~~
482 ~~linked to the fact that atmospheric inputs generally dominate the supply of PFe—deposited from Saharan dust~~
483 ~~and transported via the Gulf Stream and North Atlantic Current to the WEB (Shelley et al., 2017; Garcia Ibanez~~
484 ~~et al. (2015)), even if low atmospheric fluxes were reported during our cruise.~~

486 ~~This feature at 1000 and 2500 m between stations 21 and 26 is likely be associated to the presence of the Sub-~~
487 ~~Arctic Front, located between 49.5 and 51°N latitude and 23.5 and 22°W longitude (Zunino et al., 2017).~~
488 ~~Indeed, this front which separates cold and fresh water of subpolar origin from warm and salty water of~~
489 ~~subtropical origin was clearly identifiable at station 26 by the steep gradient of the isotherms and isohalines. The~~
490 ~~fact that the WEB was sampled close to but just after the bloom maximum is limiting any higher PFe/PAI~~
491 ~~signatures (see also section 3.3.4). The intrusion of an old LSW (Garcia Ibanez et al., this issue) at stations 21 to~~
492 ~~26 between 1000 and 2500 m with a different PFe/PAI signature could explain the smaller contribution of~~
493 ~~lithogenic PFe in this depth range as atmospheric inputs to the Labrador Sea region are relatively small (Shelley~~
494 ~~et al., 2017). This is discussed with more detail in section 4.2.~~

495 *4.2. Fingerprinting watermasses*

496 | The GEOVIDE section crossed several distinct water masses along the North Atlantic, each of them being
497 | distinguishable by their salinity and potential temperature signatures (García-Ibáñez et al., 2015; Figure 2).
498 | Based on this study, we applied a Kruskal-Wallis test on molar PFe/PAI ratios of nine water masses (Figure 87)
499 | in order to test the presence of significant differences. Water masses for which we had less than 5 data points for
500 | PFe/PAI were excluded from this test. As the differences in the median values among the treatment groups were
501 | greater than would be expected by chance; the difference in PFe/PAI between water masses is statistically
502 | significant ($P = <0.001$).

503 | As previously seen, the lithogenic imprint is dominant in the WEB, with MW and NEADW showing PFe/PAI
504 | values close to the UCC value of $0.21 \text{ mol mol}^{-1}$. Interestingly, the PFe/PAI signature of $0.36 \text{ mol mol}^{-1}$ within
505 | the old LSW_{WEB} is probably due to the effect of biogenic inputs associated with the strong bloom encountered in
506 | the Irminger Sea than in the WEB (see section 4.3.5).

507 | While it appears that lithogenic particles are dominating the water column in the ~~WEB,WEB~~ and that some
508 | water masses have a clear PFe/PAI fingerprint, it is important to discuss the origin of these signatures, which is
509 | the purpose of the following sections.

510

511 | 4.3. Tracking the different inputs of particulate iron

512 | 4.3.1. Inputs at margins: Iberian, Greenland and Newfoundland

513 | Inputs from continental shelves and margins have been demonstrated to support high productivity in shallow
514 | coastal areas. Inputs of iron from continental margin sediments supporting the high productivity found in
515 | shallow coastal regions have been demonstrated in the past (e.g. Cullen et al. (2009), Elrod et al. (2004), Jeandel
516 | et al. (2011), Ussher et al. (2007)) and sometimes, were shown to be advected at great distances from the coast
517 | (e.g. Lam et al., 2008). ~~Moreover, freshwater inputs that are usually present in these regions can also play a key~~
518 | ~~role in the global biogeochemical cycling of trace metals (Blain and Tagliabue, 2016; Guieu et al., 1991; Martin~~
519 | ~~and Meybeck, 1979). Rivers, runoff and continental glacial melt and/or sea ice melt can also supply dissolved~~
520 | ~~and particulate iron to coastal waters, thus sustaining important phytoplankton production (Fung, 2000).~~

521 | In the following section, we will investigate these possible candidate sources in proximity of the different
522 | margins encountered. Along the GEOVIDE section, sediments at margins were of various compositions
523 | (Dutkiewicz et al., 2015). Sediments originating from the Iberian margin were mainly constituted of silts and
524 | clays (Cacador et al., 1996; Duarte et al., 2014). East Greenland margin sediments were a mixture of sands and
525 | grey/green muds, while, sediments from the West Greenland margin were mainly composed of grey/green muds
526 | (Loring and Asmund, 1996). At the western end of the section, sediments from the Newfoundland margin were
527 | composed of gravelly and sandy muds (Mudie et al., 1984). The different sediment compositions of the three
528 | margins sampled during GEOVIDE have different mineralogy/composition, which are reflected in their
529 | different PFe/PAI ratios (Figure 89). While the Iberian Margin had a PFe/PAI close to UCC ratio, ~~mainly due to~~
530 | ~~seasonal dust inputs from North Africa (Shelley et al., 2017),~~ the highest biogenic contribution ratio could be
531 | seen at the East Greenland (stations 53 and 56) and West Greenland (station 61) Margins, with median PFe/PAI

532 reaching 0.45 mol mol⁻¹. The Newfoundland margin displayed an intermediate behaviour, with Fe/Al ratios of
533 0.35 mol mol⁻¹.

534
535
536 In addition to PAI, PMn can be used as a tracer of inputs from shelf resuspension (Lam and Bishop, 2008).
537 Indeed, Mn is really sensitive to oxidation mediated by bacteria (Tebo et al., 1984; Tebo and Emerson, 1985)
538 and forms manganese oxides (MnO₂). These authigenic particles lead to an enrichment of Mn in particle
539 compositions. In order to track the influence of shelf resuspension, a percentage of sedimentary inputs “%bulk
540 sediment inputs” can be ~~calculated~~ estimated using PMn/PAI ratio from GEOVIDE samples and the PMn/PAI
541 UCC value (0.0034; Taylor and McLennan, 1995) according to the following equation:

$$542 \quad \% \text{bulk sediment PMn} = 100 * \left(\frac{\text{PAI}}{\text{PMn}} \right)_{\text{sample}} * \left(\frac{\text{PMn}}{\text{PAI}} \right)_{\text{UCC ratio}} \quad (3)$$

543 This proxy is a good indicator of direct and recent sediment resuspension. We assume that particles newly
544 resuspended in water column will have the same PMn/PAI ratio than the UCC ratio leading to a “%bulk
545 sediment Mn” of 100%. -This value will decrease by authigenic formation of Mn oxides. This proxy assumes
546 homogeneity of the sediment PMn/PAI ratio through the section which is maybe not completely the case at
547 every station. In consequence, this proxy is only a tool to identify new benthic resuspension at specific location
548 and inter-comparison between several locations is not possible.—When a sample presents a “%bulk sediment
549 Mn” greater than 100%, we assign a value of 100% to simplify the following discussion. -As the Mn cycle can
550 also be affected by biologic uptake (e.g. Peers and Price, 2004; Sunda and Huntsman, 1983), this proxy is only
551 used at depths where biologic activity is negligible (i.e. below 150m depth).

552
553 *The Iberian margin*

554 Coastal waters of the Iberian Shelf are impacted by the runoff for the Tagus River, which is characterised by
555 high suspended matter discharges, ranging between 0.4 to 1 × 10⁶ tons yr⁻¹, and with a high anthropogenic
556 signature (Jouanneau et al., 1998). During the GEOVIDE section, the freshwater input was observable at
557 stations 1, 2 and 4 in the first 20 m; salinity was below 35.2 psu while surrounding waters masses had salinity
558 up to 35.7 psu. Within the freshwater plume, particulate concentrations were important at station 2; ~~with at 20m,~~
559 PFe ~~was of~~ 1.83 nmol L⁻¹. Further away from the coast, the particulate concentrations remained low at 20m
560 depth, with PFe, PAI, and PMn concentrations of 0.77 nmol L⁻¹, 3.5 nmol L⁻¹, and 0.04 nmol L⁻¹, respectively at
561 station 1. The low expansion of the Tagus plume is likely due to the rapid settling of suspended matter. Indeed,
562 our coastal station 2 was already located at around 50 km of the Iberian coast and according to Jouanneau et al.
563 (1998), the surface particle load can be observable at a maximum 30km of the Tagus estuary.
564 Besides, ADCP data acquired during GEOVIDE (Zunino et al., 2017) and several studies have reported an
565 intense current spreading northward coming from Strait of Gibraltar and Mediterranean Sea, leading to a strong
566 resuspension of benthic sediments above the Iberian Shelf, -e.g. Biscaye and Eittrheim (1977), Eittrheim et al.
567 (1976), McCave and Hall (2002), Spinrad et al. (1983). The importance of the sediment resuspension was
568 observable by low beam transmissometry value (87.6%) at the bottom of station 2. This important sediment
569 resuspension led to an extensive input of lithogenic particles within the water column associated with high

570 concentrations of PFe (304 nmol L⁻¹), PAI (1500 nmol L⁻¹), and PMn (2.5 nmol L⁻¹) (Figure 3, Table S1).
571 Moreover, one hundred percent of PFe is estimated to have a lithogenic origin (Figure 140) while 100% of the
572 PMn was estimated to be the result of a recent sediment resuspension according to the %Fe_{litho} and “%bulk
573 sediment Mn” proxies (supplementary material, Table S4S2), confirming the resuspended particle input.

574 At distance from the shelf, within the Iberian Abyssal Plain, an important lateral advection of PFe from the
575 margin was observable (Figure 4410). These lateral inputs occurred at two depth ranges: between 400 and 1000
576 m ~~as~~ seen at stations 4 and 1, with PFe concentrations reaching 4 nmol L⁻¹, and between 2500 m and the bottom
577 (3575 m) of station 1, with PFe concentrations reaching 3.5 nmol L⁻¹. While 100% of PFe had a lithogenic
578 ~~origin~~signature, the sedimentary source input estimation decreased, between 40% and ~~85~~90% of the PMn
579 (Figure 4410). Transport of lithogenic particles was observable until station 11 (12.2°W) at 2500 m where PFe
580 concentration was 7.74 nmol L⁻¹ and 60% of PMn had a sedimentary origin (Figure 949). Noteworthy, no
581 particular increase in PFe, PMn or PAI was seen between 500 and 2000 m depth, where the MOW spreads,
582 which is consistent with that was observed DFe concentrations (Tonnard et al., this issue); ~~l-y-y~~ yet in contrast with
583 the dissolved aluminium values (Menzel Barraqueta et al, subm., this issue) which were high in the MOW and
584 with the study of Ohnemus and Lam (2015) that reported a maximum PFe concentration at 695 m depth
585 associated with the particle-rich Mediterranean Overflow Water (Eittrheim et al., 1976) in the IAP. However,
586 their station was located further south of our station 1. The shallower inputs observed at stations 1 and 4 could
587 therefore be attributed to sediment resuspension from the Iberian margin and nepheloid layer at depth for station
588 1.

589 Therefore, the Iberian margin appears to be an important source of lithogenic-derived iron-rich particles in the
590 Atlantic Ocean; shelf resuspension impact was perceptible until 280 km away from the margin (Station 11) in
591 the Iberian Abyssal Plain.

592

593 *South Greenland*

594 Several studies already demonstrated the importance of icebergs and sea ice as source of dissolved and
595 particulate iron (e.g. van der Merwe et al., 2011a, 2011b; Planquette et al., 2011; Raiswell et al., 2008). The
596 Greenland shelf is highly affected by external fresh water inputs as ice-melting or riverine runoff (Fragoso et al.,
597 2016), that are important sources of iron to the Greenland Shelf (Bhatia et al., 2013; Hawkings et al., 2014;
598 Statham et al., 2008).

599 Both East and West Greenland shelves (stations 53 and 60) had high concentration of particles (beam
600 transmissometry of 83%) and particulate trace elements, reaching 22.1 nmol L⁻¹ and 18.7 nmol L⁻¹ of PFe,
601 respectively (station 53 at 100m and station 61 at 136 m). During the cruise, the relative freshwater observed
602 (S<33 psu) within the first 25 meters of stations 53 and 61 were associated with high PFe (19 nmol L⁻¹), PAI (61
603 nmol L⁻¹), PMn (0.6 nmol L⁻¹) and a low beam transmissometry (≤ 85%) (Figure 409 and Table S1). Particles
604 associated were enriched in iron compared to aluminium, as PFe/PAI ratio was 0.3 within the meteoric water
605 plume. High biological production, in agreement with PP concentrations reaching 197 nmol L⁻¹ induced by the
606 supply of bioavailable dissolved iron from meteoric water (Raiswell et al., 2008; Statham et al., 2008; Tonnard
607 et al., submitted, this issue), ~~leaded~~ led to a transfer of DFe to the particulate phase. This is in line with the fact that
608 around 30% ~~of the~~ PFe had a non-lithogenic ~~and likely biogenic~~ origin. In addition, only ~~35~~40% ~~of the~~ PMn
609 originated from resuspended sediments. Interestingly, these two proxies remained constant from the seafloor to

610 | the surface (Station 49, Figure ~~4+10~~), with around 25% of the PMn of sedimentary origin, which could be due to
611 | an important mixing happening on the shelf. The lithogenic PFe could result from the release of PFe from
612 | Greenland bedrock captured during the ice sheet formation on land.

613 | The spatial extent of the off-shelf lateral transport of particles was not important on the east Greenland coast.
614 | Indeed, no visible increase of particulate trace metal concentrations was visible at the first station off-shelf,
615 | station 60 (Figure ~~4+10~~), except at 1000 m depth, where a strong increase (up to ~~8990~~) of sedimentary PMn
616 | was seen. This is probably due to the East Greenland Coastal Current (EGCC) that was located at station 53
617 | constrained these inputs while stations 56 and 60 were under the influence of another strong current, the East
618 | Greenland-Irminger current (EGIC) (Zunino et al., 2017).

619 |
620 | To the west of the Greenland margin, lateral transport of particles was slightly more important. Noticeable
621 | concentrations of particulate lithogenic elements were observable until station 64 located 125 km away from
622 | shoreline. These particles had decreasing PFe lithogenic contribution (~~520~~) with a similar (~~257~~) sedimentary
623 | PMn content than closer to the margin. The increasing nature of non-lithogenic PFe is linked to the bloom in
624 | surface (associated with a PFe/PAI ratio of 0.30 mol mol⁻¹, ~~and~~ a PP of 197 nmol L⁻¹ at station 61 ~~and a Chl-a~~
625 | ~~concentrations of 6.21 mg m⁻³~~), with the biogenic PFe settling down along the transport of particles.

626 |
627 | Therefore, particles newly resuspended from Greenland sediments are an important source, representing around
628 | a third of the pMn pool, combined with surface inputs such as riverine runoff and/or ice-melting that are
629 | delivering particles on the shelf and biological production. Unlike the Iberian shelf, Greenland margin was not
630 | an important provider of particulate metals inside the Irminger and Labrador Basin, due to the circulation that
631 | constrained the extent of the margin plume.

632 | 633 | *The Newfoundland Shelf*

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

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

643 | The proportion of lithogenic PFe was relatively high and constant in the entire water column, with a median
644 | value of ~~67-70~~%. At station 78, ~~95-100~~% of the PMn had a sedimentary origin close to the seafloor (371 m). The
645 | spreading of the recent sediment resuspension was observable until 140 m depth where the contribution of
646 | sedimentary Mn was still 51% (Figure ~~4+10~~, Table S2). This could correspond to an intense nepheloid layer as
647 | previously reported by Biscaye and Eitrem (1977) (see also section 3.3.2). The high PFe concentration (184
648 | nmol L⁻¹, station 78, 371 m) associated with a high percentage of sedimentary PMn (95%) observed at the

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649 bottom of this station, was therefore the result of an important resuspension of shelf sediments. This was
650 confirmed with low transmissometry values of 95%.

651 ~~Despite t~~The important phytoplanktonic community present (~~maximum Chl-a= 4.91 mg m⁻³, Tonnard et al., in~~
652 ~~prep), is linked to the low~~ PFe ~~remained low of~~at 0.79 nmol L⁻¹ at 10 m, but, ~~with the~~ high PFe/PAI ratio, up to
653 0.4, and ~~the~~PP concentration of 97 nmol L⁻¹, ~~confirming~~ the biologic influence. Either the biogenic particles
654 settled quickly, and/or they were quickly remineralized. Concerning this latter process, intense remineralization
655 ~~at station 77 (7 mmol C m⁻² d⁻¹ compared to 4 mmol C m⁻² d⁻¹ in the Western European Basin) at station 77~~ has
656 been reported by Lemaitre et al. (2018a ~~and 2018b~~), which could explain the low PFe values throughout the
657 water column.

658 ~~That said, resuspended particles are were still laterally transported off shelf until station 71 (Figure 6) where~~
659 ~~PFe concentrations were higher than the background value, up to 2 nmol L⁻¹ at depths greater than 100 m.~~

661 Along the GEOVIDE section, continental shelves provided an important load of particles within the surrounding
662 water column. The three margins sampled during GEOVIDE behaved differently; the Iberian margin discharged
663 high quantities of lithogenic particles far away from the coast while the Greenland and Newfoundland margins
664 did not reveal important PFe concentrations. Spreading of particles is tightly linked to hydrodynamic conditions,
665 which in the case of the Greenland margin, prevented long distance seeding of PFe. Moreover, each margin
666 showed a specific PFe/PAI ratio (Figure ~~89~~) indicating different composition of the resuspended particles.
667 Resuspended particles represent the composition of sediment at the margin if oxido-reductive transformation of
668 iron and aluminium are considered negligible under these circumstances. Differences between margins were due
669 to the presence of non-crustal particles, ~~either biogenic or authigenic~~. Biological production in surface waters
670 ~~and authigenic formation of iron hydroxide~~ produces particles with a higher PFe/PAI content and their export
671 through the water column to the sediment increased the PFe/PAI ratio at depth. Regions where biological
672 production is intense such as in the vicinity of Newfoundland presented higher PFe/PAI ratios of resuspended
673 benthic particles. ~~These results are in agreement with the study of Lam et al. (2017), which showed the different~~
674 ~~behaviour between margins are a function of several parameters such as boundary currents, internal waves and~~
675 ~~margin sediment composition.~~

677 4.3.2 Benthic resuspended sediments

678 Benthic nepheloid layers (~~BNLs~~) (~~BNLs~~) can play a significant role in trace element distributions at depth as
679 ~~previously described (Dutay et al., 2015; Lam et al., 2015; Ohnemus et al., 2015; Revels et al., 2015)~~. BNLs are
680 important layers where local resuspension of sedimentary particles (Bishop and Biscaye, 1982; Eittrreim et al.,
681 1976; Rutgers Van Der Loeff et al., 2002) occur due to strong hydrographic stresses (~~i.e. boundary currents,~~
682 ~~benthic storms and deep eddies~~) interacting with the ocean floor (~~Biscaye and Eittrreim, 1977; Eittrreim et al.,~~
683 ~~1976; Gardner et al., 2017, 2018)~~. ~~In the North Atlantic, boundary currents were suspected to be the origin of~~
684 ~~theses stresses (Biscaye and Eittrreim, 1977; Eittrreim et al., 1976) but more recent studies demonstrate the~~
685 ~~essential role of benthic storms and deep eddies (Gardner et al., 2018)~~. Along the GA01 section, BNLs were
686 observable in each province with different strengths (Figures 3 and 12).

687

688 In BNLs located within the WEB, PFe concentrations reached up to 10 nmol L⁻¹ (stations 26 and 29, [Table S1,](#)
689 [supplementary material](#)). These concentrations were smaller than PFe concentrations encountered in BNL from
690 the Icelandic, Irminger and Labrador Basins, where benthic resuspension led to PFe concentrations higher than
691 40 nmol L⁻¹, even reaching 89 nmol L⁻¹ at the bottom of station 71 (3736 m). Moreover, in the Irminger and
692 Labrador Basins, PFe/PAI molar ratios within BNLs were higher than the ones measured within the WEB at
693 station 26 and 29. In the Irminger Basin, PFe/PAI reached 0.4 mol mol⁻¹, which could reveal a mixture of
694 lithogenic and biogenic matter previously exported. This feature was also observed in the Labrador Basin, with
695 PFe/PAI ratio ranging between 0.34 and 0.44 mol mol⁻¹. In contrast, BNLs sampled in the WEB have clearly a
696 lithogenic imprint, with PFe/PAI molar ratios close to the crustal one. Resuspended sediments with a non-crustal
697 contribution seem to hold a higher PFe content than sediments with a lithogenic characteristic. Nevertheless,
698 interestingly all BNLs present during GEOVIDE were spreading identically, with impacts observable up to 200
699 meters above the oceanic seafloor (Figure 112), as reflected in beam transmissometry values, and PFe
700 concentrations, that returned to a background level at 200 m above the seafloor. The presence of these BNLs has
701 also been reported by Le Roy et al. (submitted, this issue). Important differences of PFe intensities could also be
702 due to different hydrographic components and topographic characteristics. As previously explained, two main
703 triggers of BNLs are benthic storms and deep eddies; by definition these processes are highly variable
704 geographically and temporally, but no physical data could allow us to investigate further this hypothesis.
705 Along the GEOVIDE section, BNLs are providing high concentrations of particulate trace element in the deep
706 open ocean, contributing highly to the total trace elements budget as iron.~~Important differences of PFe~~
707 ~~intensities could also be due to different hydrographic components and topographic characteristics. As~~
708 ~~previously explained, two main triggers of BNLs are benthic storms and deep eddies; by definition these~~
709 ~~processes are highly variable geographically and temporally, but no physical data could allow us to investigate~~
710 ~~further this hypothesis.~~

711
712

4.3.3. Reykjanes Ridge inputs

713 ~~Recently, hydrothermal inputs of iron in the open ocean have been re-evaluated by (Fitzsimmons et al., 2017;~~
714 ~~Resing et al., 2015; Tagliabue et al., 2014). These studies demonstrated the importance of hydrothermal~~
715 ~~activities on the global iron biogeochemical cycle through particulate and dissolved iron fluxes. During the~~
716 ~~cruise, samples of station 38 have been collected above the Reykjanes Ridge, the upper section of the Mid-~~
717 ~~Atlantic Ridge in the North Atlantic, which has inferred hydrothermal sites from several studies conducted in~~
718 ~~the area (Baker and German, 2004). Above the ridge, high PFe concentrations were measured, reaching 16 nmol~~
719 ~~L⁻¹ just above the seafloor, while increased DFe concentrations were reported to the East of the ridge (Tonnard~~
720 ~~et al., this issue). The exact sources of iron-rich particles cannot be well constrained, as they could come from~~
721 ~~active hydrothermal vents or resuspension of particulate matter from new crustal matter produced at the ridge.~~
722 ~~According to the oceanic circulation (Zunino et al., 2017; Garcia-Ibanez et al., 2017), hydrothermal particles~~
723 ~~could have been seen in the ISOW within the Icelandic Basin. Nevertheless, at the vicinity of the ridge, scanning~~
724 ~~electron microscope (SEM) analyses of our samples did reveal a number of biological debris and clays but not~~
725 ~~the presence of iron (oxy-)hydroxide particles, which are known to be highly produced close to hydrothermal~~
726 ~~vents (Elderfield and Schultz, 1996). Their absence could thus indicate an absence of vents. However, other~~

727 proxies, such as helium-3, are necessary to claim with more accuracy the presence or absence of an
728 hydrothermal source close to station 38.

729
730 ~~Alternatively, resuspended sediments transported with ISOW flowing across the Reykjanes Ridge could explain~~
731 ~~the high PFe concentrations below 1000 m depth at station 38. This feature was associated with lower median~~
732 ~~PFe concentrations and PFe/PAI ratios (Figure 7) at station 40 (2.2 nmol L^{-1} , and $0.58 \text{ mol mol}^{-1}$ respectively)~~
733 ~~than at station 38 (6.8 nmol L^{-1} , $0.48 \text{ mol mol}^{-1}$ respectively). Moreover, PMn had a 19% sedimentary origin,~~
734 ~~constant from the bottom to 1163 m depth, a contribution that is very low for the shallower water depths.~~
735 ~~Consequently, the increase in PFe within the ISOW_{west} more likely came from sediment resuspension as the~~
736 ~~ISOW_{east} flows through the Charlie Gibbs Bight Fracture Zones.~~

737

738 4.3.4. Atmospheric inputs

739 Atmospheric deposition is an important input of trace elements in surface of the open ocean (e.g. (Jickells et al.,
740 2005). Atmospheric inputs, both wet and dry, were reported to be low during the GEOVIDE cruise (Menzel-
741 barraqueta et al., 2018, this issue; Shelley et al., 2017; 2018). In fact, oceanic particles measurements in surface
742 waters along the section did not reveal high PFe or PAI concentrations, therefore, the surface composition of
743 particles did not seem to be highly affected by atmospheric deposition at the time of the cruise. ~~However,~~
744 ~~PFe/PAI ratio was closed to the UCC one, probably due to the overall influence of atmospheric deposition in~~
745 ~~this area.~~ One pattern is also interesting to note: the surface waters of the Iberian Abyssal Plain and Western
746 European Basin, between stations 11 and 23 presented a characteristic feature with really low PFe/PAI
747 elemental ratios, of 0.11, smaller than the UCC ratio of 0.21 (Figure 6). Such low ratios have been reported in
748 the same region by Barrett et al. (2012). One possible explanation is given by Buck et al. (2010) who described
749 Fe-depleted aerosols in this area of the North Atlantic with PFe/PAI ratio below UCC ratio. However, Shelley et
750 al. (2017) found a higher PFe/PAI ratio around 0.25 in this area (their sample geo5-6). This result, highlights
751 some of the difficulties that link atmospheric inputs to water column data (Baker et al., 2016), and implies a
752 probable fractionation after aerosol deposition. In addition, there is high spatial and temporal variability of
753 atmospheric deposition (Mahowald et al., 2005) and a certain degree of uncertainty about the dissolution
754 processes of atmospherically-transported particles (Bonnet and Guieu, 2004).

755

756 ~~4.3.5. Influence of phytoplankton assemblages, remineralisation and scavenging in~~ 757 ~~the upper water column~~

758 ~~Biological activity in surface waters impacts the particle composition in the upper water column. In bulk particle~~
759 ~~samples, direct measurement of the biogenic metal fraction is not possible due to the heterogeneity of particles,~~
760 ~~and in particular, the presence of lithogenic particles (Collier and Edmond, 1984). It is however possible to~~
761 ~~estimate the $\text{PFe}_{\text{nonlithic}}/\text{PFe}_{\text{excess}}/\text{PP}$ molar ratios, based on Eq. (1) and (23), and assuming that most of the PP is of~~
762 ~~biogenic origin. As 100% of the PFe was estimated to be of lithogenic origin, stations 1 to 26 are excluded from~~
763 ~~the discussion below. Overall, the median $\text{PFe}_{\text{excess}}/\text{PP}$ molar ratios varied from 1.0 (Irminger Basin) to 38.7~~
764 ~~mmol mol^{-1} (Greenland margin) in the upper 50 m. These ratios are consistent with the few available bulk~~

765 PFe/PP ratios available in the literature (Twining and Baines, 2013 and references therein), ranging from 1 to 31
766 mmol mol⁻¹ and the phytoplankton assemblages encountered during GEOVIDE (Tonnard et al., in prep.). This
767 corresponds to a % PFe_{bio} of x to x% in the upper 50m. Interestingly, the biogenic contribution is higher in the
768 basin than in the basin.

769 Overall, the median PFe_{nonlitho}/PP molar ratios varied from 1.0 (Irminger Basin) to 38.7 mmol mol⁻¹ (Greenland
770 margin) in the upper 50 m. These ratios are consistent with the few available bulk PFe/PP ratios available in the
771 literature (Twining and Baines, 2013 and references therein), ranging from 1 to 31 mmol mol⁻¹ and the
772 phytoplankton assemblages encountered during GEOVIDE (Tonnard et al., in prep.). Indeed, the highest
773 PFe_{nonlitho}/PP molar ratio determined at stations 53 and 56 close to the South Greenland margin coincide
774 with a bloom mostly composed of large diatoms, whereas, the smallest ratios were associated with a bloom
775 mainly composed of cyanobacteria and haptophytes (Tonnard et al., in prep.). The effect of biological uptake is
776 also clearly visible when looking at PFe/PAI vertical variation, which increases from the surface to
777 approximately 100m depth (Figure 13), except in the Iberian Margin, which is under the strong influence of
778 lithogenic inputs.

781 At deeper depths, pelagic remineralisation processes influence the composition of particles (Barbeau et al.,
782 1996, 2001; Boyd et al., 2010; Strzpek et al., 2005). Taking in account remineralization depths that are derived
783 from Baxs proxy which is described and discussed in great detail in Lemaitre et al. (this issue), it is possible to
784 look at the vertical variation of PFe_{bio}/PP along the section (Figure 13).

785 Close to the IM and within the IAP, no PFe/PAI decrease that could point to a preferential remineralisation of
786 PFe over PAI could be observed within the remineralisation depth (200 to 400 m depth, Figure 13), whereas
787 preferential remineralisation of PP over PFe occurs, as reflected by increasing PFe/PP ratios (Figure 14). This is
788 probably due to the fact that remineralisation rates were low (Lemaitre et al., 2018a), and that PFe was mostly of
789 lithogenic origin, more difficult and slow to remineralize (Boyd et al., 2010). Below 600 m depth, scavenging
790 processes could explain the increasing PFe:PP ratios, from 0.30 to 0.80 mol mol⁻¹ at station 13, which is
791 consistent with decreasing dFe concentrations within this depth range reported in Tonnard et al. (this issue).

792 Within the WEB, between 200 and 500 m depth, remineralisation of PFe over PAI occurs, although reported to
793 be small (Lemaitre et al., 2018a) as reflected by decreasing PFe:PAI ratios (Figure 13), while PFe:PP ratios
794 remained constant, pointing out to similar remineralisation rates of PFe and PP. Below 600 m depth, a stronger
795 scavenging of DFe onto particles formation of Fe oxyhydroxydes (si tu peux calculer et reporter les % à ces
796 intervalles de profondeur, ça aiderait) than in IM and IAP is likely to explain the increasing ratios of PFe:PAI
797 from 0.18 to 0.30 mol mol⁻¹ and PFe:PP from 0.047 to 0.367 (Station 21), and from 0.16 to 1.05 (Station 26) mol
798 mol⁻¹. Similar patterns occur in IeB (station 32), as dFe⁻ concentrations increased (Tonnard et al., this issue)
799 therefore ruling out the possibility of PFe enrichment from scavenging.

801 Above the RR, and in the IrB, at station 38, PFe_P is remineralized preferentially over PAI and PFe_P, with
802 decreasing increasing PFe:PAI ratios from 0.46 to 0.19 mol mol⁻¹ and decreasing PFe:PP ratios from 0.24 to
803 0.04 mol mol⁻¹. This interesting ese features is are associated with moderate and high POC remineralisation

804 ~~fluxes (Lemaitre et al., 2018a) at stations 38 and 44 respectively (Figure 13), and the fact that a stronger fraction~~
805 ~~of PFe was associated with biogenic lithogenic material ($PFe_{bio} = \%$, $PFe_{litho} = \%$), easier to recycle.~~

806 ~~In the IrB, PP is preferentially remineralized over PFe and PAI, as reflected by increasing PFe:PP ratios and~~
807 ~~constant PFe:PAI ratios within the remineralisation depth. This is associated with high POC remineralisation~~
808 ~~fluxes (Lemaitre et al., 2018a) and a high proportion of lithogenic PFe.~~

809 ~~Finally, within the LB, PFe:PAI and PFe:PP remained constant within the deep remineralisation depth,~~
810 ~~extending from 200 to 1000 m depth due to the deep convection of the LSW (Lemaitre et al., 2018a). In this~~
811 ~~basin, either PP and PFe are remineralized at a constant rates, but the fact that dFe is depleted (Tonnard et al.,~~
812 ~~this issue) in this depth range points out to the influence of scavenging. Below 1000 m, PFe:PP ratios increase~~
813 ~~from 0.29 to 0.85 mol mol⁻¹, while PFe:PAI ratios still remain constant. This could be explained by the fact that~~
814 ~~most PP has been recycled due to the strongest remineralisation fluxes reported in this area (Lemaitre et al.,~~
815 ~~2018a).~~

816

817 **5. Conclusions**

818 This investigation of the PFe compositions of suspended particulate matter in the North Atlantic indicates the
819 pervasive influence of crustal particles, augmented by sedimentary inputs at margins, and at depths, within
820 benthic nepehloid layers.

821 Indeed, along the GEOVIDE section, continental shelves provided an important load of particles within the
822 surrounding water column, with PFe mostly residing in non-biogenic particulate form. The Iberian margin
823 discharged high quantities of lithogenic particles originating from riverine inputs far away from the coast while
824 the Greenland margin did not reveal a long distance seeding of PFe, due to hydrodynamic conditions. Both
825 Greenland and Newfoundland margins PFe resuspended particles were under a strong biogenic influence that
826 were exported at depth. This resulted in different remineralisation fluxes among the different provinces.

827 Scavenging processes could also be visible at depths greater than 1000 m, these effects being the most
828 pronounced within the [WEBLabrador Basin](#).

829 Finally, resuspended sediments above the Reykjanes Ridge increased the PFe composition of the Iceland
830 Scottish Overflow Water. A similar feature occurs for the Labrador Sea Water, as it flows from the Irminger
831 Basin to the Western European Basin.

832

833

834

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844

845 **References**

846

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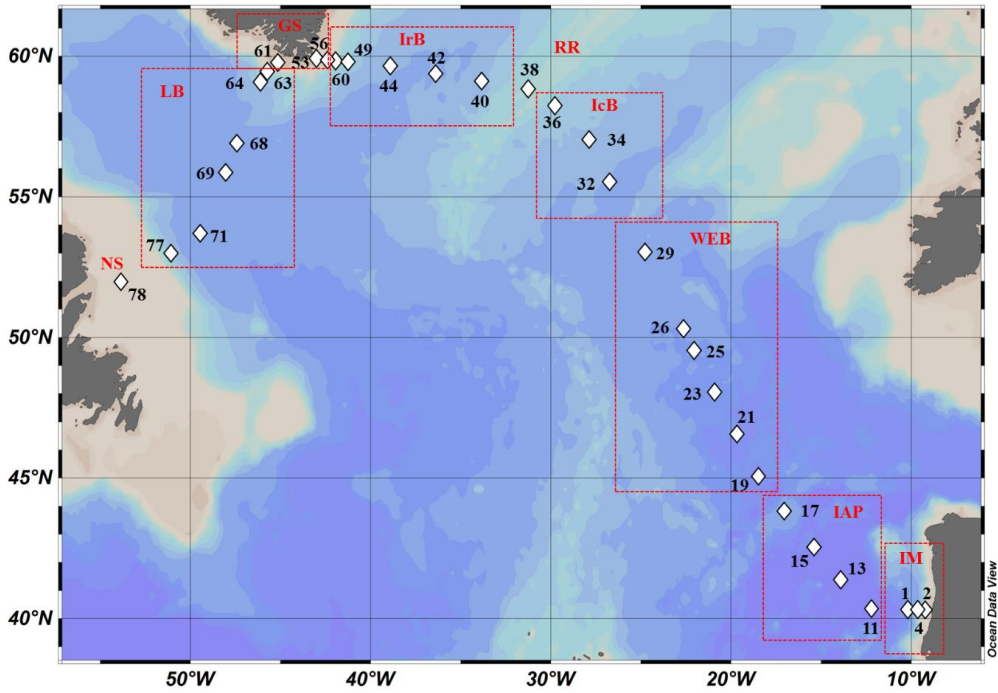
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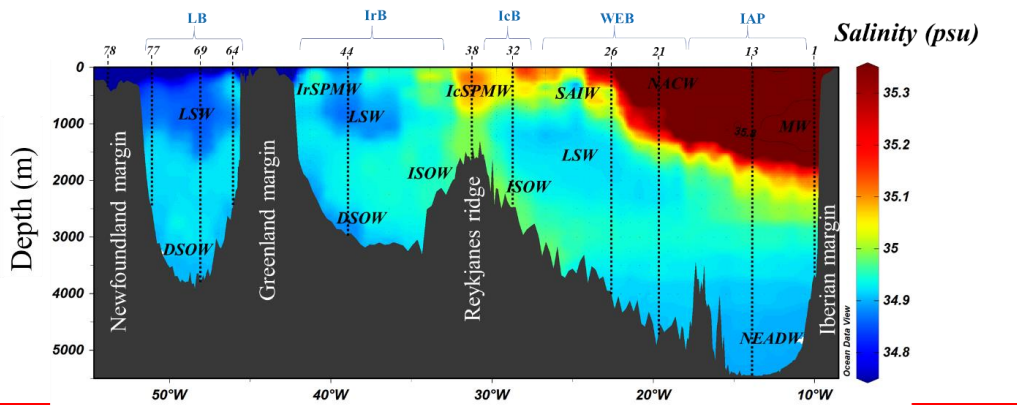
1183 **Figure 1:** Map of stations where suspended particle samples were collected with GO-FLO bottles during the
1184 GEOVIDE cruise (GA01). Biogeochemical provinces are indicated by red squares, IM: Iberian Margin, IAP: Iberian
1185 Abyssal Plain, WEB: Western European Basin, IcB: Iceland Basin, RR: Reykjanes Ridge, IrB: Irminger Basin, GS:
1186 Greenland Shelf, LB: Labrador Basin, NS: Newfoundland Shelf. This figure was generated by Ocean Data View
1187 (Schlitzer, R., Ocean Data View, odv.awi.de, 2017).



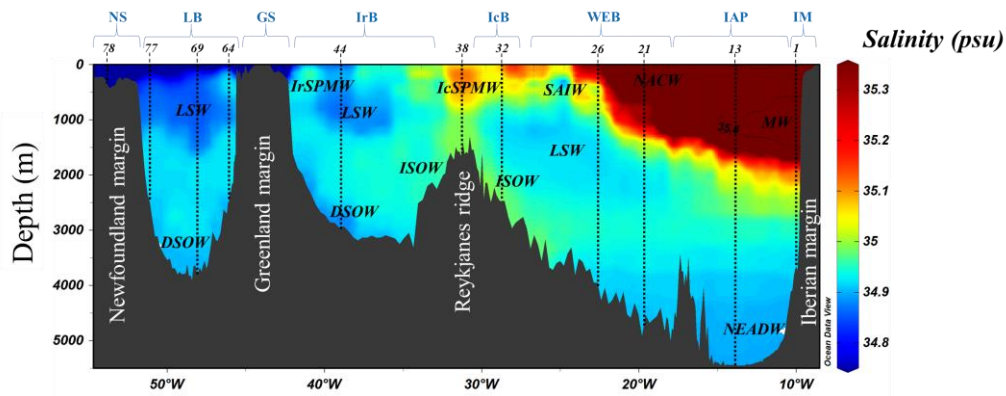
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Figure 2: Salinity section during the GEOVIDE cruise. Water masses are indicated in black, MW: Mediterranean Water; NACW: North Atlantic Central Water; NEADW: North East Atlantic Deep Water; LSW: Labrador Sea Water; ISOW: Iceland-Scotland Overflow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

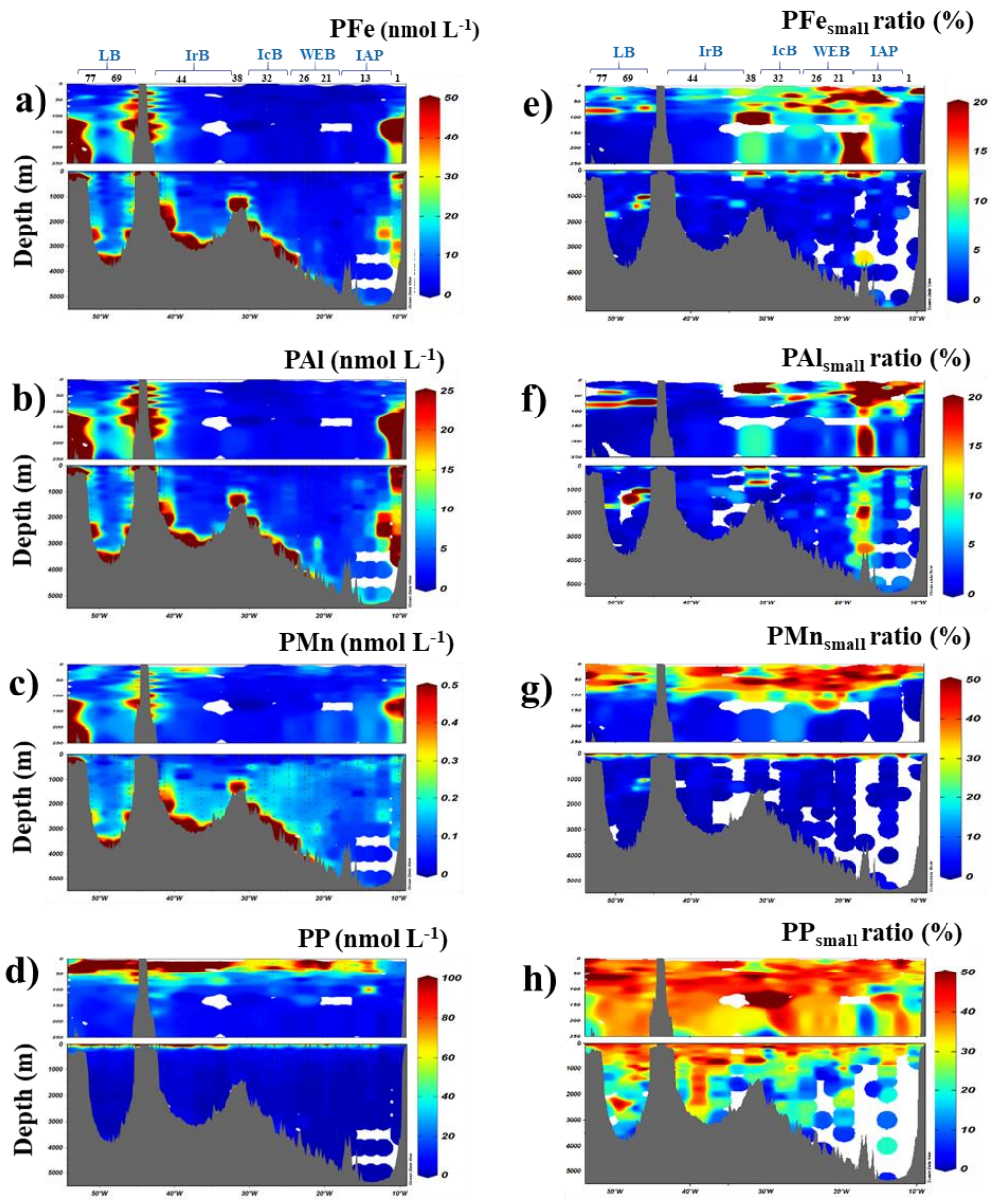


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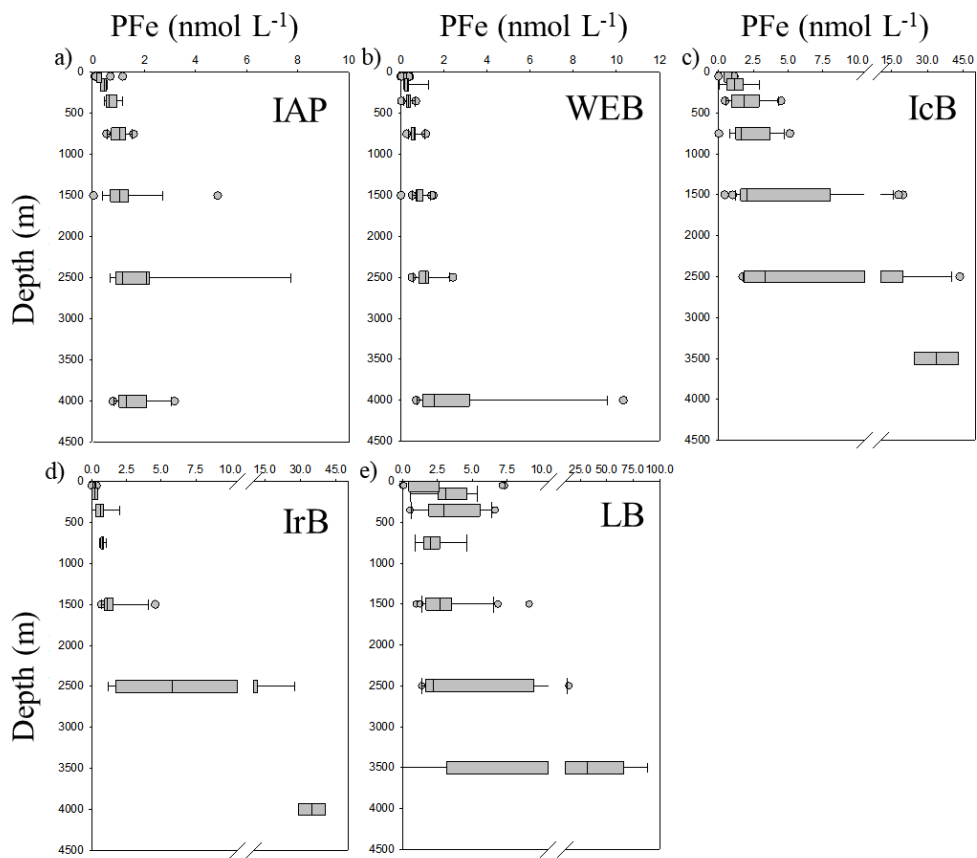
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1233 **Figure 3:** Left) Distribution of total particulate iron (a, PFe), aluminium (b, PAI), manganese (c, PMn) and
 1234 phosphorus (d, PP) concentrations (in nmol L^{-1}) along the GEOVIDE section. Right) Contribution of small size
 1235 fraction ($0.45\text{-}5\ \mu\text{m}$) expressed as percentage (%) of the total concentration of PFe (e), PAI (f), PMn (g) and PP (h).
 1236 Station IDs and biogeochemical region are indicated on top of section a. This figure was generated by Ocean Data
 1237 View (Schlitzer, R., Ocean Data View, odv.awi.de, 2017).



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1242 **Figure 4:** Boxplot figure of the particulate iron vertical profile (in nmol L^{-1}) in the a) Iberian abyssal plain (IAP),
 1243 b) Western European basin (WEB), c) Icelandic basin (IcB), d) Irminger basin (IrB) and e) Labrador basins (LB). The
 1244 left boundary of the box represents the 25th percentile while the right boundary represents the 75th percentile, the line
 1245 within the box marks the median value. Whiskers represent the 90th and 10th percentiles and dots are the outlying
 1246 data. Seven depth boxes have been used (0-100m, 100-200m, 200-500m, 500-1000m, 1000-2000m, 2000-3000m and
 1247 3000m-bottom depth).



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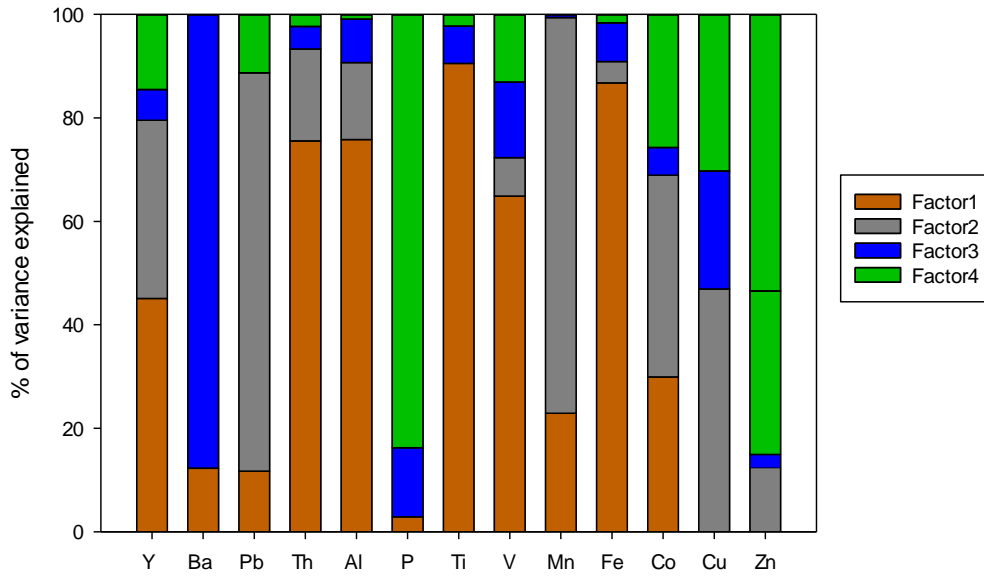
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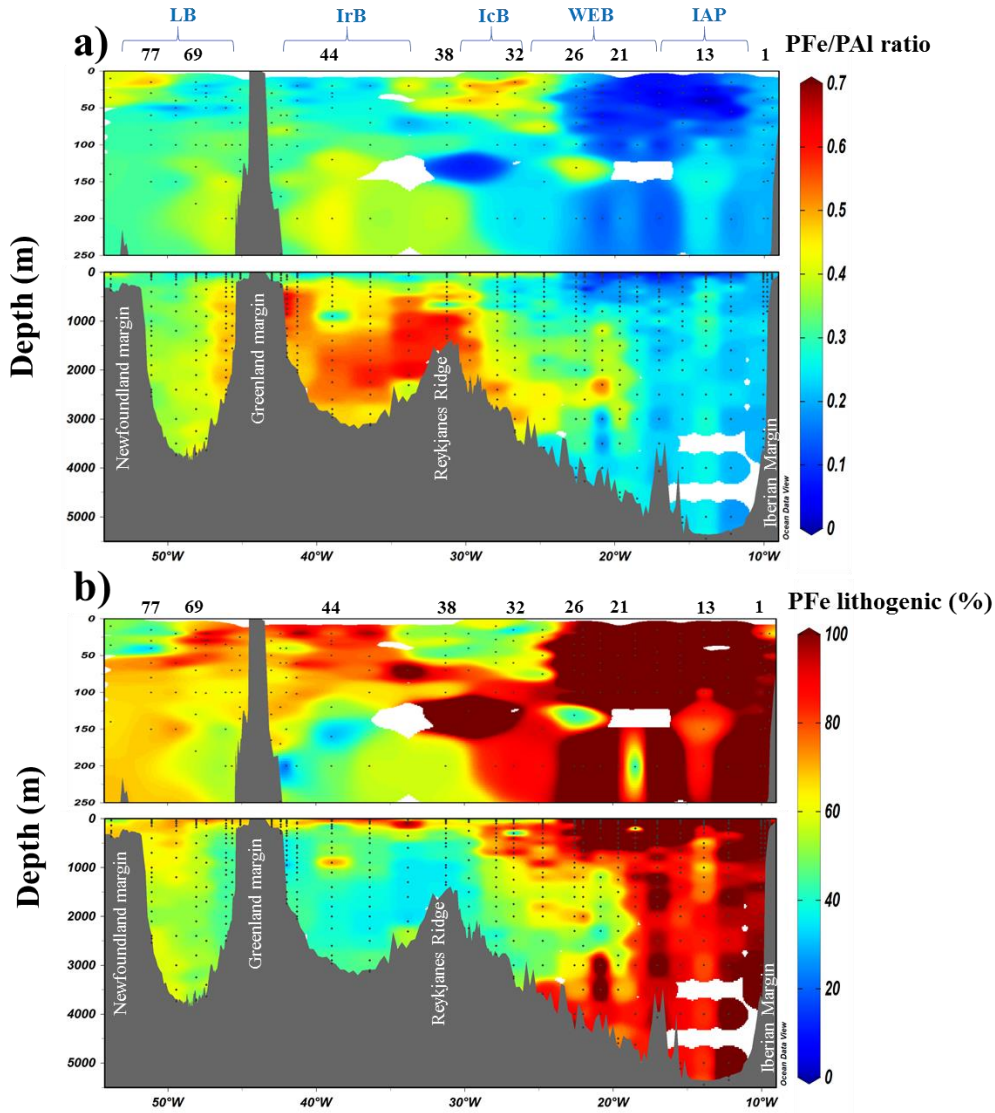
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1261 Figure 5: Factor fingerprint of the positive matrix factorisation. The four factors are represented in a stacked bar
 1262 chart of the percentage of variance explained per element.



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1282 Figure 6: a) Section of the PFe to PAI molar ratio (mol mol^{-1}); (b) contribution of lithogenic PFe (%) based on Eq. (1).
 1283 Station IDs and biogeochemical provinces are indicated above each section. This figure was generated by Ocean Data
 1284 View (Schlitzer, R., Ocean Data View, odv.awi.de, 2017).



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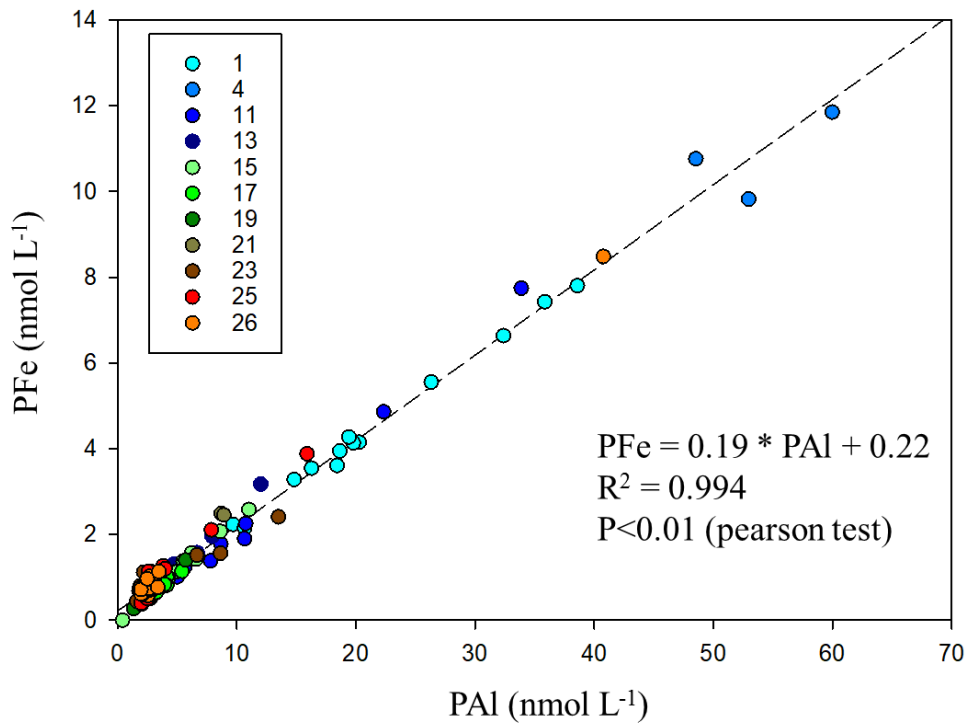
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Figure 7: PFe over PAI concentrations (nmol L^{-1}) for all stations located in the Iberian Abyssal Plain and Western European Basin. Note that the total concentrations of the two elements covaried strongly.



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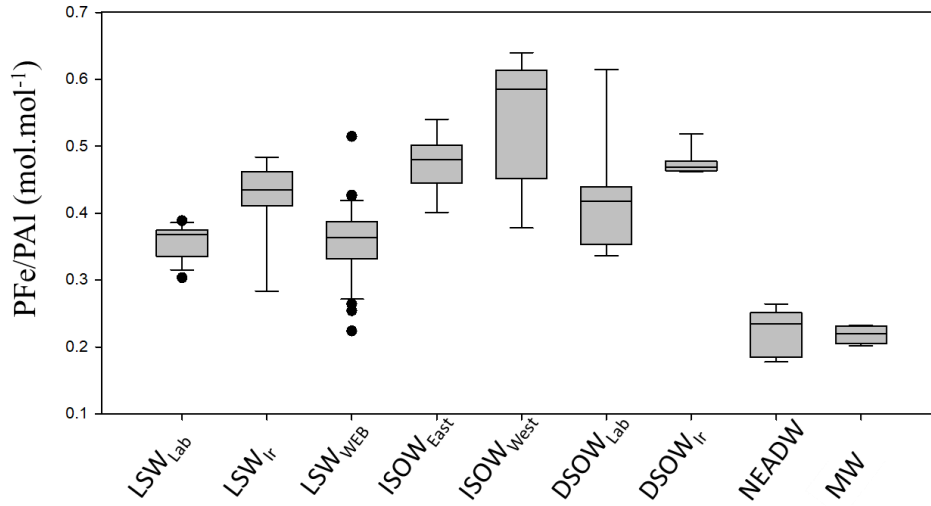
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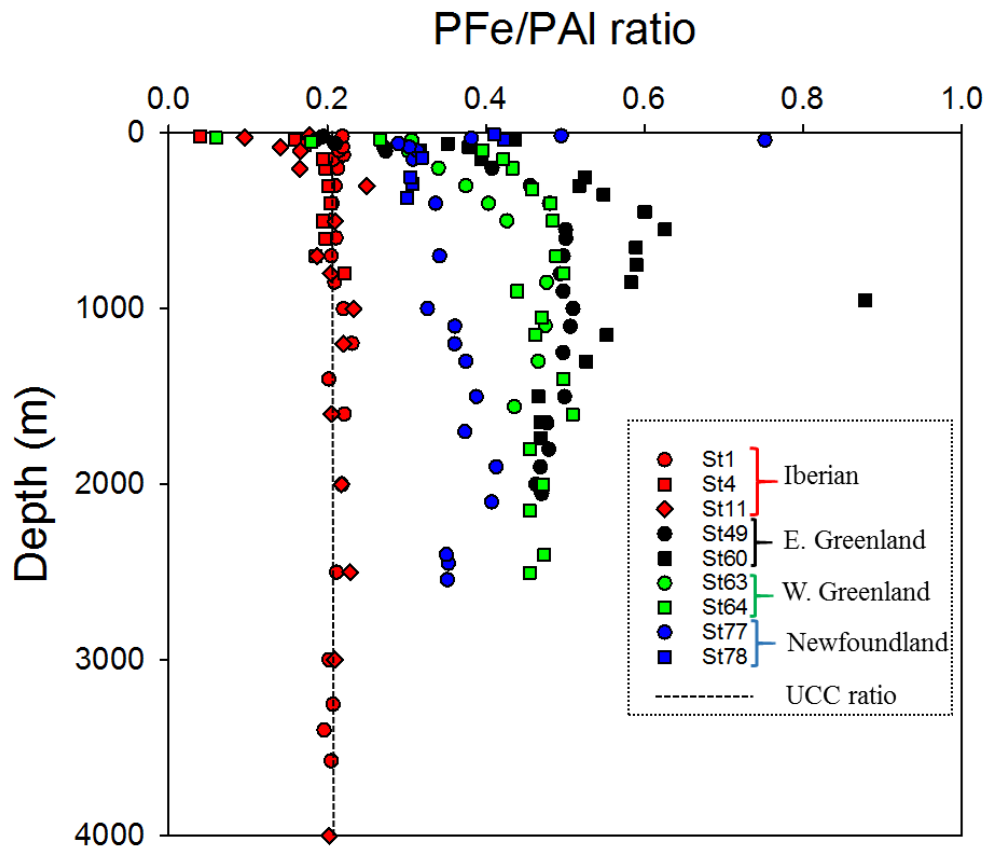
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1307 | Figure 87: Whisker diagram of PFe/PAI molar ratio (mol mol^{-1}) in the different water masses sampled along the
 1308 GA01 line. Median values for the water masses were as follows: $\text{LSW}_{\text{lab}}=0.37$; $\text{LSW}_{\text{lr}}=0.44$; $\text{LSW}_{\text{WEB}}=0.36$;
 1309 $\text{ISOW}_{\text{east}}=0.48$; $\text{ISOW}_{\text{west}}=0.58$; $\text{DSOW}_{\text{lab}}=0.42$; $\text{DSOW}_{\text{lr}}=0.47$; $\text{NEADW}=0.23$; $\text{MW}=0.22 \text{ mol mol}^{-1}$.



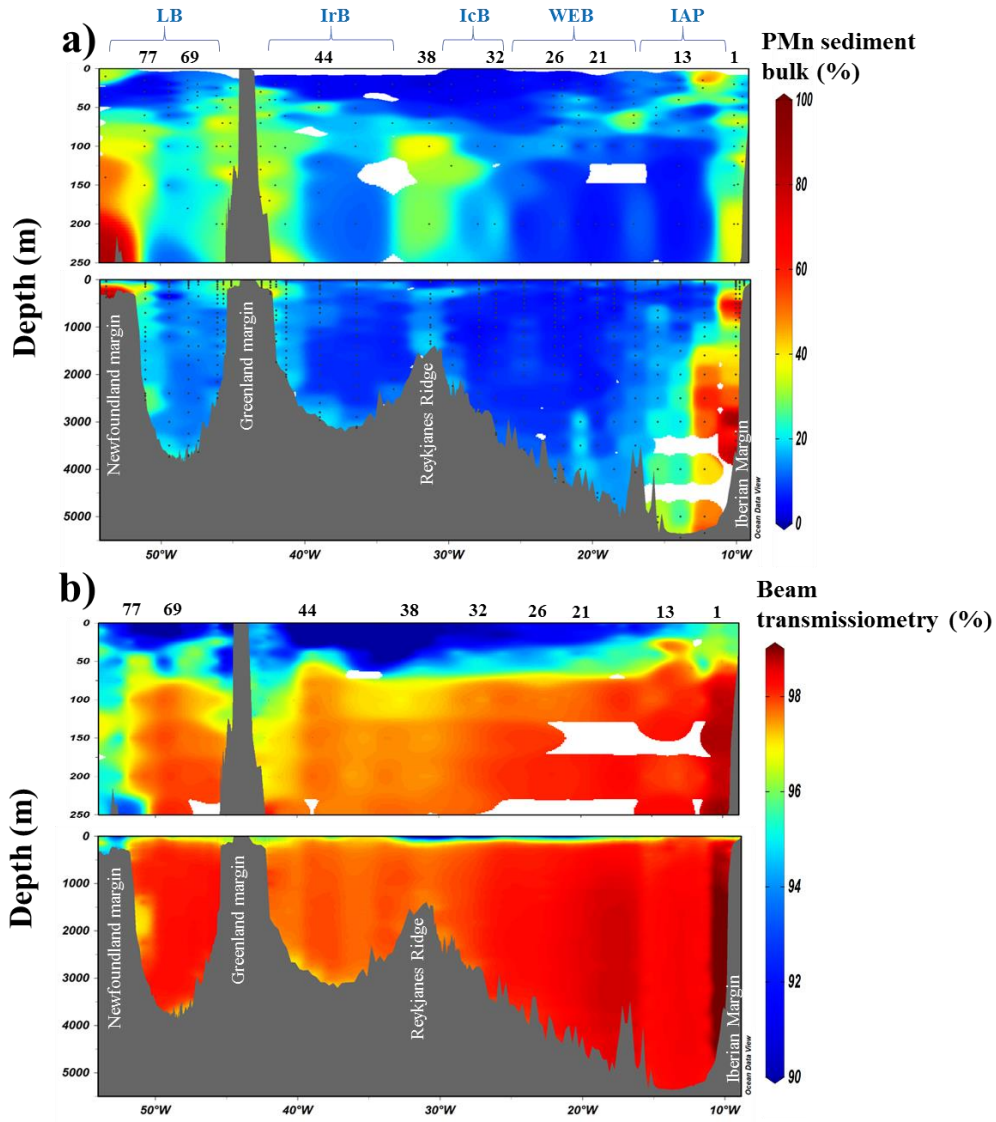
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1330 | Figure 98: Scatter of the PFe/PAI ratio at the Iberian (red dots), East Greenland (black dots), West Greenland (green
1331 dots) and Newfoundland margins (blue dots). Dashed line indicate the UCC ratio (Taylor and McLennan, 1995).



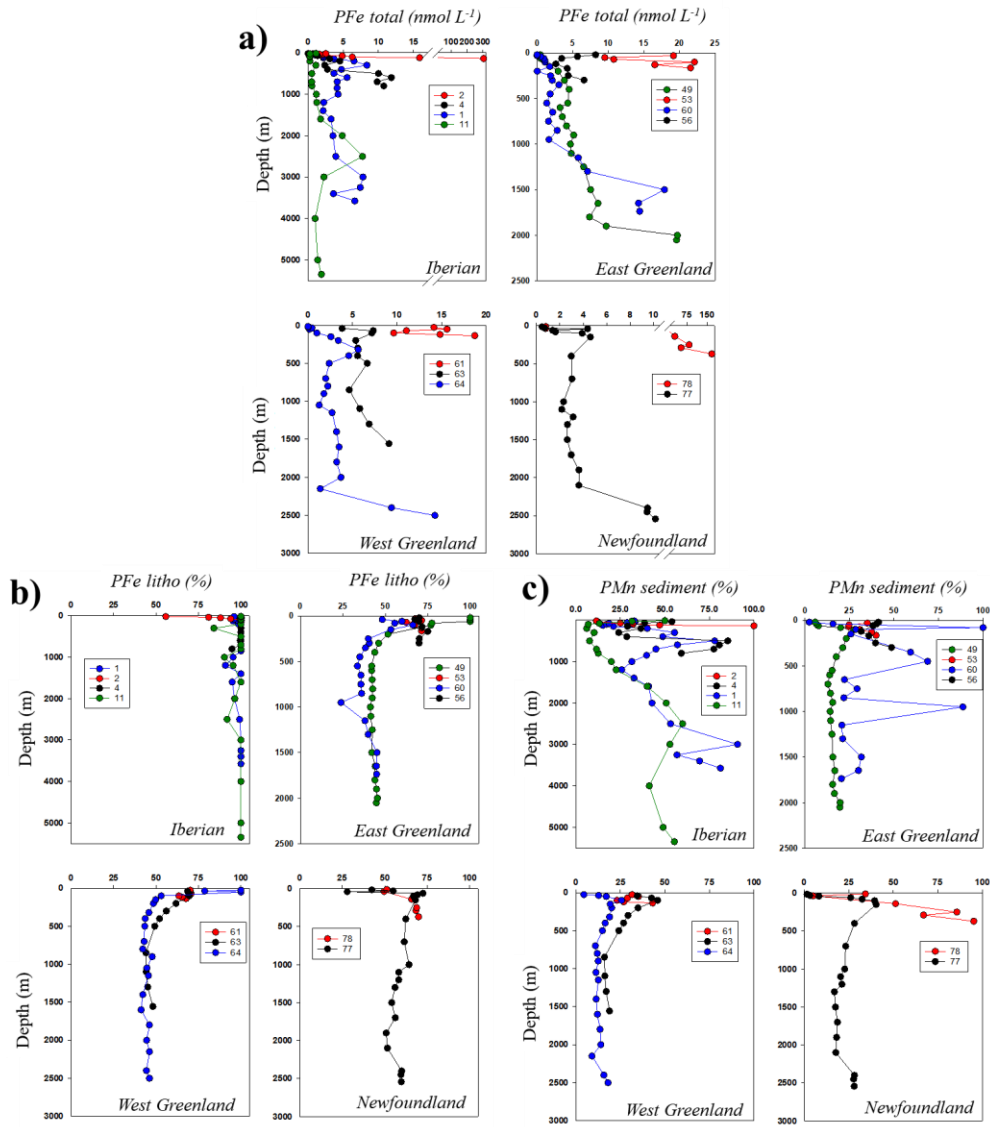
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1343 | Figure 109: Section of derived contribution of sedimentary inputs manganese bulk sediment proxy (a) and
 1344 transmissometry (b) along the GA01 section. Station IDs and biogeochemical region are indicated above the section
 1345 (a). This figure was generated by Ocean Data View (Schlitzer, R., Ocean Data View, odv.awi.de, 2017).



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1352 | Figure 140: Vertical profiles of PFe (nmol L⁻¹), lithogenic proportion of particulate iron (%), and sedimentary
 1353 proportion of particulate manganese (%) at the Iberian, East-West Greenland and Newfoundland margins.



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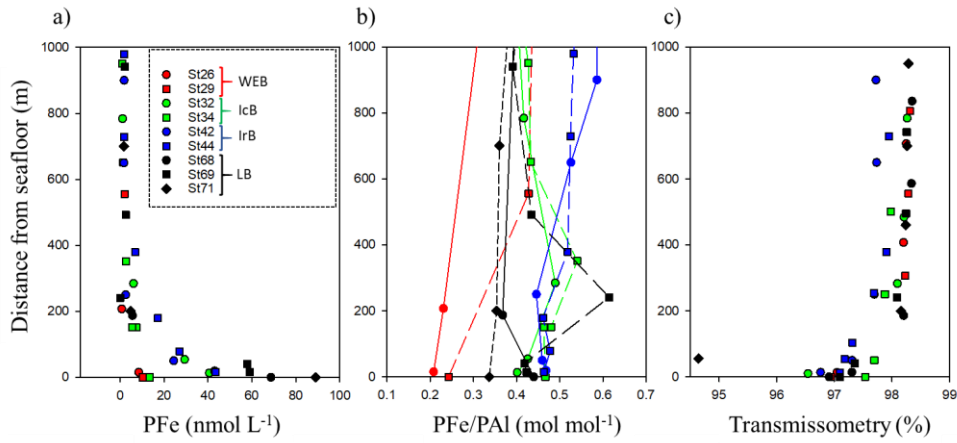
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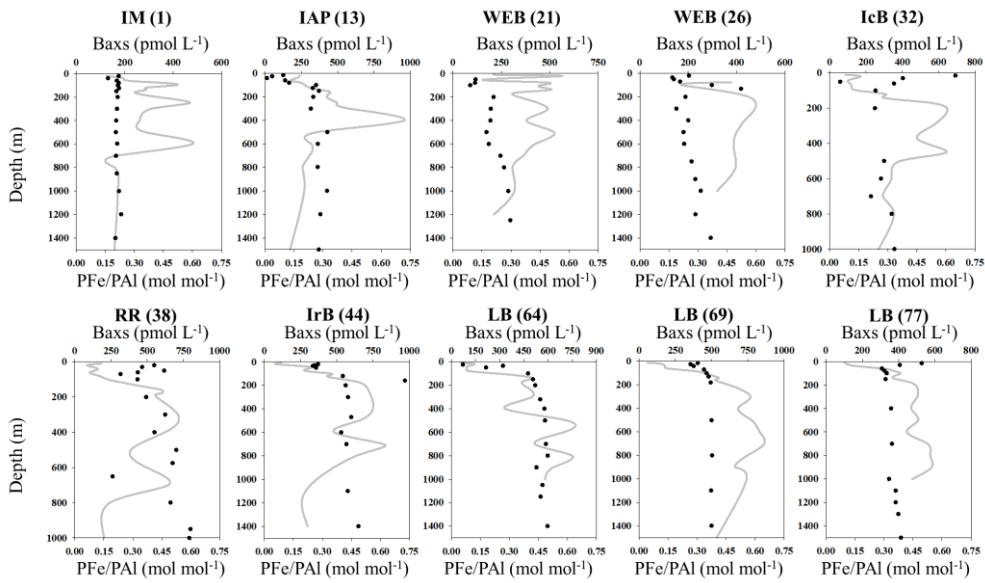
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1361 | Figure 112: PFe total (a); PFe/PAI ratio (b) and beam transmissometry (%) as a function of depth above the seafloor
 1362 (m) at selected stations where a decrease in transmissometry was recorded.



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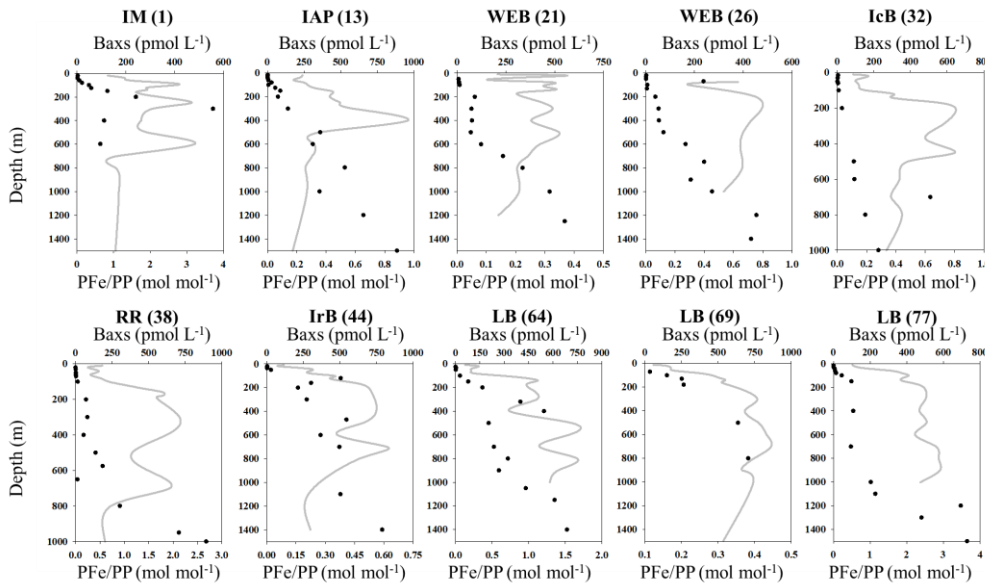
1377 | **Figure 13: Vertical profiles of Baxs (grey line, data from Lemaitre et al., 2018a) superimposed with PFe/PAI molar**
 1378 **ratios (black dots) at stations sampled in the Iberian Margin (IM), Iberian Abyssal Plain (IAP), Western European**
 1379 **Basin (WEB), Iceland Basin (IcB), above the Reykjanes Ridge (RR), Irminger Basin (IrB), and Labrador Basin (LB).**
 1380 **Note that Ba_{355} concentrations over the background level of 180 pmol L^{-1} are indicative of remineralisation processes**
 1381 **(Lemaitre et al., 2018a).**



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Figure 14: Vertical profiles of Baxs (grey line, data from Lemaitre et al., 2018a) superimposed with PFe_{bio}/PP molar ratios (black dots) at stations sampled in the Iberian Margin (IM), Iberian Abyssal Plain (IAP), Western European Basin (WEB), Iceland Basin (IcB), above the Reykjanes Ridge (RR), Irminger Basin (IrB), and Labrador Basin (LB). Note that $Baxs$ concentrations over the background level of 180 pmol L^{-1} are indicative of remineralisation processes (Lemaitre et al., 2018a).



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| | | Fe | Al | P | Mn |
|--|----------------|---------|---------|---------|----------|
| Blank (nmol L ⁻¹) | 5µm filter | 0.072 | 0.100 | 0.511 | 0.003 |
| | 0.45µm filter | 0.132 | 0.164 | 1.454 | 0.005 |
| Limit of detection (nmol L ⁻¹) | 5µm filter | 0.011 | 0.030 | 0.365 | 0.001 |
| | 0.45µm filter | 0.026 | 0.046 | 1.190 | 0.001 |
| Recovery CRM (%) | BCR-414 (n=10) | 88 ± 7 | | | 94 ± 7 |
| | MESS-4 (n=5) | 98 ± 14 | 97 ± 14 | 80 ± 30 | 110 ± 18 |
| | PACS-3 (n=8) | 101 ± 9 | 99 ± 14 | 91 ± 34 | 112 ± 11 |

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| % recovery | Al | P | Mn | Fe |
|-----------------------|---------|---------|----------|---------|
| BCR-414 (n=10) | - | - | 94 ± 7 | 88 ± 7 |
| MESS-4 (n=5) | 97 ± 14 | 80 ± 30 | 110 ± 18 | 98 ± 14 |
| PACS-3 (n=8) | 99 ± 14 | 91 ± 34 | 112 ± 11 | 101 ± 9 |

Table 1: Blank and limit of detection (nmol L⁻¹) of the two filters and Certified reference material (CRM) recoveries during GEOVIDE suspended particle digestion.

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

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

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