

Biogeosciences Discuss., referee comment RC2
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Comment on bg-2022-73

Anonymous Referee #2

Referee comment on "Tracing differences in iron supply to the Mid-Atlantic Ridge valley between hydrothermal vent sites: implications for the addition of iron to the deep ocean" by Alastair J. M. Lough et al., Biogeosciences Discuss., <https://doi.org/10.5194/bg-2022-73-RC2>, 2022

Summary: This manuscript presents very important new results for dFe (soluble, colloidal), dMn, 3-He (and 3-He excess), and dissolvable particulate Fe from a diverse array of hydrothermal plumes along the North Mid-Atlantic Ridge. There is significant uncertainty regarding the relationships among these parameters at hydrothermal sources, especially the dFe/3He-excess ratio and the complex factors underpinning its systematics. These results promise critical insights that could contribute to better global estimates of hydrothermal-sourced Fe to the oceans, itself a major climatological unknown. The study uses two (or three) methods to derive and calculate Fe/He relationships and has the scope to contribute to several outstanding questions, especially the importance and scale of plume-sampling variability and whether the use of the differentially scavengable element Mn can improve difficult-to-measure Fe/He relationships.

Several challenges prevent this manuscript from reaching its full potential. As written, the text is confusing and disjointed: an overuse of demonstrative pronouns and numerous incomplete sentences consistently interrupt the manuscript's logical flow. The methods section insufficiently describes the ratio-determining methodologies at the core of the findings. The abstract implies a major finding regarding the importance plume age but no such description is given in the text. The conclusions section includes multiple non-conclusion discussion points.

Merging of the results and discussion sections has made the manuscript especially difficult to unpack. It has forced the authors to address important discussion points (comparing the methodologies to each other and their wider context) alongside more results-specific points (variability in the plumes-as-sampled, inter-cast differences, and results-specific caveats like rising plumes). Basin-scale implications and source-fluid specific discussion points (especially the latter, being sourced from previously published results) would be better-suited in a dedicated discussion section or two, i.e. after inter-site results and inter-method differences have been presented and resolved. The "Wider implications" section 4 attempts to serve as a discussion section in many ways, but at present does so inadequately.

I attempt to address the major points in the sections below. Technical/writing points are broken out separately at the end of the comment.

Abstract: Line 17: "plume age" is mentioned as the primary driver for the ranges in Fe/He ratios, but 'age' is not once mentioned in the manuscript body. If distance from vent source or some other combination of parameters is being used as a proxy for age, either implicitly or explicitly, these relationships must be clearly stated and explained.

Methods: The "integration methods" and their relevant calculations and equations (especially those for Methods 1 and 2, but also perhaps 3) should be defined, presented, and described at some point in the Methods section proper. At least, a summary of their external sourcing and relevant assumptions should be given before the Results section begins. "Integration", I presume, is being done versus depth, but this point is not explicitly stated. Omission of these methods—or their mis-placement in other sections—makes it difficult for future users to derive their own results using Methods 1 and 2, and also to test and confirm the results being presented here.

Paragraph at Line 185: "despite the ship maintaining" is repeated twice in this paragraph, and this redundancy highlights that the purpose and ordering of this section is a bit confused. Is this paragraph serving to discuss the differences between/among the sites or the differences in the two methods? It seems to be trying to both, so consider revising this paragraph (and the ones nearby) to have clearer intent.

Specifically, it would help to discuss the inter-site differences, inter-method differences, and inter-cast/sampling system differences independently and in turn. Related to the final one, it might be clearer throughout the manuscript to refer to differences "between casts" or "between casts at the same station" rather than between "sampling systems". Different casts that are separated by time (even those using the same sampling system) will face plume-position issues. The cast's timing, rather than the system itself, is thus the source of the uncertainty for these purposes. Inter-"system" differences would be more appropriate terminology when focusing on potential contamination- or blank-related results that differ between sampling systems, i.e. TMR vs S(S)R. Many of these topics would be best presented in a dedicated results section (or even in the methods if they can be methodologically caveated).

Section 3.2: The ordering of assessments introduced here (LSS/ORP, Mn/He, Fe/He) does not match the order they are then presented and discussed in the text that follows, unless the earlier mention of LSS/ORP in Figure 3 is being back-referred to. This introductory sentence would make more sense if moved earlier in the manuscript.

Section 4 intro paragraphs (4.0?): Despite the title of this section, the introductory paragraphs (lines 325 to 350) do not significantly discuss wider "implications" of the manuscript's actual findings, at least until the very end (beg. Line 344) and then only briefly. Most of this text serves more to introduce/revisit previously published field

findings and different model designs being debated in the literature. While this text is not without merit, it might be helpful to number and rename this set of paragraphs as its own subsection (“4.1 Expectations based on prior results”), or at least move the major findings (beginning at Line 344) to the beginning and then follow through with discussing their implications more directly, as the section title promises.

Section 4.1: I had difficulty following the logic of the first two sentences beginning at Line 365. TAG has the lower vent fluid Fe/H₂S ratio—why should it precipitate more FeS₂ if ratios greater than 1 are more likely to precipitate more FeS₂ (as stated in the previous sentence)?

The section is titled “What controls near-field dissolved Fe to Helium ratios [?]”, but by the end of this section I did not feel the text had convincingly and logically addressed that question. Instead, the section presents a list of hypotheticals and loosely-connected points that dances around the findings.

Section 4.2: The major scientific conclusion of this section is that the fairly consistent 10–40 km-distance Fe/He relationships observed in Fig 5 are likely to be reflective of Fe/He ratios leaking out of the ridge into the deep N. Atlantic. State this from the very beginning, then justify it. The points summarizing these findings are not scientifically unsound as written, but they are surprisingly roundabout. The flow of the writing here is difficult to follow as the (very long) paragraph meanders amongst discussion of various assumptions, results/findings, hypotheticals, and conclusions.

The topic sentence emblematically does not clearly state or frame any of the important points regarding Fe escape from the ridges that follow. Relatedly, the final, critical sentence in this section (and potentially the paper as a whole!)—that a significantly higher Fe/He ratio may be called for in global models, does not, at present, read as directly or well-supported by the lines preceding it.

Section 4.3: The concluding sentence here is a list of wide-ranging unknowns punctuated incorrectly by a question mark. It does not especially summarize or clarify any of the preceding points about future work.

Conclusions: Sentences from Line 448 to 452 are hypotheticals about future sampling systems—not conclusions. The final sentence of the paper is especially confusing—are the findings of this paper implying different or similar Fe limits at the various sites? Consider replacing most of this section with a clearer, more concrete summary of the actual conclusions reached as a result of this paper’s new data, not hypotheticals based on what-ifs.

Table 2: This table was a bit unintentionally confusing: only half the values tabulated are

Fe/He ratios as labeled in the header. The other half are *n* values, which themselves are separated via colons (confusingly suggesting they are ratios). I suggest revisiting the table and column labeling to increase clarity. The organization of Table S1 is clearer in some respects as it groups like datatypes together (ratios, statistical parameters) allowing the two methods to be more easily compared.

It is not possible to use Figure 1 to unambiguously determine which stations are which in Tables 2 and S1. In the case of the Lucky Strike site, for instance, only one station label is shown in Figure 1, but two rows (stations/casts?) are listed in Tables 2 and S1. Consider using some combination of station/cast numbers to unambiguously label Figure 1 and both tables. Relatedly, the descriptive labels in the lower half of Table S1 (e.g. "Close N of TAG") read as overly subjective, even for a supplement.

The value of 35 nmol/fmol for Rainbow 38 (the instance with no asterisk) differs from the matching row reported in Table S1 (34 nmol/fmol). This is likely a significant figures issue? Given the importance of these data, I suggest revising both Tables 2 and S1 to ensure a consistent number of significant figures (three?) for the reported Fe/He ratio values.

Figure S1: The difference between dMn measurements ("I" = "in"?) for "surface" waters at station 25 appears at several points to be much greater than 0.2 nM, especially in the upper 100 m. I agree these differences are not especially significant at depths relevant to the manuscript, but more care needs to be taken here in describing this offset (inter-cast differences? Time of day of the casts?)

Figure S3-S9: How was the "N. Atlantic background value" for dMn determined? It's fine if it's just a "typical background" value, but the rationale should be stated somewhere.

Are the very small numbers in the grey bars atop each subfigure cast numbers or station numbers? The captions seem to imply they are cast numbers ("Main casts are..."), but the main text (Table 2) refers to them as stations. Be consistent. Either way, the numbers listed in the captions and figures here are not enough to identify which site is which (e.g. 12-15; 26, 27?) without forcing the reader to cross-reference other tables/text. Consider grouping/labeling all these supplemental sub-figures by site.

Technical/Writing and Formatting Issues:

As described in the summary, the text as written suffers from an over-use of

demonstrative pronouns ("this" "these"), especially at the beginning of sentences. I have tried to point out important instances in my line-specific comments, but the authors should revisit all uses of these terms and revise to use more specific phrasing whenever possible. Relatedly, the number of incomplete sentences and disconnected clauses is high, making it difficult to follow the logic at many points.

Line 99: Missing apostrophe in "plumes" (should be either plume's or plumes')

Line 127: Missing hyphen: "In-house"

Line 137: "Analyzed simultaneously during sample analysis" is redundant.

Line 152: Missing a hyphen: "near-impossible" (or "nearly impossible")

Line 157: "ratio's" should be "ratios"

Line 158: "off-axis" should be hyphenated

Line 174: For consistency, and even though it is not discussed at length, this third method should also probably be named (e.g. "³He-interpolation method"), rather than just "A third method..."

Line 179: Consider changing "Thusly" to "We therefore"

Lines 185-186: The word "respectively" could be interpreted as referring to either the two different integration methods or the two sites. Consider restructuring this sentence to reduce ambiguity, e.g. "...ranging from 4 to 87 at the TAG site and 4 to 63 nmol/fmol at Rainbow site."

Lines 194/195: "down to" is an awkward construction "due to" it implying depth in the ocean. Use a different term.

Line 197: Incomplete sentence at "As it can account..."

Line 202 and 203: "over the vent sites" is somewhat unclear—do you mean "across different vent sites"? "Over the vent sites" sounds like a ship position/cast difference issue, as implied by the concluding words "...when comparing different casts". As pointed out in my comment re: paragraph at Line 185, it would be best to address methodological (Method 1 vs 2), inter-site (TAG vs Rainbow), and cast/sampling system differences (TAG TMR vs TAG SR) independently and systematically.

Line 223: "over the vent sites" is again a confusing phrase here, as it implies the ship's position. It might be clearer to say "The extent to which any collected sample is representative of the core..." (or similar)

Line 240: Agreement issue: "Extrapolation...indicates"

Line 255: "its" should be "it is"

Lines 259-262: Comma usage in this sentence is confusing. Is "uncertainties associated with" relevant to both "vent fluid end-members" and the degree of removal of dMn at the two sites? In the following sentence, usage of "this" (and later "that") are unclear about what is being referenced: the sub-1:1 relationships in general, or the specifics listed at the end of the previous sentence?

Line 268: "and is" (singular) doesn't agree with the first half of the sentence.

Line 272: Agreement issue: "site-by-site differences...was not simply"

Line 283: Consider being more specific about these spatial scales right off the bat: "Over short spatial scales of under 40 km from the vent site, ..." The sentence that follows is largely redundant.

Line 286: Hyphenate "vent-derived"

Line 290: To what does "This [indicates]" refer? The outlier discussed in the previous sentence?

Line 293-295: As written, this sentence wrongly implies that the Fe/He ratios are "observing" the wide range of TDFe concentrations, rather than the authors.

Line 298-300: "West" is unnecessarily capitalized. Without additional information, it is not clear what "log K" specifically refers to here. Again, the usage of "This [could explain]..." is confusing and needs to be clarified.

Line 309: What does "This [highlights]..." refer to? The cFe/dFe ratios, or the appPFe/dFe ratios? (Or both?) Revise to be clear.

Line 316: The sentence beginning "Specifically..." is incomplete.

Lines 321-322: Two sentences in a row begin with "this" here, making the logic difficult to follow.

Line 326: "short" spatial scales is rather non-specific. Consider replacing or supplementing with the actual spatial scales (0- or 10-40 km, I presume). Also, the phrase "short spatial scales between the TAG and Rainbow plumes" implies the distance between the TAG and Rainbow sites rather than the distances over which each site's plume was sampled.

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

Line 345: Hyphenate "basin-scale"

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

Line 355: "strength to" is an incomplete thought/clause

Line 365: Try "molar ratios of Fe/H₂S >1"

Line 372: The sentence beginning "Suggesting..." is incomplete.

Line 385: The sentence beginning "Showing..." is incomplete.

Line 387: Hyphenate "Fe-rich". Explicitly state or otherwise clarify the "residence time" being referred to from the Vic reference.

Lines 389-395: How many times was Stokes' law actually used by the authors here? Twice it is mentioned, but only one calculation is discussed (though not explicitly shown), which reads as redundant.

Line 406: "This [would counteract]..." is unclear: which of the previously listed items (or all?) is being referred to?

Line 410: "This [creates]..." is unclear.

Line 414: Multiple things are listed and then referred to as "...is key to determining"

Line 415: "This [highlights]..." is, again, awkward and difficult to follow.

Line 428: "maybe" should be "may be".

Figure 1: The text labels on many of the sub-figures are on the smaller side. Font size may need to be increased for publication.

Figure 2, 4, and 5 captions: It would be clearer if the figure sub-identifiers ("(a)", "(b)"...) were listed before (rather than after) their respective text descriptions. Currently, succinct descriptors and ancillary information are mixed throughout the caption text, making these captions hard to follow.

Table S1: "Plume-integrated" should probably be hyphenated. The word "values" in the header is extraneous. The vent sites in this table (cf. comments re: Table 2 in the main text) lack station numbers and cannot be identified in the Figures or directly compared to other tables.

The text of the first paragraph of Table S1's description confusingly uses "this" several times, referring to different things each time. Please revise to be more precise. The text of the second paragraph also needs revision. Specifically, "The estimated ratios from interpolation are 'so variable'..." reads as overly subjective, and the next sentence ("Largely because of the variability...") is long and awkwardly constructed.

Figure S2 caption: "Demonstrating..." is not a complete sentence. Merge this fragment with the topic sentence here—it is the major take-away.

Figures S3-S9: Much of the in-figure text, with the exception of the y-axis labels, is very small. The font sizes may need to be enlarged for publication.

Figure S5 caption: This caption mistakenly refers to Figure 3A, but the relationships discussed are shown in Figure 4A.

Figure S10: There is a missing close parenthesis in the first sentence of the caption. Spell out "three" or use "n=3" in this sentence as well. Remove the redundant "over Rainbow" from the second sentence.

Figure S11: Font size in the grey boxes is much too small to be legible. Consider changing "which is what the main text focuses on" to something less chatty, e.g. "as are the focus of the main text".

Author contributions: The lead author is referred to as both AL and AJML.