

Dear Editor, dear Referee,

We would like to thank you for your careful reading and useful comments on our paper "Particulate Rare Earth Elements behavior in the North Atlantic". Our manuscript has greatly improved.

We carefully addressed all comments from the Referees. Referee's comments are reported in black font, and our responses are in blue font. New and/or modified line numbers are also provided. The modified parts in this new version of our manuscript appear in blue font.

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

Best regards

The manuscript by Lagarde et al. presents a unique sampled section for particulate YREE. Data like this are really of great value and are needed to better understand the marine cycle of YREE. Although I appreciate the dataset itself, there are some issues in the manuscript that need to be addressed and clarified prior to acceptance.

We thank the referee for these useful comments.

Minor comments

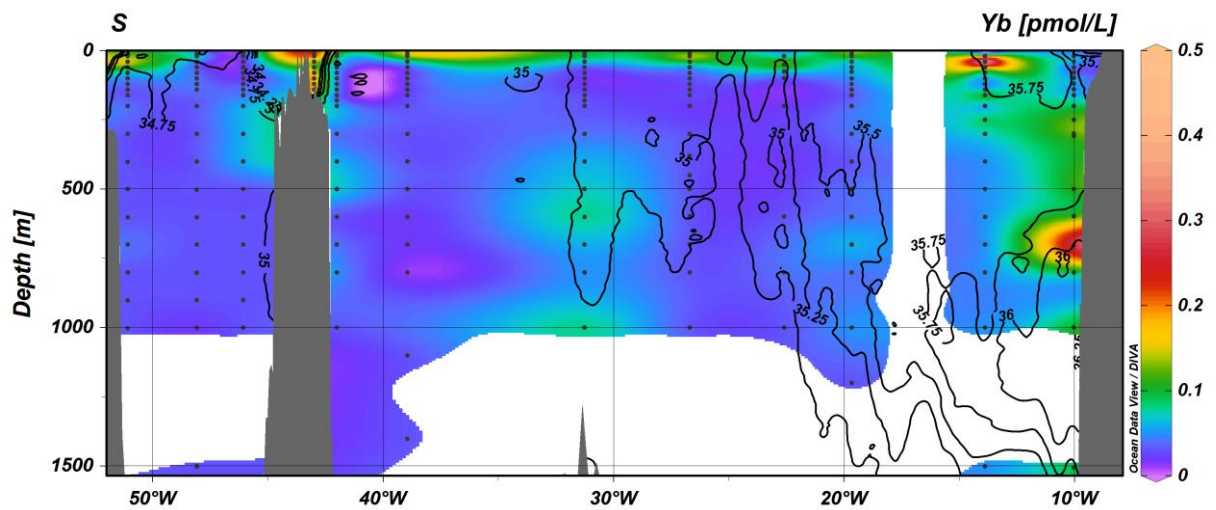
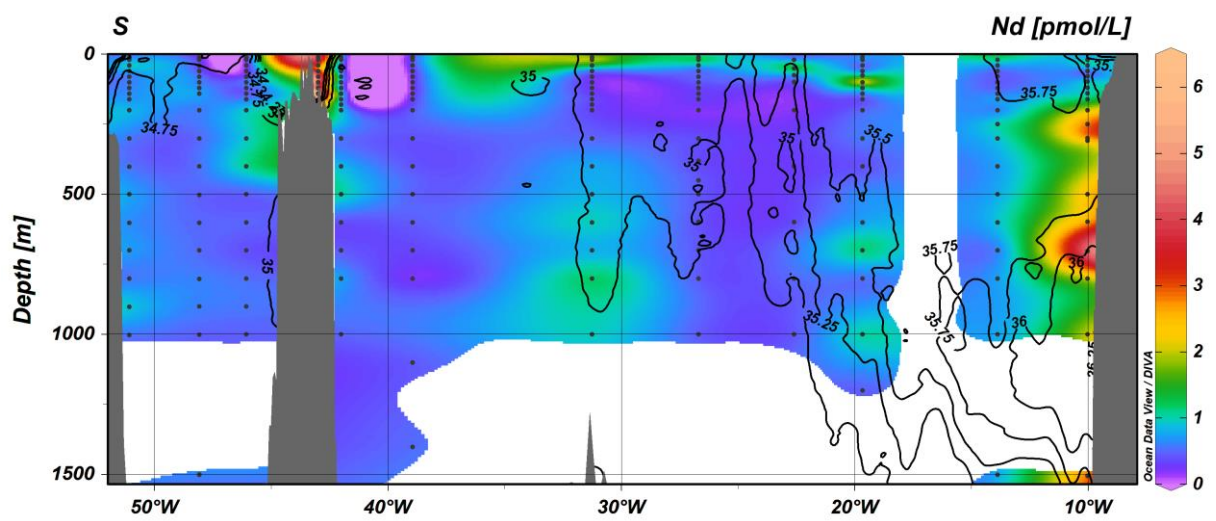
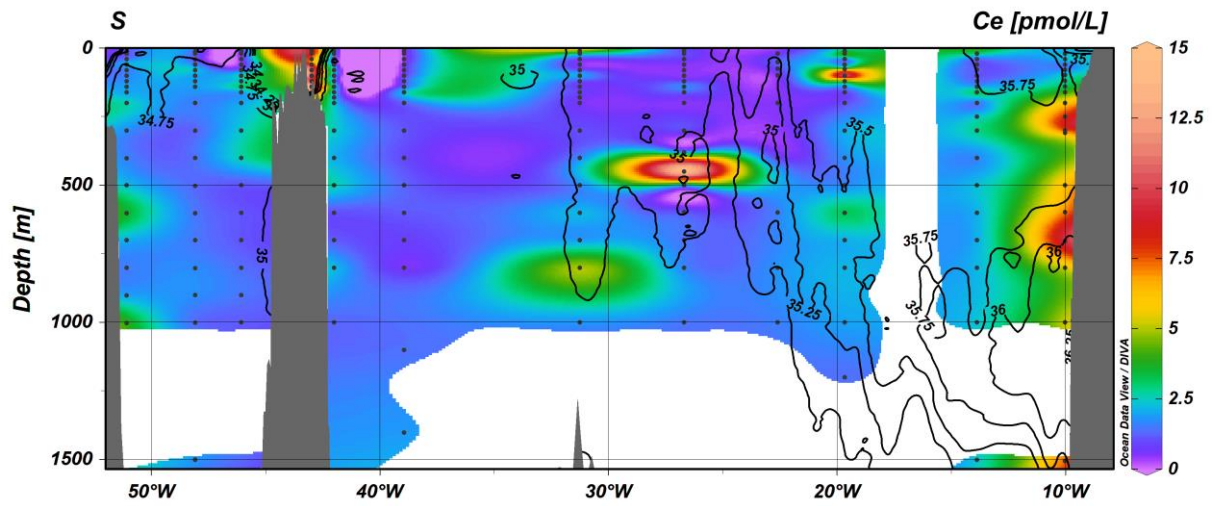
- There are quite a few orthographic and grammatical impurities. I suggest to avoid the use of 'being' and abbreviations in the beginning of sentences. The word 'one' is also often used inappropriately.

We carefully checked the orthograph and grammar.

- Figures should be reorganised and it is not always clear to which figure the captions relate. I would also suggest to make use of isolines instead of plotting 4 sections with 3 parameters. Draw salinity isolines and colour code the REE values, instead of plotting the profiles in the section. I would also strongly encourage to enlarge the upper 200m to show the features. This is the interesting depth range for most of your parameters. Also the interpolation is done differently when comparing Figure 6 and 2 A/B for which the former are without gaps while the latter section plots show white gaps.

We thank the reviewer for this comment.

We preferred to plot 4 sections with three parameters to display together the profiles and a description of the prevailing water masses using the salinity. We did try to add isolines but it rapidly came out that the figure was less readable (see examples below). The quality of the interpolations directly depends on the density of dataset. The depth and longitudinal resolutions for salinity and density are better than for PREE concentrations, so it is more rigorous to leave the gaps when PREE profiles are too distant from each other. The interpolation of Fig. 7A was revised to be the same as on Fig. 2B, 3C and 3D. We added a panel that enlarges the upper 200 m on Fig. 2B, 3C, 3D, 4, 7B and 8. For Fig. 6, the y axis is not the same because this figure shows a part of the section.



- the tables do not have units

Units were added in the table 2 and in the caption, line 821.

-some data, eg. Ac data, are not shown and not trackable by the reader, so discussions cannot be verified by the reader. For instance in lines 257-259 and 263-264.

Indeed. We are now referring to Emilie Le Roy's thesis, line 359. This work is available online.

- There are many places with missing citations. These are highlighted in the pdf.

Thank you. Citations were added when required, see revised MS.

- Parts of the discussion is found in figures in the supplement. It is awkward for the reader to keep having the supplement at hand to follow the discussion.

Figures were reorganized to prevent this. In particular, the section dealing with Ce anomalies was re-written and does not refer to the supplementary material anymore.

- The paper does not come to any conclusions is that intended in this format?

This is a mistake, we now provide a conclusion: see revised MS lines 516-547.

#### Detailed comments

L22: This needs to be formulated more clearly why this connection is drawn. Also, be consistent with PREE: When you use just REE or HREE without the 'P' it could be interpreted that you are not referring to particles.

Done, see revised abstract (lines 10-26).

L24 These 2 sentences appear out of place and attempt to state what has not been mentioned up to here. Please provide more information or leave them out.

We added more information, see revised abstract.

L29 what does this number refer to? Particle concentration, dust, algae?

It refers to particle concentration, it is now specified line 31.

L31 I know what you mean here, but this needs to be rephrased. Maybe start with "Elemental concentrations are..."

It was rephrased, see lines 36-37.

L39 Either formulate this as a hypothesis or provide a citation.

References were added line 36.

L41-49 This paragraph needs citations. There is not a single one

Citations were added, see lines 42-50.

L53 How does this quantification work then? What would be the approach using REE?

Precisions about the use of REEs were added lines 52 to 61.

L60 What does 'SP' then stand for. I assume Sub Polar - this should be added.

Yes, it is, it was added line 72.

L71 Please list them here.

The biogeochemical provinces are listed, see line 83-84.

L72 It is not clear how this diversity has been categorised. The figure does not really show the distribution of water masses. There are only labels and the reader is left to find the actual extent of the water masses.

The biogeochemical features of each region are developed in the paragraphs following L72 (line 86 in the revised manuscript). We choose to stay concise when describing features since they are described in details in Lemaitre et al. (2018b) and Longhurst, (1995). The labels represent the dominant water mass, it is specified in the manuscript lines 85, and the meaning of the labels was added to the caption and to the abbreviation table (lines 836-840). We do not provide information of the extension of the water masses because this is beyond the scope of a work discussing the PREE distributions. We are currently measuring the dissolved REE which will allow discussing dissolved and particulate data regarding the water masses.

L80 Please remind the reader where this region is geographically.

It is reminded at the beginning of the paragraph, line 93.

L101 This pool is not presented here (unfortunately).

It is a work in progress (see responses to Reviewer 1 as well).

L104 should be defined to be consistent

It is now defined lines 114 and 116.

L108 not shown in this figure.

It is now shown on the Fig. S4 in supplementary.

L110 be more precise. REE are trace metals too

Fe and Zn were given as examples, see line 181.

L113 the range given in the introduction is only 1 km wide.

This has been corrected, line 117-118.

L114 pressurised with what? Normal air? filtered air? Nitrogen gas?

It was normal air.

L115 Link? Not everybody is familiar with the GEOTRACES cookbook and its content.

The link was added line 119.

L118 was this buffered?

No, it was not buffered.

L121 Nevertheless, it should be briefly summarised here.

This section has been rewritten and clarified lines 130-136 and 188-191. We also provided a table in Fig. S4 to summarize the differences between the two sampling systems and analytical procedures performed on the filters. Results of this comparison are also provided graphically.

L126 Was the filter fully digested after this step?

Yes, it was fully digested. It was added line 142.

L141 Please give some more details

Details are provided in lines 164-165, and Fig. S2 was added in supplement to show the percentage of difference between replicates for each element analyzed.

L144 Were those values revised accordingly?

Yes, the blank was subtracted to the measured concentrations, it was added line 168.

L149 I do acknowledge that error propagation was not disregarded, however figure S2 does not provide any details, nor does the supplement.

The error propagation was reconsidered, and this part has been rewritten, see lines 169-176 and Fig. S3.

L150 what is the evidence of this 'apparent consistency'?

Particles seem to have a homogenous repartition on the filters to the naked eye. That said, we do not discuss more the hypothesis of homogeneity since we cannot quantify a difference between the two parts of a filter.

L154 It would be good to provide the reader with the main differences of the 2 methods.

This section was reworded for clarification, see lines 130-136, 188-191 and Fig S4.

L157 I do not know in which context the two studies are. What are the similarities? what are the differences?

See the rewritten section in lines 177-194 and Fig. S4.

L164 Do you really need to provide the isotope number of natural Th? Until now there are no other Th isotopes mentioned. You can state in the beginning that you will discuss  $^{232}\text{Th}$  which will be termed 'Th' throughout the text. Just a suggestion...

We prefer to keep it in, for sake of clarity.

L166 Please avoid abbreviations in the beginning of sentences.

An effort was made to delete abbreviations at the beginning of sentences.

L167 This is a bit over-simplified and lacks citations.

This short introduction to the specific behavior of Ce was completed, see lines 200-207.

L170 That is very ambiguous. Could you be more specific?

Precisions were added lines 209-210.

L174 You will need to increase the resolution of the pots to verify this as reader.

The upper 200 m were enlarged on Fig. 2B, 3C and 3D.

L176 terminology: the figure refers to sub-arctic front instead.

This is the sub-arctic front, it was revised, see line 218.

L179 provide depth range to remind the reader where this bottom is. Particularly, because you have change the range from the introduction

The depth ranges were added, see line 220.

L187 There are only PAAS normalised patterns for other PREE in the supplementary, no concentrations.

A reference to table 2 is now added in line 196. Normalized concentrations allow a comparison between concentrations. We show that they are higher close to the Iberian margin, so we decided to keep the reference to the patterns.

L197 remind the reader at which depth we are.

It is reminded, see line 237.

L208 I suggest to summarise these in a table and/or figure

It is summed in a table, added in Fig. S5.

Reference	Location	Sampling method	Nd (pmol L <sup>-1</sup> )	Ce (pmol L <sup>-1</sup> )	Yb (pmol L <sup>-1</sup> )
Kuss et al. (2001)	Along the 20°W meridian between 30°N and 60°N	Samples pumped and centrifuged from several m <sup>3</sup> of water at a depth of 7 m (n=24)	0.17 to 2.16 Average 0.67	0.2 to 4.9 Average 0.82	0.03 to 0.47 Average 0.13
Tachikawa et al. (1999)	Tropical northeastern Atlantic (20°N, 18-31°W)	<i>In-situ</i> pumps at 3 stations (an eutrophic (E), a mesotrophic (M) and an oligotrophic (O) sites), filtration of 30 to 995 L	E: 0.7 to 10.5 M: 0.3 to 2.6 O: 0.1 to 0.5	E: 2.5 to 24.6 M: 1.0 to 5.5 O: 0.4 to 1.1	E: 0.04 to 0.5 M: 0.02 to 0.09 O: 0.05 to 0.03
This study	Subpolar North Atlantic (40-60°N, 10-55°W)	Niskin bottles	0.1 to 6.1	0.2 to 16.3	0.01 to 0.50

L213 This is a bit confusing to me. Your transect within this region is rather meridional. Do you mean the southern part by 'to the east'? Please clarify. Also while checking this in your table, I noticed that you did not provide any units.

South east would be more exact, it is specified line 258. Units are provided in Table 2.

L214 This is not a discussion. This is just a comparison of 2 different studies in two different regions. Tachikawa's study is more than 20 degrees further south than your southernmost station. If you want to compare the studies, you have to be more thorough in explaining the differences from your study. As it is now, it reads as if these study areas are very close to each other actually comparable.

It seemed interesting to us to compare our results to this study because it described PREE behavior in three very different contexts, with a station located in an eutrophic area, another one in an mesotrophic area and the last one at an oligotrophic site. These authors highlighted differences in PREE distributions and Ce anomaly profiles between the three sites, revealing different processes depending on the type of the station. It provides a wide range of concentrations to compare our data. Our stations can be compared to eutrophic/mesotrophic/oligotrophic types, and we discuss our results in the light of these sites, especially in the part about the Ce anomaly, because comparable features are observed. Finally, to our knowledge, even located 20° south of our most southernmost station, this study is the closest study dealing with PREE in the literature. For all these reasons, it seemed important to us to provide details about the study at the beginning of the discussion.

L230 Can you explain the reader, why this fractionation might occur? You missed to explain the reader a bit on REE chemistry in the introduction (e.g free metal vs stable complex)

Information about REE chemistry in seawater were added in the introduction, lines 52-60.

L233 Which phases do you mean. Please repeat, because you have introduced a couple of more phases in lines 230 and 231.

It was repeated, see line 282.

L245 it's not yet percent unless you multiply by 100. Like this, I would term it 'fraction'

It was multiplied by 100, see line 297.

L246 you cannot subtract concentrations in different pools from each other. You have to factor them by the lithogenic fraction. In this case  $[REE]_{authi} = ([REE]_{total} - F \times [REE]) / (1-F)$ . Or the other way around the  $[REE]_{total} = F \times [REE]_{lith} + (1-F) \times [REE]_{authi}$

It was revised, see lines 295-297.

L250 not correct - see comment on equation (3)

It was revised, see line 300.

L252 What does this mean. You only chose LREE without Ce? This is OK, but (1) explain why and (2) you don't need to write '...on the one hand ...on the other hand...'

We choose not to take Ce into account because of its distinctive behavior. It was added line 303-304, and '...on the one hand ...on the other hand...' was deleted.

L257 by how much is this excess? It is not visible in the figure.

This excess is of a few per cent most of the time, and can be up to 550 % at the surface of station 1. The excess of a few per cent is within the uncertainties, when higher excess suggests authigenic Th. We developed the discussion about it lines 308-314.

L261 this is correct and you need to point out - assuming you talk about concentrations in the water column - that you can only compare the pattern, but not the absolute numbers.

Yes, only patterns were compared, not absolute concentrations.

L262 They're flat because you determined your lithogenic REEs via UCC, which is pretty much PAAS in the distribution pattern.

This is right, that part was deleted. Instead, we used the total flat patterns of station #1 and #13 to assess the validity of the UCC as a lithogenic source (lines 325).

L263 where do you show that?

It is not shown here as it was done by Rudnick and Gao (2014).

L265 Of course, as you compare dust from a specific region with your data I would not expect matching patterns.

Also if you would have determined your lithogenic REE via dust normalisation the pattern would be flat too. I think this is a circular argument.

We deleted this argument and replaced it : we use the UCC as a lithogenic reference because the GEOVIDE are is surrounded by shields and extended crust, and dust inputs were not significant during the GEOVIDE cruise (Shelley et al., 2017). See lines 325-328.

L270 Will you discuss about this?

Details were added lines 319-322

L276 Which maxima do you refer to? You talked about little LREE enrichment, but not about maxima (of what?).

It was specified line 336.

L278 bracketing and where were these samples taken from? The same region? What the values?

Samples were collected during the same cruise with the clean sampling system (samples used for the comparison in 2.3.2). For PFe, the values were also 100%. For PMn they were about 40% between 200 m and 250 m, and ranged between 60% and 75 % from 500 m to 1000 m.

L281 You do not have the data to prove that. station 53 is above the shelf. Station 51 to the east - still close to the continent - does not show this enrichment. To the west station 64 is already far away from the shelf. Or what do you mean by eastern end of the section?

This is a comparison with the intermediate nepheloid layers observed along the slope at station #1, at the south east of the section. It was reworded lines 358.

L291 where are these fractionated patterns to find and which 'other processes are at play'?

These are the patterns of station #77 presented in Fig. 5, and the other processes are discussed in the following parts. Fractionation by diatoms and precipitation of Fe and Mn hydroxides were added here as examples, lines 352-353.



L293 This paragraph jumps geographic regions and compares them amongst each other but differences are not discussed at all.

This paragraph was deleted as it was not helpful for the discussion.

L295 I'm confused here. Why do you have flat patterns in figure S3 and fractionated patterns in fig. 5?

Patterns were flat in Fig. S3 because they represent the lithogenic fraction, the patterns for total PREE were represented on Fig. 5.

L306 It is rather consistent with the high lithogenic fraction, but not really visible in PREE data.

This has been corrected, see line 362.

L310 You talk about data that the reader has no access to - therefore difficult to verify

See answer to minor comment 4.

L325 This section is very vague and not convincing. I am not sure what the aim of this discussion is.

L335 was it?

L338 You should show that and convince the reader that there is a relationship

L349 where do you have the residence time from?

L352 This is basically a discussion about supplementary material. Why is this not shown in the main paper?

For the five previous comments: the section was completely rewritten, see lines 379-441.

L360 there are a few statements in this paragraph without back up of citations.

Citations were added, see lines 442-483.

L647 Suggestion: Combine pMe and sal in one section plot using isolines for one parameter. You can still show your stations by slightly increasing the sample dots. I would also strongly encourage to zoom into the upper 250m where you have the highest variation in pMe.

See the answer to the second minor comment.

L661 Same here: Please zoom into the dynamic upper layer

The upper 200 m were enlarged on Fig. 2B, Fig.3C and D, Fig. 7B and Fig. 8.