Dear editor,

Hoffman and Monreal et al. produced a highly valuable dataset and manuscript on Fe-binding ligands and microbial communities in hydrothermal (HT) plumes at sites over the Mid-Atlantic Ridge. The manuscript has a focus on siderophores, and particularly the genetic techniques targeting siderophore-production related transcription are novel and influential. In general, the manuscript is a pleasant read, with little to remark in terms of editing.

Having said that, this reviewer must express some puzzlement on the particular focus of the manuscript on siderophores alone, given the broad and detailed dataset. In the supplement, the authors detail a comprehensive set of results representing multiple ligand classes. However, the manuscript fully omits all but the strongest ligand class, and seeks to correlate this subset to siderophores specifically, which in turn is an exceedingly small subset of the prior. In the introduction it is stated that the authors seek to explain siderophores specifically for the first time in HT systems (line 75). However, much of the manuscript proceeds to discuss the siderophores as the main component of the ligand pool at these sites, with strong implications for ocean systems extrapolated. This stands in contrast to the admitted maximum 4% representation of this ligands class by the siderophore fraction in the present dataset, and while underestimation is (rightly) expected, firm assertions made here do not seem sufficiently supported by the depth of analysis of the dataset presented here.
It would therefore be my suggestion to either revise the manuscript to widen the scope of the analysis to the entire width of the dataset (i.e., include L2 and L3 classes and describe their roles more completely, as to further motivate the assertions made on the L1 class), or walk back the assertiveness of statements to their implications for the specific parts of the microbial community these relate to, as opposed to Fe cycling in bulk.

More detailed responses to specific parts of the manuscript follow, in the hope these will explain these concerns more fully.

As a secondary and smaller scale remark, citations in the manuscript can be considered a little light on diversity, and it would be a credit to the authors if they were to cite the trace metal / ligand research community a bit wider.

I would strongly support publication of this dataset, as it is of great value and quality. However, I would recommend revision of the manuscript to refocus it to better leverage the dataset's wider merits.

Line 76
'has been proposed' does burden this statement with a requirement of citation

Line 89-91
Does mere presence indeed imply the gravity of the role?

Line 102-103
I see no dFe-ligand ratios, or ligand concentrations for that matter, in Lough et al. 2022.

Line 104~
When coupling was lacking for L2 and L3 – was L in excess of dFe? From table S2 it looks like this was the case for all but vents 'BS' and 'TAG', where dFe was found in excess. This would merit more discussion. The weaker ligands are brought into relation with inorganic speciation, are Fe-oxy-(hydr)oxides implied? Would these reasonably be reflected by L2+? How? Or particulate inorganics? Please elaborate.

Line 111-112
Surely prior work can be reflected in a broader sense...

Line 113-121
In summary – L1 ligands did not correlate with the HT fluid proxy, weaker ligands did, but did not correlate with dFe, as stated in the prior. Were they in excess? The weaker ligands would benefit from more discussion here, these groups do not 'cap' dFe, but they surely remain relevant to its cycling. This section proceeds to discuss L1 ligands more or less alone and in terms of local microbial production, is this supposition staved by microbial presence?

Re: distance from the nearest vent – it would be helpful to have this as part of table S2, as the reader is now tasked to work this out from S1 and Figure1. The correlation or lack thereof with for the different ligand classes would benefit from elaboration as well in order to better support the assertions in section 2.1

159-161
"compelling evidence that microbial production of ligands is responsible for at least some portion of the tight coupling between L1 and dFe"
A strong statement, though this reviewer feels that *compelling evidence of coupling* on the one hand and *'...some portion...'* at max 4% representation of the L1 pool on the other are at odds...
More comprehensive discussion is warranted here before this statement is made thusly. The identification of siderophores, their probable underestimation from the summed totals, and their role exemplifying (local) microbial contributions to the ligand pool are all worthwhile, but this extrapolation is too reductive.

Line 213
Please cite literature to reflect siderophore production in situ at HT plumes being believed to be less widespread than found here.

241
"...will aid in constraining the biogeochemical importance of microbial feedbacks in impacting the hydrothermal dFe supply to the deep ocean."
This sentence seems overly complicated. Please clarify what is meant with (...) *the biogeochemical importance (...) in impacting (...)*

Lines 257-271

What equilibration time was employed for these analyses? If longer or overnight, has there been any measurement to ascertain if the formation of $Fe(SA)_2$ was a factor at the 10nM SA addition level and an influence on these measurements?