

Response to Reviewer #1:

This short paper provides 18 $\delta^{56}\text{Fe}$ in chiton (marine molluscs living in the near shore coastal environment) that accumulate Fe biominerals in their radula's teeth. Different species have been analysed (including two from the same site) at 4 locations (2 in the Atlantic, 2 in the Pacific). The authors compare their values with published seawater data, even though they are far away from the chitons sampling time and location.

Data seem to be of good quality and the article is generally well written and illustrated. The authors acknowledge honestly several times that their study is preliminary. There exist indeed few data on $\delta^{56}\text{Fe}$ in marine environments so any new dataset is welcome. This is why I would recommend publication.

1. I do however have a significant concern regarding the discussion when the authors try to interpret the differences in $\delta^{56}\text{Fe}$ measured in these samples. They present three speculative hypotheses in an imbalanced way: e.g. the feed regime is preferred to explain the differences in two species from the same site whereas this hypothesis is not better supported by data from this paper or from previous studies. In the main text, the three hypotheses should be equally discussed (as in Fig.3). Organising the discussion into sub-sections would help. Abstract and conclusions should be modified accordingly. Some other points could be also clarified (e.g. seawater sites).

We agree with the reviewer that in the original manuscript we emphasized the feeding hypothesis, although the other hypotheses discussed are equally possible. In the revised manuscript we will address this imbalance and we will explicitly point out that at present the data do not favor a particular pathway.

My detailed comments are listed below.

2. Title: add in brackets after "...in marine invertebrate (chitons, mollusca)..." to have more accurate information on the study which is very restricted to this type of invertebrate only.

We agree with the reviewer that the proposed title is more accurate and it will be changed accordingly in the revised manuscript.

3. Abstract and conclusions should present the three hypotheses to interpret the data.

We agree with the reviewer that this would be informative. In the revised manuscript we will briefly summarise the different hypotheses in the abstract and conclusions.

4. P.5537 line 25. Please provide a range of sample size: how much dry weight of radula and Fe have been processed for a single analysis?

The amount of Fe in the analyses ranged from 30 μg to 840 μg and this will be mentioned in the methods section of the revised manuscript.

5. P.5538. Specify whether Apex has been used with or without membrane.

We used Apex without a membrane. This will be mentioned in the Methods section of the revised manuscript.

6. P.5539 Line4:typo: delete one 'the'.

This will be corrected in the revised manuscript.

7. P5542,Line8:typo: 2σ instead of $y\sigma$.

The reported values are in fact for 2σ . We will correct the typo in the revised manuscript.

8.P 5542 and 5543 “Assuming that the isotopic difference between *T. lineata* and *M. muscosa* does indeed reflect their contrasting diets” is indeed highly speculative, so going a step forward, i.e. finding an explanation on why red algae would have a different isotopic signature than green algae, goes very far since no algae data were measured. Similarly P. 5543 can the authors provide a reference to support this statement: “relatively high Fe(II) concentrations in the eulittoral zone and low Fe(II) concentrations in the sublittoral zone.”?

We agree that this section is speculative but we think the discussion is worthwhile; it has long been known that differences in oxygen and carbon isotopic signatures exist between red and green algae (Anderson, T.F. and M.A., Arthur, 1983. Stable Isotopes of Oxygen and Carbon and Their Application to Sedimentologic and Paleoenvironmental Problems. In: Stable Isotopes in Sedimentary Geology, Arthur, M.A., T.F. Anderson, I.R. Kaplan, J. Veizer and L. Land (Eds.). SEPM, Short Course Notes Vol 10, Georgia, pp: 1-151.). These represent 'vital' kinetic fractionation effects and it is conceivable that such differences also may exist for Fe isotopes.

*As for Fe(II) concentrations in the littoral and sublittoral zones, measurements in marine settings show a steep reduction in Fe(II) concentrations in the top 10 m of the water column, and this is attributed to light attenuation and the drop in photoreduction of Fe(III) to Fe(II) (e.g., Shaked, Y. (2008). Iron redox dynamics in the surface waters of the Gulf of Aqaba, Red Sea. *Geochimica et Cosmochimica Acta*, 72(6), 1540-1554). However, we cannot be certain that this is also the case in the Washington site in our study. In the revised manuscript we will include additional citations and rephrase this sentence to make it more cautious.*

9. Fig. 2. What are the distances to the chitons' sampling sites the locations of SW signatures d) and e)? Unclear also if 500km apply for a,b and c. See also my comment on table 1 where this information could be provided.

To clarify, we will add a reference in the text reporting the Bermuda SW sampling location [John, S. G. and Adkins, J. F.: The vertical distribution of iron stable

isotopes in the North Atlantic near Bermuda, *Global Biogeochem. Cycles*, 26, GB2034, doi: 10.1029/2011GB004043] and modify the figure caption as follows in the revised manuscript:

Blue squares are published surface seawater isotope analyses of dissolved Fe from locations as close to the chiton sampling sites as available data permits. Data reported by Rouxel and Auro, (2010) for three sites located off the north-eastern Atlantic coast of North America are (a) Vineyard Sound on Cape Cod, Massachusetts, USA (-0.82 ‰); (b) Waquoit Bay on Cape Cod, Massachusetts, USA (-0.55 ‰); (c) Connecticut River estuary in Long Island Sound, Connecticut, USA (+0.04 ‰). These three sites are located within less than 150 km distance from each other, on average about 500 km south of the chiton sampling site at Grand Manan Island, New Brunswick, CA). Data for the North Atlantic (d) (+0.3 ‰; sampled about 100 km southeast from Bermuda, John and Adkins, 2010; John and Adkins, 2012) are compared with the Bermuda chiton sampling site. The closest available coastal seawater Fe isotope data to compare with the Puget Sound chiton sampling site (Washington, USA) is from the San Pedro Basin (e) (0 ‰; John and Adkins, 2010), which is located off the Atlantic coast near Los Angeles (California, USA), about 1500 km south from Puget Sound.

10. In Fig.3 Here, the three potential processes yielding to Fe isotopic fractionation are presented in an equal way. The legend correctly underline that those 3 mechanisms are not mutually exclusive. However, in the text, mechanism a (and in a less extent, b) is preferred and this is imbalanced. This is especially true in abstract and conclusion, where only hypothesis involving diet is provided.

We agree with the reviewer that there is a certain imbalance in the discussion of the different fractionation pathways. In the revised manuscript we will correct this imbalance and we will point out that at present the different pathways are equally possible.

11. Text on Fig.3 is too small.

This will be corrected in the revised manuscript.

12. Table1. Provide exact locations: latitude and longitude of sampling sites.

We agree with the reviewer that more information would be useful. Unfortunately, the exact sampling locations were not recorded when the samples were stored and archived at the Peabody Museum.

13. Provide also locations and $\delta^{56}\text{Fe}$ of seawater data (a,b,c,d,e) that are compared with chiton isotopic signatures.

This will be included in the revised manuscript.

14. Eventually four small maps of each site could be helpful.

Again, we agree that this would be useful but the exact sampling locations were not recorded.