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Reply on RC2

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Author comment on "The impact of hydrothermal vent geochemistry on the addition of iron to the deep ocean" by Alastair Jason Mackenzie Lough et al., Biogeosciences Discuss., https://doi.org/10.5194/bg-2022-73-AC2, 2022

The detailed review from reviewer 2 is much appreciated and we are grateful for their efforts in helping to improve this manuscript and reach its full potential. We are happy to make grammatical corrections and amendments to sentences in the revised manuscript.

We are also happy to make adjustments to the structure of the manuscript where it is appropriate to do so to improve the flow. Our rationale for combing the results and discussion was to keep the manuscript relatively short, so it remains more accessible to researchers planning future studies of hydrothermal systems. This is also the format of many recent ocean sciences articles published in biogeosciences.

Comment on Line 17: Throughout the manuscript we refer to distance which is frequently assumed to be a proxy for age*i.e.* the further away from the site of venting the older the age of the plume is (Fitzsimmons et al., 2017; Resing et al., 2015; Saito et al., 2013). As we show with our results from separate casts directly over the vent sites, distance from the seafloor is not a reliable proxy for age. However, on the wider sampling scales of 40 km it is safe to assume that stations further away from the vent sites are sampling older plume waters. We will clarify this throughout in the revised manuscript.

Comment on calculations for Fe/xsHe: We do not feel it is necessary to include equations for integration as this is a common place calculation within this subject and a concept that is usually taught before higher education. We will amend the text to specify that integration is done versus depth. We separated the different ways of calculating Fe/xsHe from the methods section as we decided the methods section should be specific to sampling and analytical methods.

We will delete the repeated phrase and revise the paragraph (starting at L185) to focus on differences in Fe/xsHe caused by the shifting of the plume relative to the position of the ship.

We will change the text to discuss between casts rather than sampling systems and focus the text to clarify that the source of uncertainty comes from the timing of casts rather than the casts themselves as reviewer 2 rightly points out.

In section 3.2 the order of assessments refers to the order used to establish how successful the sampling cast has been in capturing the full extent of the hydrothermal plume over the vent site. Step 1, assessing the sensor profiles is done at sea at the time of sampling hence why it is referred to earlier. Whereas steps 2 and 3 can only be done once the sample analysis is complete, which is why they are introduced in section 3.2.

We will move the findings to the beginning of the paragraph for L325 to L350.

We apologise for this mistake on L365, it should read "Fe/H_2S <1" as FeS_2 nanoparticles will be more prevalent when sulphide is enriched relative to Fe.

We can change the title of section 4.1 to "what controls ridge axis dissolved Fe to helium ratios" to be more specific". The main two points of this section are:

- When we look at our results in comparison to Fe^{2+} oxidation (paragraph 1) and Fe/H_2S of the vents (paragraph 2) these parameters cannot fully explain differences in dFe/xsHe, we therefore suggest these differences are primarily controlled by organic ligands (end of paragraph 2). As we have ruled out the possibility of differences in inorganic chemistry being the controlling factors.
- The dFe/xsHe ratio measured within the ridge valley at the scale of 10-40 km is similar between sites to that used in global biogeochemical models. However, there is significant variability in the appPFe/dFe which has implications for the way hydrothermal Fe is modelled. Which leads into the next section.

We will revise section 4.1 for clarity however given reviewers one and three did not take issue with this section we think a complete re-write is unecessary.

The consistency of dFe/xsHe at the 10-40 km scale is stated at the end of section 4.1 which leads into 4.2. We will move it to the start of section 4.2 for clarity.

We will edit the first paragraph of section 4.2 to emphasise that TDFe/xsHe was higher than dFe/xsHe and we therefore need to investigate the possibility that this difference will persist as plume waters are transported beyond the ridge. As this will impact the values used in global biogeochemical models.

The question mark at the end of section 4.3 will be removed. Two of the four points listed in this sentence (the frequency of vent systems and the variability in the hydrothermal ligand source) are the subject of the preceding paragraph.

Sentences from Line 448 to 452 begin with conclusions based on our repeat sampling at the same vent site which highlights how variable depth profiles of a hydrothermal plume can be between casts taken hours apart. When it is usually assumed that one profile over a vent site is enough to constrain the concentration profile of a hydrothermal plume in the water column. This is an important conclusion for the community and those that will be planning future sampling campaigns. We therefore feel it is prudent to recommend possible solutions to the difficulties of measuring Fe and He together in plumes so that other researchers can consider the technical issues when planning to study hydrothermal plumes using multiple sampling systems.

To clarify the final sentence of section 5, similarity in the near field dFe/xsHe relative to the vent Fe/xsHe shows there is a limit on the amount of Fe released from vents that can be converted into dissolved Fe in the water column. However, as a result of the scatter in the near-field dFe/xsHe we cannot say whether or not the this limit was higher or lower between the TAG and Rainbow vent sites, within the range of dFe/xsHe values measured (4-38 nmol/fmol).

We will revise section 5 in order to make the conclusions clearer.

We will swap table S1 for table 2 in the revised manuscript. Table S1 will now show the number of samples used for the integrations to keep this information separate and avoid confusion.

For figure 1 there are two points on the map at lucky strike, but they are so close together that they appear as one on the map. We will add station numbers in brackets to figure 1 and in table 2 and table S1 so they are more easily relatable. We will change the descriptions to include the distance e.g. change "Close N of TAG" to "29 km N of TAG (S26)"

We will revise the data in tables and report the data to 3 significant figures as requested.

For figure S1, we will corret the typo ("I" = "in") and add further description to the caption that this offset could be the result of differences in the time of day that sampling took place.

The N. Atlantic background value was determined from dMn measurements of waters at the same depth range to the samples collected in this study but from the GEOTRACES equatorial Atlantic (GA03) and western Atlantic (GA02) at open ocean stations away from any margin sources. Background dFe was determined in the same way. We will add text to explain this in the manuscript at L181 where the background values used are stated.

The numbers in the grey boxes of supplementary figures are station numbers not cast numbers. We will correct this in the caption. We will change the labels to the new descriptions e.g. change "Close N of TAG" to "29 km N of TAG (S26)" that will be added to the tables. This should make comparison between these figures and the tables more straight forward.

Technical/Writing and Formatting Issues:

We will go through the manuscript and correct all of the points reviewer 2 has highlighted in their line by line comments and revisit all uses of the terms mentioned to see if more specific phrasing can be used to make it easier to follow the logic.

Line 99: This will be corrected in the revised manuscript.

Line 127: hyphenated "In-house"

Line 137: "Analyzed simultaneously during sample analysis" is will be deleted.

Line 152: hyphenated "near-impossible"

Line 157: "ratio's" changed to "ratios"

Line 158: "off-axis" hyphenated

Line 174: The naming will be changed in the revised manuscript.

Line 179: Changed "We therefore" to "Thusly"

Lines 185-186: We will restructure sentences to separate the TAG and Rainbow data as in the provided example and elsewhere in the manuscript where the term respectively has been used.

Lines 194/195: "down to" corrected to "due to"

Line 197: This sentence will be revised.

Line 202 and 203: We will adopt the example naming strategy throughout to make it clearer when we are discussing differences between methods, sites and sampling casts.

Line 223: We will add the provided example in this instance and look to use more precise language where the term over the vent site is used.

Line 240: This will be corrected in the revised manuscript.

Line 255: "its" corrected to "it is"

Lines 259-262: The mentioned uncertainties cover all of the possible reasons why points on the graph fall above or below the 1:1 line. We will edit the sentence to be more direct removing usage of "this" and "that".

Line 268: "and is" will be replaced with which and the sentence revised.

Line 272: This will be revised

Line 283: We will edit and combine these two sentences to be more succinct as suggested

Line 286: Hyphenated "vent-derived"

Line 290: The outlier in the previous section is in brackets. The "this" refers to the drop in dFe/xsHe between the vent stations at 0 km and the stations in the 10-40 km range which is the main subject of the sentence.

Line 293-295: We will reword the sentence and remove the term observing.

Line 298-300: Log K refers to the oxidation rate constant of Fe(II). We will revise the sentence to "had anomalous Fe(II) oxidation rate constant values (log K),"

Line 309: The difference in the appPFe/dFe ratio, we will revise to make this clearer.

Line 316: This sentence will be revised for clarity

Lines 321-322: Second this will be changed to which.

Line 326: We will go through the manuscript and replace the terms short, distal and near field with specific distances.

Line 336 (and 355): Hyphenated "Fe-binding ligand[s]"

Line 345: Hyphenated "basin-scale"

Line 349: Hyphenated "particulate-dissolved Fe exchange" and "smaller-scale"

Line 355: removed "to"

Line 365: corrected to "molar ratios of $Fe/H_2S > 1$"

Line 372: This sentence will be revised for clarity

Line 387: The residence time refers to the time it takes for tracer lagrangian particles within a physical mesoscale model to exit the ridge valley. This will be clarified in the text.

Lines 389-395: Stokes law was only used once to assess the potential for FeOOH particles to settle out of the plume during further dispersion beyond the sampled 0-40 km range. We will edit the text to make this clear.

Line 406: It refers to both, will change "this" to "these forces"

Line 410: This sentence will be deleted

Line 414: "are key to determining"

Line 415: Revised to "It follows"

Line 428: Corrected to "may be"

Figure 1: This formatting follows the biogeosciences journal word document template

Figure 2, 4, and 5 captions: We will revise the caption and move letters before the description.

Table S1: See previous comments on revising this table, the term values will be removed

Table S1's caption:This section will be revised to for clarity

Figure S2 caption: This will be revised as recommended.

Figures S3-S9: The font sizes of figures will be increased to make them more visible.

Figure S5 caption: This mistake will be corrected

Figure S10: This will be revised as recommended.

Figure S11: The font will be increased to make the numbers clear and caption edited as recommended.

Author contributions: It should be AJML, this will be corrected in the acknowledgements.

Fitzsimmons, J.N., John, S.G., Marsay, C.M., Hoffman, C.L., Nicholas, Sarah L., Toner, B.M., German, C.R., Sherrell, R.M., 2017. Iron persistence in a distal hydrothermal plume supported by dissolved–particulate exchange. Nature Geoscience 10, 195.

Resing, J.A., Sedwick, P.N., German, C.R., Jenkins, W.J., Moffett, J.W., Sohst, B.M., Tagliabue, A., 2015. Basin-scale transport of hydrothermal dissolved metals across the South Pacific Ocean. Nature 523, 200-U140.

Saito, M.A., Noble, A.E., Tagliabue, A., Goepfert, T.J., Lamborg, C.H., Jenkins, W.J., 2013. Slow-spreading submarine ridges in the South Atlantic as a significant oceanic iron source. Nature Geoscience 6, 775-779.