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 #1’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.

In this manuscript, Lagarde and colleagues present a substantial dataset for suspended particulate REEs and Y from a 2014 cruise in the NE Atlantic. The authors are commended for publishing these data, as there are too few high-quality REE data sets for ocean particles, and the interpretations have the potential to greatly increase the understanding of biogeochemical processes in general, and the ocean chemistry of REEs in particular, relative to inferences from dissolved data alone. One liability to keep in mind is that the residence time of particulate phases is generally much shorter than that of the dissolved pool, so these kinds of datasets are much more like short-term “snapshots” of distributions, especially in the upper water column, than long-term averages of regional distributions. This difference might be worth pointing out explicitly in this paper.

We thank the reviewer for his/her detailed review and valuable comments. Regarding the “short-term snapshot” of the reported distributions of PREEs, we are now emphasizing it in the abstract and in the conclusion, lines 12-13 and 519-520.

Overall, the paper is well organized, and the figures are appropriate. This constitutes a substantive addition to the long history of effort to understand the processes affecting the distribution of REEs in the ocean, via inferences from (mostly dissolved) oceanic distributions, combined with laboratory studies that are not the focus of the current paper.

We thank the reviewer for this comment.

I do have a fairly long list of comments and criticisms, that in sum probably suggest major revision. I list these below, in order through the manuscript. Fortunately, only a couple of the comments refer to what I deem to be serious misinterpretations of the data. These are comments #25 and 26 below (marked with *).

Thank you for the careful reading of our paper and these comments. We answered to each one of them and reported the lines modified in the manuscript after your comments.

A substantial fraction of the other comments are related to clear wording, and confusions caused by vague or inaccurate use of words and phrases. These are very important, because they are relevant to the specific processes being discussed. I have also commented extensively through the first part of the manuscript (see attached marked-up pdf, looking carefully for the small Adobe editing marks), with numerous revisions of the English word use, syntax, or grammar. However, at some point I felt that there were too many editorial needs in the use of English, and I refrained from most revisions in the interest of time, simply marking the problematic spots in yellow highlight. I leave it to the more experienced authors with good command of English (e.g. Planquette) to spend a few hours carefully improving the writing to avoid incorrect word usage and awkwardness, with the goal of making this paper much more readable. The problems with the highlighted sections should be quite obvious. One tip is that the frequent use of “one” or “ones” in a comparative sentence almost always makes for awkward English.

Following this comment, we spent a lot of time rewording the manuscript.

Once the revisions to the main body of text are completed, the authors should re-consider the content of the Abstract. As it stands, it makes the point about the long-distance transport of INLs, which I argue below is not well substantiated, and it also ends on an unsatisfyingly uncertain note with regard to Ho/Y ratio observations. I suggest deleting this last part (and perhaps the final section of the paper – see below), to end the Abstract on a stronger note.

The patterns and the Ce anomaly clearly point out to a dominance of REE absorption processes in the first meters in contrast with scavenging processes at deeper depths. The abstract rephrased.

1. Line 100: Methods: Were Ba and Th-232 measured on samples taken from both kinds of sampling bottles? Were conventional bottles measured by one lab and GO-FLO samples by another lab? Later it says Y was used to compare the two procedures (but also the collection method?). Not clear starting at line 100. This should also be made clear in Table 2. The authors should state what exactly is being reported here vs. related results from the same cruise that are reported in other publications. I found this description of sample types and who measured what to be confusing.

Particulate Ba and ^{232}Th concentrations were first determined in particles collected with Niskin bottles at the Royal Museum for Central Africa, Tervuren, Belgium then at LEGOS, Toulouse, France on the remaining leaching solutions. Ba and Y concentrations were also determined in particles collected with Go-Flo bottles at LEMAR, Brest, France. The differences between the two methods and the results of the lab intercalibrations are provided in the Supplementary material (Fig. S4). The section was revised to make it clearer (lines 113-196).

2. Line 117: Samples were rinsed with MQ water. There is possible loss of adsorbed elements as water should have been pH 5.6 if equilibrated with air. Previous workers have used NH4OH to adjust pH of rinse water to~8-9 to avoid this potential loss of adsorbed elements. Can the authors argue that results were not biased by loss of material from the particulate samples?

As the reviewer pointed out, there is indeed possible desorption (or even adsorption) of elements when rinsing with Milli-Q water or any solution actually. However, this desorption process is element- and filter- dependent.

Concerning Y, we were able to perform a comparison between concentrations measured on samples collected with Niskin bottles and with Go-Flo bottles. Samples collected with GO-FLO bottles were not rinsed (see Gourain et al., 2019), the excess water being drawn off with a syringe. The agreement is very good (see answer to comments n°4 for more details).
Furthermore, a previous study conducted at LEGOS (Arraes-Mescoff et al., 2001) investigated the dissolution of REE and Th following the incubation of large particles filtered from seawater during 24 hours. Results showed that after 24h no dissolved REE could be measured (i.e. below detection limit) (their Fig. 2) and a slight increase in particulate ^{232}Th concentrations after 12h (their Table 3). They also showed that these concentrations did not exceed 0.4 ppb after 10 days, which remained less than 0.5% of the initial PREE concentrations (their Table 4).
The rinsing time during GEOVIDE was very short, and a volume of less than 5 mL was used (Lemaitre et al., 2018b), so the material loss is supposed to be negligible.

3. Line 127: Analytical Methodology: The text implies that 2.0mL from a 3.0mL total digest solution was used for REE, Y, Ba and Th analyses. But what was the dilution of this solution for analysis? Was HF included in this solution, and was an HF-compatible ICP-MS introduction system used?

The leaching solution was not diluted for most of the samples, only a few samples were diluted with HNO_3 0.32 mol L^{-1} (prepared from Merck nitric acid 65%, EMSURE® distilled again at LEGOS to get the purest product, regularly controlled) by a factor between 1.3 and 1.5, because the archive solution volume was too small to allow for ICPMS analysis that requires at least 2mL. HF was not included in this solution, therefore no specific introduction system was required. See lines 142-144 in the manuscript for details.

Were standard curve solutions match to the acid mixture and concentrations in the (diluted?) samples?

Yes, standards were prepared by dilution of a stock solution in 0.32 mol L^{-1} HNO_3 with ca. 0.1 ppb of In and of Re. This information is now included in lines 159-161.

What was the % correction for oxides for each of the REEs, especially those generated by Ba and the LREE?

Major interferences of Ba oxides and hydroxides affect the Eu and Gd isotope masses. BaO interferences represented a maximum of 0.4% and of 0.3% of the signals of the measured Eu and Gd isotopes respectively and occasionally reached 10% for Eu for seven samples. For the other REEs, oxides contributed to less than 0.1% of the signal. Hydroxide interferences are one order of magnitude less than oxide's interferences. This information is now provided in lines 155-156.

How were isobaric interferences avoided;

In low resolution, isobaric interferences were corrected by the software of the ICP-MS (Method Editor, Thermo Fischer Scientific), using another mass of the same element (not interfered, 161Dy in the example below) to calculate the number of counts that are interfering the desired measurement. For example, for interferences of ^{161}Dy on ^{158}Gd, the correction is:

Counts(^{158}Gd) =counts(mass158)-counts(mass161) x abundance(^{158}Gd)/abundance(^{161}Dy)

They are listed together with the element interfered in the answer of the next comment. Note that the isotopes that we analyzed were selected to minimize these interferences. All the equations used for these corrections were checked in the method of the instrument before analyses, and that it is possible to custom the isotope used for correction.

it may be useful to list in a table the isotopes analyzed.

Done. Listed below are the isotopes analyzed. We did not consider relevant to add them in the main text; however, if the editor and referee wish, we could add them as supplementary material.

Y89
In115 (Sn115)
Sn118 (only used to correct In115 from Sn115 contribution)
Ba137
La139
Ce140
Pr141

Nd143
Nd146
Sm152 (Gd152)
Eu151
Eu153
Gd158 (Dy158)
Tb159
Dy162 (Er162)
Dy163
Ho165
Er166
Er167
Tm169
Yb172
Lu175
Re185
Th232
Dy161 (only used to correct Gd158 from Dy158 contribution)

What mass resolution settings on the HR-ICPMS were used for the various analytes?

All measurements were performed in low resolution mode (see line 151 of the corrected MS)

The reported 20-30% uncertainty in final PREE concentrations seems very high. What was the largest contributor to the uncertainty? Fig. S2A implies that the largest source of error was cutting the filter exactly in half (or sample heterogeneity on the filter surface). The highly variable and relatively large error associated with the measurement (Fig. S2A) is odd because cps should have been quite high given the sample volume and final digest volume, unless the primary digest solution was over-diluted (if so, why?).

Indeed, assuming a homogeneous filter loading, the largest contribution to the uncertainty is cutting the filter exactly in half, as shown in Fig. S2A. The different contributions are summed in the following table that replaced the Tab. S2 in the manuscript as Fig. S3.

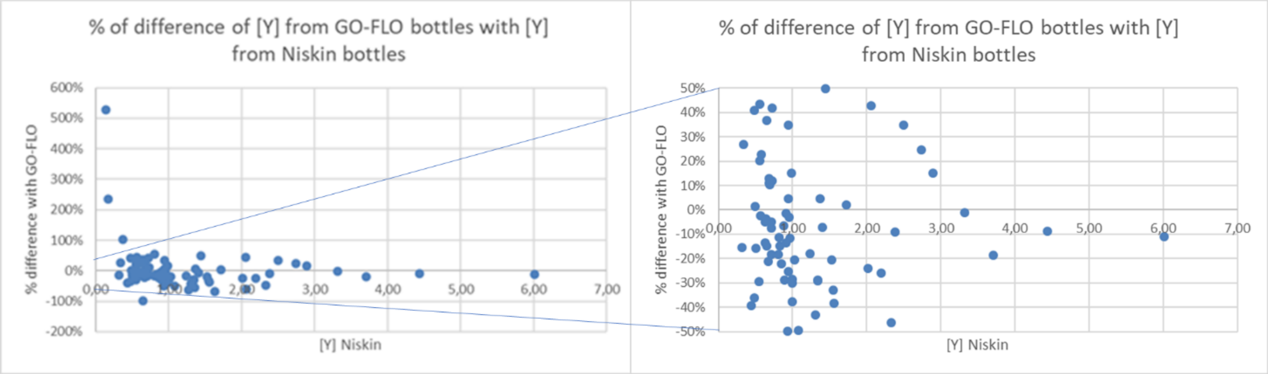
Source of error	Determination	Mean % of the concentration
Volume of leachate	2sd calculated on the weight of all archive volume after 13 mL of HNO_3 0.32 M were added	0.6%
Volume taken for ICP-MS analysis	Average 2sd calculated on weighted replicates for a sample	0.005%
ICP-MS measurement	2sd calculated on 5 spectra measured for a sample	3.3%

Reading between the lines, one could guess that a dry-down step was not desired, so digests may have been diluted to acceptable acid concentrations for the ICP-MS introduction, leading to low counts per second and high uncertainties based on signal counting statistics. Alternatively, filter blanks may have been high and/or variable (this is not mentioned – what was the range of % filter blank?). This needs much more explanation, because 20-30% uncertainty is very high, and is be-lied by the relative smoothness of the profiles shown Fig. 3 for example. This stated measurement uncertainty should have yielded noticeably “bouncy” vertical profiles. I think the data are not as uncertain as the authors' assessment, which may be more theoretical than empirical, since true sample replication was not practical.

There was a dry-down step and only few samples were diluted when there was not enough leaching solution left (see lines 145-146 in corrected MS). The chemical blanks represented 0.01% to 5% of the measured concentrations, rarely reaching 30% for Y, Lu and Th (lines 166-168 in corrected MS). Filter blank was determined by leaching an unused clean filter following the same protocol as for the samples. Taking your comment into account, we choose to not consider the contribution of the uncertainty on the fraction of filter analyzed to the final concentration error. The data set and the profiles are corrected. We provide additional information in lines 169-176.

4. Line 155: To compare concentrations between the two analytical labs, the regression slope is only partially helpful. Please give the mean % difference for all samples, and indicate if this % shows any trend with sample concentration (e.g. higher concentrations agree better?).

Following the reviewer's comment, we provide below the mean % difference for all samples:



The figures above (twice the same plots, different scales) show that the highest concentrations agree better. Lowest concentrations of Y show the largest difference between the two labs. The median percentage of difference is 21%, for concentrations ranging between 0.3 pmol L^{-1} and 6 pmol L^{-1}. Four samples were excluded between the first and the second graph: one sample collected with a GO-FLO bottle (station #32 at 300 m) which had a concentration below 0.01 pmol L^{-1} and showed a difference of -100% with the sample taken at the same station at the same depth with a Niskin bottle. Three samples collected with Niskin bottles had significantly lower concentrations than samples from the GO-FLO bottles, below<0.3 pmol L^{-1}: at station #1 at 60m and at station #64 at 500 m and 900 m. These differences suggest an unidentified bias during the sampling and/or analytical protocols.

5. Line 159: Why did Y agree much better between the two sampling systems (and labs?) than Ba, which has much higher particulate concentrations? Was this related to the filter type used for each sampling effort, or differences in the digestion methods used by the two labs? This comparison is again quite unclear. If this cannot be explained in simple terms, please put all the information in a table, with columns of collection bottle type, filter used, digest method, analytical method, lab where analyses were made, and final results from each lab, etc.

A table is now provided in the supplementary material (Fig S4) and recaps the sampling systems, digestion procedures and intercomparison of measured concentrations.
Lemaitre et al. (2018a) explained the higher Ba concentrations measured in samples collected with Niskin bottles than in samples collected by GO-FLO bottles by the different filter types and the chemistry used. Samples collected with Niskin bottles were collected on 0.4 μm polycarbonate filters, while samples collected with GO-FLO bottles were collected on paired 0.45 μm polyethersulfone and 5 μm mixed ester cellulose filters. Different filters can lead to

different concentrations even when the chemistry is the same (Planquette and Sherrell, 2012). Furthermore, a more concentrated HF solution was used for the chemistry on polycarbonate filters.

Although impossible to prove at this stage, it is possible that Y is less sensitive than Ba to the filter material, and/or to HF.

6. Line 167: "Ce oxidation onto particles" suggests a poor understanding of whether pCe is dominated by adsorbed Ce4+ or by an independent phase e.g. CeO2. If Ce forms or forms within an independent oxidized authigenic mineral, then "prevents ad-sorption" is not the right phrase. Please clarify and expand this explanation of the unique behavior of Ce.

We carefully reworded this section. See lines 200-207:

The specific behavior of Ce is due to the occurrence of its IV oxidation state in addition to the III oxidation state common to all the REE. Two mechanisms for Ce oxidation have been proposed so far: a microbially mediated oxidation in seawater under oxic conditions that leads to formation of insoluble CeO_2, more particle reactive than Ce(III) (Byrne and Kim, 1990; Elderfield, 1988; Moffett, 1990, 1994; Sholkovitz and Schneider, 1991) and an oxidative scavenging onto Mn oxides particles (De Carlo et al., 1997; Koeppenkastrop and De Carlo, 1992). These two processes act in addition to the general scavenging process that affects all the trivalent REE by surface complexation, thus leading to the Ce enrichment in particles and its stronger depletion in the dissolved phase compared to other REE.

7. Line 171: This section describing pCe distributions is very hard to follow because no figures are referred to. Also the terms epipelagic and mesopelagic need to be re-defined by depth intervals as a reminder to the reader, so that "bottom of the epipelagic" can be understood relative to the depth scale of Figure 2.

We referred to Fig 2 at the beginning of the section (line 212), and added more references to each figure (PCe profiles on Fig. 2A and PCe section on figure 2B). Epipelagic refers to the depth range of 0-200 m while mesopelagic refers to the depth range of 200-1500 m (lines 117-118 and 220).

8. Line 184: The Station 44 maxima at 120m and 160m are defined by only one point each, so I think it is quite possible that they are uniquely contaminated with Ce, unless the contextual data can provide a clue as to a possible source in this region at those depths. See further related comment on Ce anomalies below.

At Station 44, PCe concentrations are 1.8 and 3.3. pmol L^{-1} at 120 m and 160 m, respectively. While higher compared to the concentrations above and below, they remain in the range of PCe concentrations measured along the section, and similar maxima are observed at station #32 at 140 m and at station #38 at 160 m. We investigated a possible carry-over contamination from the previous sample which cannot be excluded even if the Perspex systems were carefully rinsed between each sample. That said, station #38, which preceded Station #44 was not especially rich in PCe compared to other PREEs. Finally, when there is contamination with PCe other REEs are usually affected, like La for example. Looking at our dataset, a specific contamination in Ce seems very unlikely.

9. Line 200: Section 3.4 has a big problem because the heading says Nd/Yb but Fig.4 shows Yb/Nd, the inverse. One of these headings is wrong, and I suspect it is the section heading. I would expect that near-surface particles have a greater biogenic component and a smaller crustal component, so that Yb/Nd will be higher, reflecting the LREE-depleted seawater source for the particulate uptake, which is only partially compensated by preferential LREE removal by biological particle production, and by particle scavenging in general. In other words, I would expect PAAS-normalized REE patterns for particles in the euphotic zone to be "seawater-like",

but somewhat less LREE-depleted. Thus my guess is that Fig. 4 is correct, and the text throughout section 3.4 is wrong. Please correct this. The following two comments should be taken in this light.

Thank you for this comment. Indeed, the section heading was wrong. We corrected this mistake by harmonizing the use of Yb_N/Nd_N in the text and the figure (lines 239-250).

10. Line 202: I don't see any values of Yb/Nd of 0.01 in Fig. 4. Visually, it looks like the minimum value is about 0.2.

It has been corrected, see line 244.

11. Line 204: It is stated that the highest Yb/Nd value is in the epipelagic of Sta. 21, but Figure 4 shows a single subsurface value three times as high in the epipelagic of Sta.13. This needs to be corrected. The last sentence of this paragraph says that at this relative high Yb/Nd point at Sta. 21, four of the LREE are also at high concentration. This sounds odd, because high LREE would be expected to drive HREE/LREE, and thus Yb/Nd, to low ratios, not high. If this sentence is highlighting a surprising result (high HREE/LREE at high [LREE]) then this should be pointed out. I can see from Fig.2A that PCe has a single point maximum – is that single point the one that generates the high Yb/Nd?

The data point at station #13 at 40 m was categorized as an outlier at line 244. The last sentence of the paragraph is about the minimum at 100 m, it was specified line 248 to avoid confusion.

12. Line 231: "react preferentially with biogenic phases". Can the authors be more specific? Are they suggesting that Yb would be adsorbed preferentially to Nd on POM? On bSi? This phrase is too vague. Best to be more specific about the phase invoked, and to cite references appropriately.

This assumption is based on the work of Akagi et al. (2013), and is now detailed in lines 278-280 in the corrected manuscript:

In the Bering Strait, Akagi et al (2011) also observed a strong association between particulate HREE and biogenic silica collected in sediment traps. This specific BSi control on HREE behavior is discussed in section 4.6.

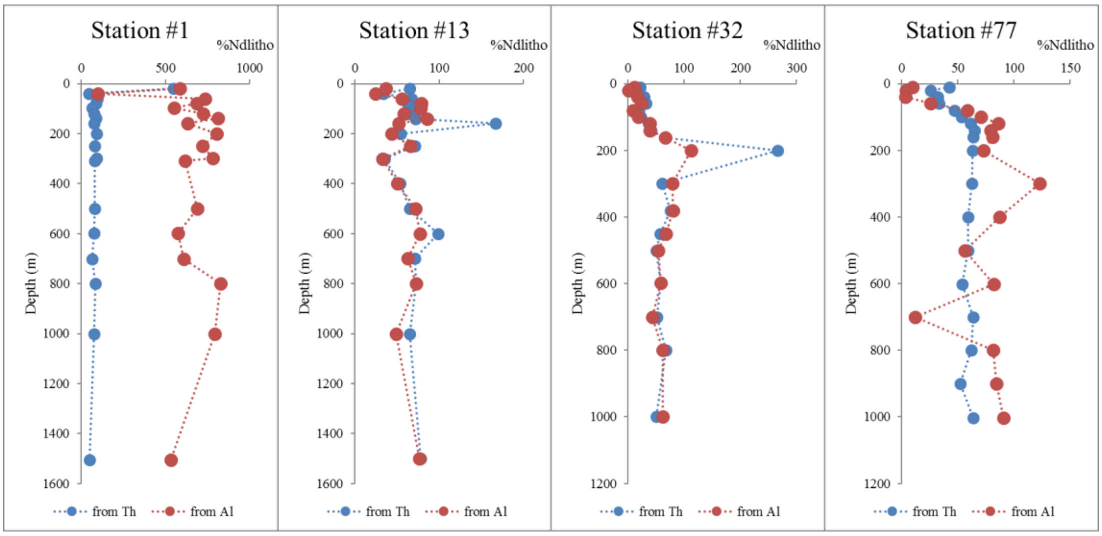
13. Line 241: This is the first place in the manuscript where I finally understood that the samples being discussed were collected in the standard Niskin bottles. This should be abundantly clear in the Methods. See comment above.

See answer to comment n°1, we provided more details in lines 113-196 and summarized the different sampling systems, chemistries on filters with the associated measured element in Fig. S4. We hope it is now clearer.

14. Line 244: Rock types in the crust are likely more variable in Th content than in Al content. A rough estimation of the uncertainty in the %Lithogenic fraction calculated in this manner should be presented. Are the uncertainties large enough that the %Lithogenic should be viewed only as a relative scale?

We did not want to use Al as lithogenic tracer because it was demonstrated that it could be incorporated in the biogenic silica. In addition, i) ^{232}Th is less soluble than Al, as shown by its shorter residence time (25-55 years (Roy-Barman et al., 2019) versus 200 years (Hayes et al., 2018)); ii) PAl was not measured concomitantly to our samples, while ^{232}Th was; iii) ^{232}Th is less prone to contamination than Al. Moreover, previous observations allowed us to assume that ^{232}Th was relatively homogeneous in rocks and sediments: 1) Chase et al. (2001) showed that ^{232}Th concentration in lithogenic sediments sampled in the South Atlantic ocean was constant at around a value of 10 ppm 2) This value is close to the median concentration of 10,5 ppm of the upper crust reported in Rudnick and Gao, 2014 and used in this study 3) the GEOVIDE area is surrounded by shields and extended crust in majority of Caledonian fields (https://earthquake.usgs.gov/data/crust/type.html), with a relatively homogeneous geochemistry (Cocks and Torsvik, 2006; Rudnick and Gao, 2014). For all these reasons, we considered that ^{232}Th was a reliable tracer of the lithogenic fraction.

However, we also estimated the lithogenic contribution calculation using PAl concentrations measured in samples from the clean rosette and obtained a good agreement for most of the samples and stations. Discrepancies are discussed below.



.

The error calculated on the lithogenic fraction varies from 0.7% to 6% when the error on the Th concentration only (3.3% on average) is considered. Propagating the error of ±0.5 ppm on Th concentrations in the upper crust increases this error to an average of 5.9% (ranging from 4.8% to 10.4%). Thus, overestimated lithogenic fractions below 106% are falling within this uncertainty (ie station #38 at 160 m). When the estimated lithogenic fraction was higher than 100%, we attributed it a value of 100%. Comparing with the lithogenic proportion calculated with Al shows that sometimes the use of Th overestimates the lithogenic fraction (for example at station #13 at 160m) but the calculation with Al also indicates a maximum at the same depth. At station #32 at 200 m, Al data are also significantly lower than Th data (113 % vs 200%), and allow us to assess a value of 100 % for the lithogenic fraction.

At stations #1 at 20 m, Th has likely been scavenged, and is present in authigenic fraction. Such “rapid Th scavenging” has already been reported by Hayes et al. (2015) in particles and by Robinson et al. (2008) in sediments. Hayes et al. (2015) proposed a correction using the partition coefficient of Th that is assumed to be the same for ^{232}Th and ^{230}Th, but we do not have data to do it, so we set the values to 100%. At the surface of station #77, where a diatom bloom occurred during the cruise, the lithogenic fraction calculated from Al is lower than the one calculated from ^{232}Th (10% for Al and 40% for ^{232}Th), suggesting an authigenic source of Al.

To conclude, even if the used of ^{232}Th as a lithogenic tracer sometimes include a bias that can lead to an overestimation, it remains a better lithogenic tracer than Al for our data set. We added this discussion in lines 307-314 in the corrected MS.

15. Line 258: A finding of >100% lithogenic fraction using the Th-232 method suggests that ALL estimates of lithogenic fraction may be overestimates, and are at least probably not underestimates. This should be acknowledged in the text as a potential unidirectional bias in % lithogenic fraction.

We acknowledged it line 311.

16. Line 262: The patterns of PAAS normalized lithogenic fractions are called “flat” but the plots in Fig. S3 are on a log scale. This hides the fact that most of the patterns are MREE-enriched. Could the authors comment on this observation? I encourage plotting REE patterns on a linear scale whenever possible; this highlights the quality of the data and inter-element pattern details more clearly.

We agree with the reviewer regarding the MREE enrichment. Associated Gd and Eu anomalies were calculated. However, we decided to not present them in the manuscript because the associated errors prevent clear interpretation of the anomaly profiles. In consequence, we kept the “classical” pattern representation with a log scale, as used in the literature in the manuscript, and added the patterns with a linear scale in Fig. S7 B to the patterns represented with a logarithmic scale (Fig. S7 A in corrected supplementary).

17. Line 276: The enrichment in LREEs is interesting, and this enrichment appears larger where the absolute pREE concentration is higher (greater depths). The authors imply that this is because of the “lower solubility” of the LREE relative to the HREE, and that this depth difference in LREE enrichment is caused by the adsorbed fraction. Yet the % lithogenic increases with depth as well. Can the authors eliminate the possibility that the LREE-enrichment is a function of the difference in REE composition of source rocks (or the fraction of source rocks that survives chemical weathering) and PAAS? Also, as noted in comments on the pdf, “solubility” is not the right term to use to describe the LREE, if the process being referred to is adsorptive scavenging, not the solubility of a unique solid phase. Admittedly, this chemically inaccurate language is used often in the marine chemistry community.

We interpreted the increase of the lithogenic contribution with depth as an effect of remineralization, leading to a loss of authigenic material and by consequence an increase of the lithogenic fraction. As the preferential scavenging of LREEs relatively to HREE is a well-documented behavior in seawater, we assumed that the LREE enrichment observed in the particles is the symmetric of what happened in the dissolved pool of LREE (Garcia-Solsona et al., 2014; Tachikawa et al., 1999a).

18. Line 276: “these maxima”. It is not clear here that you are referring now to the Iberian margin maxima in % lithogenic fraction. This is because the start of the paragraph refers to BOTH margins. Please add words to make it clear that you are shifting your focus to the Iberian margin here. And the Fig. 6 caption needs to explain the white arrows in the figure.

We clarified this point in the text (line 335) and changed the Fig. 6 caption to provide information about the white arrow.

I think they are supposed to show a density similarity between relative maxima in the profiles from the various stations, but for Sta. 26, for example, the arrow from Sta. 21 does not point to a relative maximum; the maximum is one depth lower at 200m.
Similarly, the lower arrow pointing from Sta. 26 to Sta.32 shows the % lithogenic increasing from _60% (not a relative max) to _100% at Sta. 32. I don’t see how the lithogenic fraction could increase unless biogenic particles are preferentially lost to sinking (not likely) or another source of lithogenic particles exists at Sta. 32. I suspect that these differences are all related to the uncertainties inherent in the Th-232 normalization. The authors need to do more work to justify their interpretation that Iberian margin suspended particles are advected NW along isopycnals. Where are currents going at various depths? What about the possible influence of the broad shelf-slope region around the British Isles?

The section is presented for Nd only, as an illustrative REE and because in the near-future we will be able to trace the sources of lithogenic material with its isotopic composition.
The point located at 200 m at station #26 is on the same isopycnal as the point located at 700 m at station #21 (σ_0= 27.25), and there is no datapoint on this isopycnal at station #1.
Our stations are located in currents that form the North Atlantic Current and are flowing northward (Zunino et al., 2017), preventing influence from the British Isles.
In addition, we are currently working on the mechanisms of sediment resuspension along the Iberian margin on one hand and on the propagation of these intermediate nepheloid layers on the other hand. In order to better quantify these mechanisms, we use a circulation model (NEMO with a resolution of 1/12° and 50 vertical layers) but interpreting them farther was beyond the scope of this paper. However, this work was presented at Ocean Sciences (San Diego), confirming that there is no influence of the British Isles.

19. Line 303: “no particular lithogenic contribution” is used to describe agreement between the PFe and PMn results from Gourain et al., as compared to the PREE results presented here. But Sta. 53 HREE have a substantially higher %lithogenic than Sta. 51 (Fig. 5), indicating that there IS in fact an increase in lithogenic fraction at Sta. 53 only. So the phrase in quotes does not represent an observation “in agreement with our results”. This needs clarification and re-wording. The word “particular” is not clear in the above – do you mean “unusual”? Note also that authigenic contributions of Mn and Fe from shelf sediments may mask an increase in lithogenic Mn and Fe, when looking only at %lithogenic as a metric. This may be less true for REE, especially HREE, leading to the differences between Sta.’s 51 and 53 as noted above.

Regarding the reference to Gourain et al. (2019), we meant that the lithogenic contribution remains on the Greenland shelf and that no nepheloid layers were observed along the slope, in contrast with the Iberian margin. Correction is added in line 357.

20. Line 307: “At station #13 at 200m, no lithogenic maximum is identified”. The authors need to clarify whether they are still referring to Ac-227 data or are now referring to REE data. Indeed, Fig. 6 shows this depth to be a relative min. in %lithogenic Nd, but there are relative max’s just above and below. Please clarify language so it is fully clear which data you are referring to. Also, please use “lithogenic” and “%lithogenic” appropriately. It is possible to have high lithogenic concentrations but low %lithogenic, for example near a margin where weathering particles might combine with higher biogenic particles resulting from high productivity. In the quote above, I think you mean “%lithogenic”.

Yes, it is %lithogenic and not the absolute lithogenic concentration. It refers to ^{227}Ac, and this is indicated in line 362.

21. Line 308: “merging of the two maxima observed eastward”. Please clarify where the two maxima are and do you mean eastward of Sta. 13 or some other station? Also, Fig. 4 does not show anything about isopycnals; should this be citing Fig. 6?

The two maxima we are referring to are those observed at station #1 (200 m and 240 m), and we are now directing the reader to Fig. 5 and Fig. 6. See line 335 for modified text.

22. Line 322: The concluding paragraph should sum up the REE results. As it stands, it restates more general processes at ocean margins that have been established through previous studies. It would be better to summarize the central findings related to REE marine geochemistry.

We are now summarizing the results in lines 375-377.

23. Line 327: “less prone to desorption”. This phrase is not fully accurate because my understanding is that Ce, once oxidized, has a higher Kd, which describes an equilibrium state

where adsorption and desorption rates are equal, but distribution of Ce is more strongly in favor of the solid surface. See for example Ohta and Kawabe, GCA, 2001. Also, the literature contains some discussion whether adsorption or coprecipitation best describes the association of Ce(IV) with authigenic Fe and Mn oxyhydroxides. The nature of the anomalous redox behavior of Ce deserves a more complete introduction here, including its relative Kd via adsorption to MnO2 and FeOOH, from abundant published experimental data. It is important to understand as well as possible which major authigenic mineral phase is most responsible for REE adsorption or coprecipitation, and for Ce oxidation.

This part has been rewritten with a more complete introduction on the Ce special behavior, see lines 379-391.

24. Line 329: “authigenic Ce adsorbed on”. An adsorbed species must first be in solution, and “authigenic” usually refers to a mineral in solid phase, so this wording is incorrect. Whether Ce(III) is first adsorbed, then oxidized, or whether a discrete mineral (CeO2?) forms independently or in a co-precipitation process with a more abundant oxide is to my mind an open question, but the authors may wish to briefly summarize their understanding of the literature on this point. This reinforces the importance of the terminology referred to in the last comment.

To clarify these points, this part has been rewritten, see lines 379-391 for modified text.

25.* Line 338: The paragraph starting on this makes some assumptions that I think may not be true. First it is stated that Ce oxidation only occurs below the surface layer. In fact, without dissolved REE data, the authors cannot prove the Ce does not have a positive anomaly relative to the dissolved REEs.

The reviewer is perfectly right: the normalization to PAAS indicates that at the surface of productive areas a similar REE pattern to the seawater one is observed in particles, and then this is attenuated with depth. The observation of a more pronounced Ce anomaly below the surface layer does not necessarily mean that Ce oxidation did not occurred at lower depth. We are now softening this assumption in line 410.

The authors seem to have been caught in the trap of interpreting the Ce anomaly in absolute terms rather than in relation to the dissolved pool from which the REEs are adsorbed onto or absorbed into the biogenic particles. This dissolved pool has itself a very negative Ce anomaly throughout most of the ocean. Very likely then, the only way to achieve a positive Ce anomaly in marine suspended matter is via advection of sedimentary particles that have had very long time periods during which to undergo substantial REE cycling and fractionation, or as the authors suggest, preferential loss of the strictly trivalent REEs upon POM remineralization.

Thanks to your comment, we first noticed a mistake in our calculations of the Ce anomalies. Because of an error in the PAAS normalization, the presented Ce anomalies were larger than what they really are. The figure and the text of the manuscript were corrected. Please accept M. Lagarde sincere apologies for this mistake.
We do not agree on the ubiquity of the “very” negative Ce anomaly in the dissolved pool. Full depth published REE patterns show flatter shapes at the surface. More particularly, Greaves et al. (1994) and Tachikawa et al. (1999) reported surface patterns with quasi flat REE patterns (in particular no or slight HREE enrichment) following Saharan dust inputs.
Tachikawa et al. (1999b) also observed the formation of a positive Ce anomaly in suspended particles while settling through the water column. They explained this positive Ce anomaly by an adsorption of trivalent REE on newly formed Mn oxides (Moffett, 1994) without anomaly close to the surface. At greater depth, after particles began to settle, CeO_2 is adsorbed onto particles. This is consistent with a Ce oxidation slower than Mn oxidation (Moffett, 1994). These

anomalies then increase with depth by desorption of strictly trivalent REEs. This mechanism is also discussed in detail in de Baar et al (2018).
For example, at the surface of station #32, there is a negative anomaly of 0.36. The observed increase of this anomaly to a value of 1.1 (at 160 m at the same station) requires to increase the PCe concentration of 0.7 pmol L^{-1} to a concentration of 2 pmol L^{-1}. At this station, dissolved Ce concentrations are higher than 6.3 pmol L^{-1} in the first 150 m, and below 5 pmol L^{-1} below 450 m (preliminary results of our ongoing work on GEOVIDE dissolved REEs). Thus, a positive anomaly of PCe is likely to happen.

The authors also suggest, however, that a strong (positive) Ce anomaly could result from high particle concentrations. But a higher concentration of reactive surfaces would affect all REEs similarly; I don't see how high particle loads by themselves would lead to preferential Ce oxidation and retention on the particles.

This is only a suggestion. As the greatest positive anomalies occur in productive areas and identified lithogenic inputs, we suggest that higher particle concentration induces greater surface exchange and oxide formation.
It is possible that a higher bacterial activity enhances Ce oxidation (thereby forming CeO_2), in the areas of high productivity oxidation (de Baar et al., 2018; Moffett, 1990) and lead to high positive anomalies.

The authors assert that a positive Ce anomaly is not observed in the ARCT and NADR regions because export is strong and particle residence time is short, but Fig. 7 does show positive Ce anomalies at various depths in these regions, so this assertion seems untrue and needs more thorough examination and explanation. If Ce is preferentially removed under all scavenging scenarios, then it is reasonable to expect that a productive region with rapid export would deplete dissolved Ce in the mixed layer in both absolute and relative terms, through the course of a weeks-long bloom, leading potentially to strongly negative Ce anomalies in the dissolved state and increasingly negative (though positive relative to the dissolved pool) in the near-surface particles sampled at some advanced bloom stage. I encourage the authors to examine whether this scenario has been shown or disproven in other productive regions.

Yes, the exact proposition is that a positive Ce anomaly is not observed at the surface in the ARCT and NADR regions (in the upper 100 m), because seawater-like patterns are observed in these areas. The hypothesis of a preferential scavenging of Ce is supported by the attenuation of the Ce anomaly with depth, until it is close to the absence of anomaly with a value close to 1 at 200 m. This scenario has been shown by Moffett (1990) who observed weaker anomalies where the particles export was more intense. He suggested that the kinetics of exchanges between the dissolved and the particulate pool rely on the time they are in contact. If particles are removed faster than the Ce oxidation occurs, the anomaly will be weaker than in areas where particle have a residence time closer to the equilibrium of the reactions that leads to preferential scavenging of Ce. This was included to the manuscript, lines 408-413.

Even if Ce oxidation were not favored in the sunlit ocean, I would guess that the dissolved REE pool would show a fairly strongly negative Ce anomaly, inherited during previous deep winter mixing. In sum, I would expect for most oceanic regions that Ce anomaly to be negative for biogenic particles in the euphotic zone in general, unless there were an admixture of authigenic particles (perhaps resuspended from shelf sediments) overwhelming the biogenic effect. The authors may be able to refute this idea based on published data, in which case this should be stated as part of the discussion in this section. These two particle sources are often mixed in highly variable ratios in ocean margins, and I would think that alone would make total particulate REE data difficult to interpret. I think this is the reason why the authors have so much trouble in the last few sentences of this paragraph seeing consistent correlations between Ce anomaly and Mn behavior, particle concentration, particle residence time, etc.

Finally, I think the very sharp strong single-point maxima in Ce anomaly (Fig. 7A, B; e.g. Sta. 32, 440m and Sta. 13 600m) are very likely a result of Ce-specific contamination. Unless the authors can justify these oceanographically surprising features, those data points should be deleted from the graphs, and the data table values marked as likely contaminated values. See similar comments below.

We cannot exclude random contamination in Ce during the sampling, and we do not have a clear explanation. These data are not included in the graphs. They are reported under brackets in Table 2. See lines 437-440 in the revised MS.

26.* Line 360: The opening sentence of this paragraph seems incorrect to me, and gets the reasoning behind the interpretation of REE patterns in biogenic particles off on the wrong foot, affecting the rest of this discussion section. The LREE's likely have a larger lithogenic fraction than do the HREEs NOT because the LREE's are not preferentially taken up by (or onto) the biogenic particles, but simply because the surface seawater dissolved REE pool is so LREE-depleted relative to PAAS.
The degree of LREE-depletion cannot be known for the stations investigated here because no dissolved REE data are presented, but even with preferential LREE scavenging, a mixture of crustal minerals and biogenic surface particles would always show the observed larger lithogenic component for the LREE, because the preferential LREE uptake on the biogenic particles cannot come close to compensating for how depleted the LREEs are in the dissolved pool.
This depletion might be especially true for a surface layer that has already seen substantial growth and export in the preceding weeks, which could cause the surface layer to be even more LREE-depleted than it was immediately following winter mixing. I encourage the authors to "borrow" dissolved REE data from elsewhere in the Atlantic (no one measure them on GEOVIDE??), assume a degree of preferential LREE uptake based on published laboratory adsorption experiments or papers showing both dissolved and particulate data for euphotic zones in other regions, and do the calculation themselves. Without looking exhaustively at the literature, it is evident from dissolved REE data near Bermuda (see deBaar et al., GCA, 2018, Fig. 10) that dissolved Nd/Yb decreases from deep water up to the surface. In sum, a "seawater-like" pattern does not necessarily imply a unique LREE/HREE fractionation, or absorption vs. adsorption, it simply means short-term uptake from a very LREE-depleted pool, without significant admixture of authigenic minerals or refractory lithogenic particles, which are so important to the total PREE patterns below the surface layer. I think it is a mistake and is misleading to refer to "HREE enrichment" because PAAS-normalization has little relevance for biogenic marine particles, and this term implies preferential HREE uptake, relative to LREE. I also think that the speculations about REE patterns implying some kind of control by biogenic carbonate vs. Silica is poorly reasoned and not convincing; I would argue that the vast majority of REEs in biogenic particles (mostly living cells in productive surface waters) is associated with organic matter, as is true for nearly all trace metals. In sum, I strongly disagree with the interpretations in section 4.4 and I urge the authors to reconsider and rewrite this entire section.
As a related side observation, I see from Fig. 5 that the REE pattern of near-surface particles from Sta. 53 shows high concentrations (relative to Sta. 51) and strong LREE-enrichment (shown log scale). This station was dismissed in the first sentence of the section as distinct from most other stations which form the basis of the discussion in Section 4.4. But the question remains how this LREE-enrichment might occur. Is this a result of preferential LREE scavenging from a "flat" dissolved REE pattern, or does it reflect mineral particles from sediments or the continent that are already LREE-enriched, for example the authigenic products of previous long-term particle-seawater interactions?

At the surface, even if LREE are depleted by comparison to PAAS, LREE concentrations are still higher than HREE concentrations due to their natural abundance (de Baar et al., 2018; Fig. 10). We also know that dissolved REE concentrations are higher than particulate REE concentrations which represent only 5% of the total Nd (Jeandel et al., 1995). We are currently performing the analyses on dissolved REE samples collected during GEOVIDE. Preliminary results also confirm that dissolved Nd concentrations are 5 to 65 times higher than particulate Nd concentrations. Particulate LREE represent 3% to 5% of total LREE pool (data for stations #1 to #32, ongoing work). Therefore, a short-term uptake mechanism would be in favor of LREE at the surface if the REE distributions are driven by adsorption processes. With depth, the dissolved patterns show a decrease of the DNd/DYb ratio, by preferential adsorption of LREE on oxides and hydroxides on particles. In addition, lithogenic inputs by dust or resuspended sediments can lead to an LREE enrichment in surface waters (Greaves et al., 1991, 1994; Tachikawa et al., 1999a for dusts, station #53 and #1 are an example for lithogenic inputs).

The association of REE with biogenic silica and calcium carbonates is still under debate in the literature (de Baar et al., 2018 and references therein; Patten and Byrne, 2017). The link between biogenic matter and REE is mostly established by the observation of a correlation between REE and major nutrients (de Baar et al., 2018). However, only few works are documenting the REE concentrations in the different particulate fractions yet: Akagi, (2013) and Akagi et al. (2011) suggest a main control by the biogenic silica, while de Baar et al. (2018) are in favor of a control by the soft material (traced by P and N).

This incorporation of REE in particles with a seawater-like pattern by association with soft tissue is what we observed in the NADR region, where a coccolithophorids bloom occurred during the cruise. Yet, the PYb_N/PNd_N ratio observed in the ARCT area is contrasting with the PYb_N/PNd_N ratio of the NADR region. This ratio is maximum where the diatom bloom occurred, while the REE patterns in the Labrador Sea (ARCT) are less similar to a seawater pattern than in the NADR region. This could be explained if one assumes that the bloom is senescent and the lithogenic inputs are higher. Then, HREE seems more particle-reactive at the surface of the ARCT region than at the surface of the NADR region, when LREE seems to be less depleted at the surface due to a higher lithogenic component. This is consistent with the theoretical work of Akagi (2013), and seems to indicate that HREE are more linked to Si cycle than LREE.

Station #53 is dismissed at the beginning of the section as it is not considered as an "open-sea" station because it is located on the Greenland shelf and subject to high particles inputs from Greenland that results in a dominant lithogenic signal. This lithogenic fraction is not as high as at station #1 and is higher for HREE than for LREE. LREE are found in higher proportion than HREE in the authigenic fraction, suggesting a preferential scavenging of LREE, unlike what happens at the stations discussed in this section. The roughly constant Ce anomaly around 1 confirms that a lithogenic origin is more probable than LREE-enriched authigenic products.

27. Line 426: The meaning of "dynamic scavenging" used here and above, and how it can generate a positive Ce anomaly, should be explained further. My understanding of the authors' meaning is that that successive cycles of adsorption and desorption accompanied by progressive Ce oxidation, can increase the Ce anomaly until it is strongly positive. But could remineralization of organic matter, and loss of the associated REE, leaving MnO2 and other authigenic oxyhydroxides as a greater fraction of the overall particulate REE, have the same effect, as long as refractory lithogenic particles (no Ce anomaly) were not an important part of the mix? This is not the same process as repeated cycles of adsorption and desorption on a constant particle population.

The reviewer understanding is right, we meant that a positive Ce anomaly was generated by successive cycles of adsorption/desorption of REE, with less desorption for Ce. A remineralization of organic matter would not prevent a preferential adsorption of Ce on oxides and hydroxides to occur. The amount of adsorption sites would modify the intensity of the Ce anomaly and depends on the particle modification through time. Remineralization would modify the intensity of the anomaly too, with a decrease of the negative Ce anomaly imprinted by organisms from seawater, but cannot generate a positive Ce anomaly without adsorption of Ce on particles.

28. Line 429: “and then a stronger scavenging of REEs”. It is not clear what this phrase means. Is this proposed to be the second step occurring at this depth interval, after “intensive exchanges”, or does “then” mean further down the water column. I’m not clear how ones achieves the combination of high PHo enrichment and strong positive Ce anomaly. Could the responsible processes be occurring independently, involving different particle types within the suspended particle mixture? Overall, it seemed to me that this section dutifully follows a pattern of discussion points in other REE papers from the senior author’s group, and is interesting on theoretical grounds, but did not advance understanding of the relative behavior of Ho and Y in the ocean to a significant degree.
The variations in Fig. 8 seem barely interpretable in any cohesive way. The authors should reconsider whether this section truly adds to the impact of the paper. To my reading, it makes the paper end on a somewhat vague note.

Following this comment and the previous ones, this section has been entirely reworded. We agree that Fig. 8 does not bring important information and we deleted it. Instead, we proposed a general conclusion in lines 516-547.

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