Answers to reviewer 1

1) I think Figure 1 still needs some work. One of the previous reviewers recommended adding a scale bar to Figure 1C, which the authors say they have done, but I could not find it. By scale bar, I assume that the reviewer was referring to distance in km in addition to the latitude and longitude data that was already included. In addition, the Petrohue river does not appear to be labeled, while the other two rivers are labeled in different font sizes. And the symbol for the hydrological station is difficult to see in the figure. Perhaps a solid black triangle would be better. In the caption, the ash plume extent should be stated as "approximate" as it represents a full week of coverage for a dynamic feature.

Answer: Changes have been made to the new figure 1. A scale bar (in km) has been placed in addition to the lat/long, all three major rivers are labeled at the same font size and the station symbols are now bolder against the background. In the description and text is stated that the ash plume extent is an approximate representation for a full week of coverage of a dynamic feature (new lines 139-140)

2) I think the collection of the ash sample still needs more clarification. The eruption is described in lines 144-145 as taking place in two pulses on 22nd April and 23rd April. The ash sample was not collected until 6th May. In the response to reviewer 3 the authors state that the ash sample was collected after the third eruptive pulse. The timing of this third pulse should therefore be included in the eruption description to clarify to readers that there was not much time between deposition and sample collection. (The authors do mention an ash plume on 6th May in the caption of Figure 2, but details of this third pulse should also be mentioned in the text when discussing the eruption timing).

Answer: at the beginning of section 2.2 the timing of the sample collection of ash is clarified. We extent the introduction to mention the smaller 3rd eruption and now refer clearly to the main/first eruption throughout the text except where explicitly referring to the smaller 3rd eruption (new lines 147-150).

3) With the inclusion of Table 1, there should be some reference to it in the text, preferably before getting into details about the different leaching experiments. I also suggest splitting the water type (DI water, seawater, etc) and the measurements made (trace metals, alkalinity) into separate columns in Table 1.

Answer: Table 1 was modified to show the information in a clearer way with new columns and fewer rows and is referred to through the methods section (e.g. lines 177, 215)

4) There is still a problem with Figure 4 and/or Supplementary Table 2. The station-by-station data for diatoms in the figure do not match those in the table. According to Supp.Table 2, the circles in Figure 4 represent diatoms at Stn.A, the triangles Stn.B, and the squares Stn.C. This problem was brought up in the previous round of review and the authors stated that it had been addressed, so it may be that the updated version of the figure somehow didn't get included. It needs to be addressed, along with any associated changes needed in the text regarding diatom abundance at the different stations (e.g. Line 326).

Answer: Sup. Table 2 was correct, we apologize we had mis-interpreted a prior comment and amended the description of fig 4 in the text rather than the legend on the figure – the error was in the figure, with A/B/C not labelled correctly. Figure 4 has been changed to display the correct labels.

5) The supplementary material for this manuscript includes five tables and three figures and yet reference is only made to one supplementary figure in the text (four of the tables are mentioned with regards to data availability). It would make sense to include references to the appropriate supplementary materials in the main text.

Answer: All tables and figures are now properly mentioned in the text.

6) In section 4.3 the authors no longer discuss the SW Atlantic area marked on Figure 8; instead referring to the area further south and shown on the supplementary figure. I think the better approach here involves first discussing the two Pacific areas, as they have done, and then comparing the two Atlantic areas – the one in Figure 8 that did not show any change in chlorophyll-a and then the one further south where location on the shelf and lower satellite coverage complicate interpretation.

Answer: Significant changes have been made to section 4.3. to highlight the two impacted areas and Fig. 8 with Sup. Fig 3 are now discussed together in a logical order as suggested (new lines 682-722)

Technical comments:

Line 49 – The symbol for fluoride should be F-, not Fl-. This was brought up by Reviewer 2 previously. Also at Line 227 and Table 2.

Answer: We have corrected this through all the text.

Lines 68-72 – Suggest changing this sentence to "A pulse of Fe(II) release upon addition of Calbuco ash to seawater made it an unusually efficient dissolved Fe source. The fraction of dissolved Fe released as Fe(II) from Calbuco ash (~18-38%) was roughly comparable to literature values for Fe released into seawater from aerosols collected over the Pacific Ocean following long range atmospheric transport".

Answer: Suggestion accepted, now in current lines 68-72.

Line 96 – change to "some freshwater bodies"

Answer: Suggestion accepted, current line 96

Line 104 – Remove the first "North Atlantic" reference from this sentence and change the year of the Eyjafjallajokull eruption to 2010 (noted in the previous round of reviews).

Answer: Suggestion accepted, current line 104 now reads "In contrast to the 2010 Eyjafjallajökull plume over the North Atlantic"

Lines 106 – The use of "largely" in this sentence is made redundant by the use of "predominantly". Suggest removing "largely".

Answer: Suggestion accepted, current line 106 has "largely" removed.

Line 115 – Suggest changing the las part of the sentence to "..and leaching experiments carried out on ash collected from the fjord region, to investigate the inorganic consequences of ash deposition to natural waters".

Answer: Suggestion accepted. New sentence can be found on lines 114-116.

Lines 124-128 – I'm confused as to why the Petrohue is identified as the major river of the system (line 125) when the Puelo is the largest in terms of flow. Perhaps the first sentence could be adjusted to "The Calbuco volcano is located in close proximity to Reloncavi Fjord".

Answer: Problem resolved due to mistake on naming the Petrohue as the major river. Now reads "The Calbuco volcano (Fig. 1) is located in a region with large freshwater reservoirs and in close proximity to Reloncaví Fjord."

Lines 161-163 – Ash was collected on 6 May. How long after deposition was this? Also clarify that the weather was dry throughout the period between deposition and collection. At present it is ambiguous – just the day preceding ash collection.

Answer: We clarify details concerning the ash collection which was whilst the 3rd eruption (small) was ongoing, that the ash collection site was 30 km away from the volcano and that the ash was collected over a 24 h time period (i.e. freshly deposited ash from the 3rd eruption). Yes, the weather was dry throughout this time period (the time period plasticware was left outside for, and the day prior to this) see new lines 147-150 and specifically 163 "On 6 May (2015, Cochamó, Chile, approximately 30 km from the volcano) after the third, and smallest, eruptive pulse of ash from the Calbuco volcano (Fig. 2, A), and with the volcano still emitting material, ash was collected using a plastic tray wrapped with plastic sheeting (40 × 94 cm). The plasticware was left outside for 24 hours until sufficient ash (~500 g) was collected to provide a bulk sample. Ambient weather over the period of ash collection, and the preceding day, was dry (no precipitation)."

Line 176 – The range of concentrations selected was justified in comments to a reviewer as being broadly in line with previous studies. This justification could be included in the text for all readers.

Answer: In response to this, and comments from another reviewer concerning best practices from prior work, we include Sup. Table 1 showing all prior seawater leach work, and introduce a new paragraph evaluating the best options concerning leaches to investigate Fe release under conditions that might mimic natural conditions. New lines 178-213.

Line 178 – Specify that the addition was of concentrated HCl.

Answer: Suggestion accepted. Line 233

Line 181 – Give the (approximate) seawater temperature.

Answer: Suggestion accepted (now line 237), now approximate temperature is shown with reference to Sup. Table 1.

Lines 183-184 – Suggest changing to "For these experiments, a pre-weighed mass of was added to 250ml South Atlantic seawater and manually shaken for approximately one minute, using an expanded concentration range of 0.2-4000 mg L-1"

Answer: Suggestion accepted. New lines 240-243.

Line 196 – change to "flow-through".

Answer: Suggestion accepted. Line 253

Lines 215-217 – The inclusion of the size distribution of the ash is a good addition. In the response to reviewers, the authors also make the argument that the <63um size fraction has the greatest surface area and so the choice of size fractions includes both the dominant size fraction by mass (250-1000um) and the dominant size fraction by surface area (<63um). A couple of lines making this argument should also be added to the manuscript to clarify for the reader.

Answer: Suggestion accepted. New lines 277-279 "The dominant size fraction by mass was thereby the 250-1000 µm fraction which was analyzed in addition to the finest fraction (<63 µm) with the greatest surface area to mass ratio."

Line 227 – Change Fl- to F-.

Answer: Corrected (new line 289).

Lines 234-240 – There is unnecessary repetition here. The first list of parameters could be removed, giving a sentence that reads "High temporal resolution (hourly) in situ measurements were taken simultaneously in the Reloncaví fjord (Fig. 1 C, North Patagonia Oceanographic Buoy) at the surface and at 3 m depth, using two SAMI sensors that measured spectrophotometric CO2 and pH (DeGrandpre et al., 1995; Seidel et al., 2008) (Sunburst Sensors, LLC), and an SBE 37 MicroCAT CTD-ODO (SeaBird Electronics) for temperature, conductivity, depth and dissolved O2, as per Vergara-Jara et al., (2019)."

Answer: Suggestion accepted, we have shortened this section (new lines 296-300) "High temporal resolution (hourly) in situ measurements were taken in the Reloncaví fjord (Fig. 1 C, North Patagonia Oceanographic Buoy) at 3 m depth using SAMI sensors that measured spectrophotometric CO_2 and pH (DeGrandpre et al., 1995; Seidel et al., 2008) (Sunburst Sensors, LLC), and an SBE 37 MicroCAT CTD-ODO (SeaBird Electronics) for temperature, conductivity, depth and dissolved O_2, as per Vergara-Jara et al., (2019)."

Line 254 – "an" instead of "a"

Answer: Suggestion accepted, line 316.

Lines 259-262 – Reword as "Additionally, as part of a long-term monitoring program at station C (Fig. 1), chlorophyll-a samples were retained from 6 depths (1, 3, 5, 7, 10 and 15 m) on 6 occasions during March-May 2015. Chlorophyll-a was determined by fluorometry after filtering 250 ml of sampled water through GFF filters (Whatman), as per Welschmeyer (1994)."

Answer: Suggestion accepted, lines 319-324.

Line 266 – "…for comparison to prior reported data integrated to 10m…"

Answer: Suggestion accepted, line 327.

Line 267 – I don't think it is necessary to reference Figure 4 here, particularly as Figure 3 has not yet been referenced.

Answer: Suggestion accepted, reference to Figure 4 removed.

Line 275 – Similarly, there is no need to reference Figure 7 here, when earlier figures have not yet been referenced.

Answer: Suggestion accepted, reference to Figure 7 removed.

Line 327 – Is station B the more freshwater influenced? Station A is further upstream in the fjord and close to the second largest river flow input. Station C is located close to the largest river flow input. Is there data to back up station B being more freshwater influenced?

Answer: Changes were done to correct this, because the wording was ambiguous. A is in the inner-fjord although C sits at the mouth of a river outflow, so the term 'freshwater influenced' was ambiguous as it. (new line number 390-399), we now refer to the 'innermost station' (A) for clarity.

Lines 329-330 – Reword this to state that highest chlorophyll concentrations were measured close to Station C on that date (rather than at C, including two nearby measurements).

Answer: Suggestion accepted, new line 391 "The highest measured chlorophyll-a concentrations were on 30 April at Station C,".

Line 358 - Change Fl to F (also Table 2). Also, elsewhere in the manuscript all of these chemical species are written as ions, which is what is measured by the technique, and should also be written as such here for consistency.

Answer: Suggestion accepted (new line 421).

Line 366 – The authors responded to a reviewer comment about brackish water leaching results not being included in Table 2 because the higher original concentrations of major ions in brackish water make it difficult to observe changes. A statement to that effect could be added here for the general reader.

Answer: Suggestion accepted. New lines 430-432 states: "Major ion analysis was only conducted in de-ionized water as no significant changes would be observable for most of these ions in brackish or saline waters under the same conditions."

Lines 376-377 – In the methods the ash loading for these leaching experiments is described as 2-50 mg/L, and in Figure 6 it is given as mg ash added, but here it is described as 0.1-6 mg/L. I think this latter case is supposed to be mg/100mL or just mg added. Please change units or numbers for consistency.

Answer: Suggestion accepted, numbers, units corrected "The net release of dissolved metals proceeded with varying relationships with ash loading over the applied gradient (2-50 mg L^{-1})" (new lines 441).

Lines 377-383 – No mention is made here of where Fe fits into this grouping.

Answer: This section is expanded slightly to better discuss all elements, new lines 440-459.

Figure 7 (left panel) – Now that only Calbuco ash is included in the figure, the x-axis label should be changed to "ash loading", rather than "dust loading".

Answer: Suggestion accepted, X-axis label corrected as "ash loading / mg L-1".

Figure 7 (left panel) – the lines fitted to each dataset (2 weeks, 4 months, 9 months) are not mentioned in the figure caption.

Answer: Suggestion accepted. They are now mentioned explicitly in the caption (new line 474).

Figure 7 – The caption (and description in the methods) indicates a 30-minute time-series of Fe(II) measurements, but the x-axis of the right-hand panel clearly extends to beyond an hour. Please clarify in the text and caption.

Answer: Suggestion accepted, clarification introduced, now Figure 7 legend read as "Figure 7. Fe(II) release from Calbuco ash into seawater. Mean Fe(II) released into South Atlantic seawater over a 30 minute leach at 5-7°C (left). The same batch of Calbuco ash was subsampled and used to conduct experiments on 3 occasions after the 2015 eruption (2 weeks, 4 months and 9 months since ash collection). The lines are power law fits, with associated equations shown in the legend. The 3 time-series of Fe(II) concentrations following ash addition is considered collectively by normalizing the measured concentrations (right), such that 1.0 represents the median Fe(II) concentration measured in each experiment. All experiments were conducted for at least 30 minutes, those conducted with 4/9 months old ash were extended for 1 hour. The black line shows the mean response over 34 leach experiments with varying ash loading, the shaded area shows ± 1 standard deviation. The initial Fe(II) concentration (pre-ash addition at 0 s) in all cases was below detection and thus the detection limit is plotted at 0 s (open circle). Source data is provided in Supplementary Table 2.".

Line 415 – "Concentrations of up to 4.0 nM Fe(II)" are not evident in Supplementary Figure 2 (which should be referenced) or in the data in Supplementary Table 1. Perhaps 4nM represents one value from replicates of the 4 month old ash, 2mg/L ash loading experiment (2.17+-1.07 nM Fe(II) in the table), but that should be acknowledged in the text.

Answer: Suggestion accepted, yes the 4.0 nM refers to a measured concentration, but not a mean (which is what is displayed in Sup. Table 2). For clarity, we refer to the mean Fe(II) concentrations that were observed, new line 491 "Elevated Fe(II) concentrations (mean 0.8 nM, Sup. Table 2) were evident at this temperature (5-7°C), which represents an intermediate sea surface temperature for the high latitude ocean."

Line 419 – reference Supplementary Figure 2 also.

Answer: Suggestion accepted, reference placed e.g. new lines 237, 495.

Line 431 – in response to a comment from Reviewer 2 the authors provide a reference (Torres et al 2007) to support their interpretation of UV Aerosol Index as a measure of dust/ash aerosol loading. Why not cite that reference here in the text?

Answer: Suggestion accepted, reference placed at section 2.6, line 334.

Lines 436-438 – reference the appropriate supplementary figure here.

Answer: Suggestion accepted, reference placed, line 468.

Figure 8 – dashed vertical lines (22nd April) in panels G and H are hard to discern. Perhaps red would be a more visible colour to use.

Answer: Solid red lines are now used throughout to represent the eruption dates.

Line 471 – change “ash fall” to “ash deposition event” to avoid confusion with the fall season, mentioned in the line above (or use autumn instead of fall for the season).

Answer: Suggestion accepted, now instead of “ash fall” is “ash deposition event”, throughout including new line 553.

Line 491 – it is stated that the diatom bloom (peaking 14th May based on data available) occurred several weeks after the eruption. This adds to the confusion about collection of the ash sample, which was carried out a week earlier on 6th May. But in response to reviewers the authors state that this collection took place after a third eruptive pulse of ash. This suggests an additional ash deposition event closer to the bloom peak. Please clarify.

Answer: As per previous comments, we clarified that a 3rd (small) eruption occurred after the two major eruptions. Specific to the above comment, new lines 561-564 “Peaks in diatom abundance were measured at two stations on 14 May one week after the third (small) eruptive pulse, and measured chlorophyll-a concentrations were highest close to Station C on 30 April (Fig. 4)”.

Line 544 – See my general point about changes needed to Figure 4 – currently the figure does not show a more modest diatom abundance at Station A, as stated here, but rather at Station B. Also, the authors again state that this date (14th May) is 3-weeks after the eruption – should it be three weeks after the main eruption (see line 491 comment)?

Answer: As the figure 4 was labeled wrong with respect the correct station labels A/B/C, the data does show (Supp. table 3) that station C & B had higher diatom abundance than station A. As above comments instead of referring to an “eruption” we now clearly distinguish the major eruptions on 22/23 April from the later, small eruption on 30 April. (new line 629) “Considering the more modest peak in diatom abundance at the most strongly ash affected station (Station A, Fig. 4) and the timing of the peak diatom abundance 3 weeks after the main eruption,”

Lines 570-574 – I believe this sentence is added based on a reviewer comment about previous work. In the response to reviewers the authors argue that these lower pH experiments may not be applicable at seawater pH and I think that caveat can be added to this sentence.

Answer: Clarified, new line 658- reads “Alternatively Fe(II) could be released from more crystalline Fe(II) phases. Prior work, at much lower pH (pH 1 H2SO4 representing conditions that ash surfaces may experience during atmospheric processing, but not in aquatic environments) suggests that short-term release of Fe(II) or Fe(III) is determined by the surface Fe(II)/Fe ratio which may differ from the bulk Fe(II)/Fe ratio due to plume processing (Maters et al., 2017).”.

Answer to Reviewer 2

Vergara-Jara and co-workers' present study is based on collected samples of ash fallout from the 2015 Calbuco volcano emission. Their leaching experiments were performed on two different sets of samples: (i) South Atlantic seawater (bulk ash leaching for trace metals (Fe, Cd, Pb, Ni, Cu, Co, Mn) and Fe(II), and (ii) DI water and Aysen fjord water (sizefractionated ash samples; <63 μm and 250-1000 μm) for total alkalinity (Aysen fjord, DI water) and major ions (DI water). In brief, the authors did a multitude of different leaching methods in the present work and looking at the present work one more time, I feel that the study is not that focussed. I firmly believe that the present study would have benefitted from a more focussed approach from the authors regarding their leaching experiments.

One of the critical aspects that I am still not entirely convinced with is some of the parameters/protocols used for leaching experiments conducted in the present work. Different leaching experiments were done on a different set of water samples (S. Atlantic seawater, brackish water from the Aysen fjord, DI water), making the results non-comparable, especially with previous such studies (Jones and Gislason, 2008).

Reply: This is a generic criticism that affects all work conducted on volcanic ash, not just this study. There are only 2 leaching experiments that can be done on water in a completely comparable way; synthetic seawater, or de-ionized water. These have no environmental relevance at all and are particularly problematic for Fe. The objective of this study was not to compare ash to other eruptions, but to understand environmental changes in marine environments around the region where ash was deposited.

Fe(II) leaches in deionized water, or synthetic seawater at room temperature would be meaningless as the features of natural water that moderate Fe(II) release are not replicated in either case, so we disagree with this comment. The details of the water selected for any leach should be matched to the hypothesis. If the experiments were designed to compare specific details of inorganic chemistry contrasting ashes from different volcanoes, yes we agree a single standardized synthetic seawater leach would be appropriate, but that is not what we were doing herein.

To clarify to what extent our work is/is not comparable to prior work we include a new paragraph describing the rationale, and the set up of prior work which investigated ash additions to seawater (new lines 172 - 214).

The study was also largely focused on (biased towards) explaining the results of the trace metal leaching experiments done on S. Atlantic seawater.

Reply: This is the main (possibly only) mechanism that could explain an increase in offshore primary production, as was detected by satellite derived chlorophyll (Fig. 8) so yes this is of course a focus of the text. We now explain this rationale in lines 172-214.

Although the authors performed their leaching experiments on different ash size fractions (<63 μm and 250-1000 μm) on brackish water and DI water, they could not extract any major conclusions out of these experiments. Also, the discussion section primarily focussed on the results of leaching experiments done with the S. Atlantic seawater. So, considering all this, I am not entirely sure why the authors performed these different ash size-fractionated (<63 μm and 250-1000 μm) leaching experiments on a different set of samples (brackish water from the Aysen fjord, DI water).

Reply: As stated in the introduction (new lines 172-214), we want to investigate to what extent the ash could affect primary production over the spatial scale here the ash was deposited which covers a

range from fresh, to estuarine, to offshore waters. Hence the main mechanisms that could plausibly affect primary production in these waters (release of bioavailable N, Si, Fe, Mn; moderation of carbonate chemistry) should be investigated in conditions that represent these waters. Changes in the concentrations of ions and changes in the carbonate system could only really potentially affect primary production in freshwater (or perhaps brackish water) systems, as the reviewer pointed out previously. Conversely, trace metal fertilization could only really positively affect primary production to a large extent in offshore marine systems. So the experiments are designed to quantify these potential shifts in a meaningful way. Ionic leaches in seawater would be pointless, and conversely trace metal leaches in deionized water to quantify Fe release in seawater would be of dubious environmental relevance.

When last reviewed, I had some concerns on how (and why) the authors defined different ash size fractions: <63 μm (fine fraction) and 250-1000 μm (coarse fraction) for their leaching experiments. Although the authors have tried to justify that they have used different ash size fractions "following the recommendations of Witham et al., 2005", I, specifically, did not find any such recommendations on these particular size fractions by Witham et al., 2005. Further, in connections with their leaching experiments with the DI water, even though the authors have claimed that "DI experiments provides additional opportunity for comparison with similar studies (e.g., Jones and Gislason, 2008)", the authors have not utilized this opportunity by discussing these comparisons.

Reply: Prior literature does not specify which exact size fractions should be used, merely guidelines concerning the consideration of size fraction as an important consideration. Our rationale for these specific size fractions is now stated in lines (278-280) "The dominant size fraction by mass was thereby the 250-1000 μm fraction which was analyzed in addition to the finest fraction (<63 μm) with the greatest surface area to mass ratio".

The results from the earlier study of Jones and Gislason, 2008 have been simply provided in Table 2, without discussing whether the similarities are valid or not. Additionally, (i) the leaching experiments performed by Jones and Gislason, 2008 were on a different ash-size fraction (45–125 μm), and (ii) the leaching experiments of Jones and Gislason, 2008 were conducted in Teflon single pass plug flowthrough reactors (different from the present work). In light of these points, I do not think that authors' comparisons on their DI leaching experiments with earlier results of Jones and Gislason, 2008 would be valid. In fact, for making their comparisons to be validated against an earlier study (Frogner et al., 2001), Jones and Gislason, 2008 used the same experimental methods (I am quoting it here: "To allow direct comparisons with previous work, we have attempted to apply the same methods as used in Frogner et al. (2001)" from Jones and Gislason, 2008). So, at least, if the authors of the present work have performed their leaching experiments with DI water with an intension to compare their results with previous findings, they should discuss any similarities (or, discrepancies) in these comparisons in more details

i.e. to answer: why do they expect their results to be any similar or different from previous studies? What about different size fractions used (in contrast to Jones and Gislason, 2008)? What about differences in leaching experiments?

Reply: As stated in the introduction, the main aim herein is to investigate the environmental effect of the Calbuco eruption, not to conduct an extensive comparison to every other eruption for which ash is available. Several extensive reviews have been written on this subject going into detail about what is, and is not, known concerning the effects of size, time, age, leachate on measured ionic and trace metal leaches which are cited extensively throughout. We do not think it is appropriate to repeat this discussion herein as it is ancillary to the main aim of the paper, particularly concerning the potential

effects of ash (change in alkalinity, and ion availability) that it is obvious from the results did not have any environmental effects on phytoplankton following the Calbuco eruption. Nevertheless, we have expanded the discussion of to what extent a comparison is/is not valid for the leaches (trace metal) that potentially did have environmental effects. See new Supp. Table 1 and new lines 172-214.

There is a very long discussion to be had concerning the precise design of any experiment to investigate any aerosol dissolution in seawater which we do not think is useful to discuss extensively herein, a general comment on the difference between the continuous plug flow reactor used by previous researchers and mixed reactor (bottle experiments) used here for example, is that flow through systems used a filter to retain sediments in place, which is susceptible to clogging, and hence limits its use to larger particle sizes which will not clog the system (the problem of clogging is something alluded to in the methods of Jones and Gislasson). As a consequence, previous or otherwise standard methods may be inadequate for assessing the effects of the most relevant fine particle sizes, which are generally more abundant and are certainly more widely deposited over larger geographic areas. Ultimately, residence time (and not reactor design) is the basis for the limited comparison between this experiment with previous experiments. Meanwhile, we can say that residence time was very similar compared to previous studies, and the concentrations and rates were within the same range.

I also have major concerns regarding the processing of the S. Atlantic seawater used for trace metal leach experiments. The authors have not mentioned whether the S. Atlantic seawater sample was filtered or unfiltered. Besides, the authors' protocols for seawater processing deviated significantly from Jones and Gislason, 2008, wherein much more robust protocols for seawater processing were followed. Upon collection, Jones and Gislason, 2008 processed their seawater samples by filtering (through 0.2 μm filter; to remove particular matter) and subsequent irradiation with UV light (to kill the remaining biota) before storing the samples in the dark. Such robust seawater preservation methods (before leaching experiments) were found to be missing in the present study.

Reply: For clarity we now include details of all prior work conducting ash leaches in seawater (Supp Table 1 and new lines 179-206). We disagree that the 'robust' technique above would have been appropriate to use herein for two reasons.

UV irradiation of seawater will destroy any natural organics present, it will also lead to high concentrations of compounds like H2O2 which will take weeks to months to decay back to background concentrations. The chemistry of Fe in seawater is controlled almost entirely by the concentration and nature of the organics present – organics which the reviewer previously commented we should think carefully about. There are of course circumstances under which it would be desirable to UV-treat seawater, but we do not think this is one of them as the cycling of Fe and Fe(II) would be interfered with.

The Atlantic water used was filtered (new lines 222), it was oligotrophic to start with, and the concentrations of any bacteria present after storage in the dark for > 1 year would be insufficient to make meaningful differences during leach durations of 10 minutes (this would apply even if water were unfiltered and leaches were conducted immediately for such a short leach duration).

Regarding the trace metal leaching the protocols with the collected ash, the authors added pre-weighed ash into 100 ml of S. Atlantic seawater and gently mixed the suspension for 10 minutes. How did the authors decide on this particular mixing time (10 minutes)? Did the authors change the suspension mixing durations to see the impact of ash-interaction with seawater for a reduced or prolonged interaction duration?

Timings vary widely in prior work, but all studies show that the major processes occur practically instantaneously such that there is little difference between initial leach timings of 10 minutes, 1 hour or 2 hours (see new lines 210-214) "A short leaching time (10 minutes + filtration) was adopted to minimize bottle effects and recognising that most prior work suggests a large fraction of Fe release occurs on short timescales (minutes), followed by more gradual changes on timescales of hours to days (Duggen et al., 2007; Frogner et al., 2001; Jones and Gislason, 2008)."

I am also not sure why some of the parameters were changes during the manuscript. E.g., although the coarser size fraction was defined as 250-1000 μm fraction in the earlier part of the manuscript (section 2.3, Table 1), the same coarser fraction is defined as >1.0 mm later in the manuscript (section 3.3, Fig. 5).

Answer: We apologize, there are only size fractions considered "<63" and "250-1000", the later was incorrectly referred to as ">1000" in one place in the prior work.

In view of the above considerations, I think that a significant amount of work is needed on the manuscript before making it suitable for publication with the "Ocean Science."