nature portfolio

Peer Review File

Cycling and persistence of iron-bound organic carbon in subseafloor sediments

REVIEWER COMMENTS

Reviewer #2 (Remarks to the Author):

This work explored the persistency of reactive-iron associated organic carbon and its behavior in continental slope sediments, then updated the global budget of FeR-OC. This work is significant. Some statements, however, need more explanations or calculation to support. See the comments below for details.

Line 23: a comma is missing.
Line 29: use the right format of scientific notation here and throughout the text.
Line 45: add “an” before “active electron acceptor”.
Line 58: “Nevertheless” is odd here and can be removed.
Line 109: Fig. 2a is not cited yet.
Line 117: provide a number or a range to show how low it is.
Line 123: the average content of FeR-OC of 0.03% is contradictory to the statement on Line 119 where the minimum FeR-OC content is 0.04%.
Line 125: the average FeR-OC/DOC of 5.2% is is contradictory to the statement on Line 120 where the minimum FeR-OC/TOC is 10%.
Line 141-146: I agree with the authors that microbial activities are the most likely reason for the phenomenon that in both cores, lower values are observed in SMTZ sediments. But I don’t follow how a glacial-interglacial cycle has something to do with the reasons proposed for the lower values. More explanations are needed here.
Line 244: what is non-FeR-OC? Why is FeR-OC with a percentage of 10-20% in TOC preferentially remineralized? I don’t get the point. More explanation is needed.
Line 269-272: using the numbers provided here, “a substantial fraction” the authors claim is less than 4%. Is this “substantial”?
Line 288: Based on the molar ratio of FeR-OC to FeR and the two mechanisms, the proportion can be estimated to support “a large proportion” the authors proposed. Otherwise, the proposal is weak.
Line 329: “shelf” should be “slope”?
Line 331: change “is” to “are”.
Line 331-334: a conjunction is needed for the two sentences.
Line 354 & 357: provide the number of data to be averaged.

Reviewer #3 (Remarks to the Author):

The manuscript investigated the long-term variations in content and isotope of TOC and FeR-OC, emphasizing the impact of SMTZ in FeR-OC preservation. Based on the basically unaltered FeR-OC/TOC ratio long-buried sediments, this study provided an estimation of global budget of FeR-OC in Quaternary marine sediments. The topic is new and innovative, and experimental and analytical designs are convincing. The results provide an important experimental basis for understanding the role of sedimentary FeR-OC in regulating long-term carbon cycle.

1. Title, the current title could not be completely reflect the topic and the present study, such as the items of digenesis and microbial community.
2. Before the process of SMTZ, how the FeR-OC change and/or response towards the reduction process of dissimilatory Fe and Mn reduction?
3. The impact of SMTZ in FeR-OC preservation was more significant in the sediment core QDN-14B nearby cold seeps due to the more abundant microbial occurrence; however, this degradation mechanism could not be suitable for the slope sedimentary setting, thus is the microbial evidence also be increased in the normal slope core with SMTZ section? In addition to the support from the remineralization of FeR-OC, what is the role of methane release for this specific microbial community structure?
4. Line 143-154. As the authors mentioned, the sediment provenance could alter the characteristics of FeR-OC and TOC. Although the impacts of provenance were not apparent in the studied two cores located in continental slopes, provenance was non-negligible factor in the shelfs due to exposure and submersion cycles on glacial/interglacial timescales. Therefore, is the extrapolation of the stable FeR-OC/TOC ratio to shelf and delta/estuary reasonable or rigorous?
5. Lines 212-216. Re-adsorption of isotopically depleted DOC to Fe was considered to result in the negative δ13CFeR-OC values and low FeR-OC content. From the perspective of geological evolution, the sediments below the SMTZ have experienced the Fe reduction and DOC re-adsorption before burial in present depth, but the FeR-OC content and isotope characteristics are similar to those of sediments above SMTZ. Did some isotopically enriched OC adsorb to the FeR after passing through the SMTZ? If so, some of the FR-OC maybe not only formed at the redox interface as proposed in lines 288-289.
6. Line 293-319, 339-360, the global estimation of Fe-OC over a larger spatial and temporal scale should be important, however, much of these explanation and discussion as mentioned here could be shorten to make the draft more focusing.
7. Table 1 present clear comparisons of content and carbon isotope between non-SMTZ and SMTZ. Statistical analyses (e.g. one-way ANOVA) are recommended to guarantee the reliability of the comparison, just like you did in Figure 4.

Reviewer #4 (Remarks to the Author):

Chen et al. present a pair of detailed short-core datasets which they use to use to investigate the coupling of FeR and OC in marine sediments. This is a nice study, and looks to be scientifically sound. The findings are generally quite interesting, especially the suggestion that microbial remobilisation of FeR occurs in the SMTZ. However, I feel other than these data, the manuscript does not represent enough of a step forward to warrant publication in Nature Communications in its current form. My reasons for this are as follows:

1. The title, and the abstract discuss 'geological timescales', while the term 'long-buried sediments' is used in line 103. However, a study which investigates 100 kyr does not fit the bill to me. Persistence of FeR -OC over much longer timescales (up to tens of millions of years) has now been

demonstrated in numerous publications (Faust et al., 2021; Longman et al., 2021, 2024). Some of these studies have not been cited in this work, so emphasising this as an aspect of novelty seems wrong. I would adjust the title and the primary narrative of the manuscript to better reflect the more interesting data – what goes on through the SMTZ and below, and interactions of FeR and OC during burial.

2. The compilation of data for the section discussing the scale of the marine sedimentary FeR-OC sink is lacking a large number of studies. There was a recently published updated dataset of FeR-OC in such environments (Longman et al., 2022), which cites a lot of work not considered here, and a lot of environments not considered. Furthermore, the existence of that study, which provides estimates of the amount of FeR-OC buried every year, means this study as presented does not represent something fully novel.

3. What is novel is trying to estimate the total FeR-OC pool in quaternary sediments. However, I cannot see the reason behind making this calculation. The authors are aiming to estimate the amount of sediment which is microbially active, but the only microbial activity they demonstrate to have an impact on feR-OC is in the SMTZ. As they themselves show, this zone in limited, and does not represent the entirety of quaternary marine sediment. The cores studied here have SMTZ thicknesses of only up to a metre. Even in this study they only represent around 15% of the sediment. Outside of this there is no evidence for variability in FeR-OC driven by microbes. I really don't think any sort of extrapolation of FeR-OC based on the LaRowe estimate of Quaternary sediments makes much sense when trying to estimate what is impacted by microbes in the SMTZ. I would suggest a better way to investigate this would be to focus on the SMTZ – I am no expert on this, but are there estimates of thickness available globally?

4. However, even if the adjustment to reflect only the SMTZ is made, the fact that data show the FeR-OC values are similar above and below the SMTZ in one core (and likely in the other if deeper samples were available) suggests nothing of much consequence is ongoing. Yes, there may be some remineralisation, but overall if we are indeed concerned about the long term sink, it doesn't look like the SMTZ and microbes really affect this. This is definitely not an issue, but framing it as a finding of importance makes little sense to me.

5. I'm not sure where the links to 'deep life' come from. Yes, FeR-OC can be a large labile reservoir of OC, but what suggests deep life? I assume the microbiology results? If so, make the link here clearer. However, if it is the microbiology, I see no evidence for this – you see some really interesting microbial activity in the SMTZ, but nothing to suggest this goes on in the 'deep' subsurface. Also, I don't think 900 cm is really 'deep'. There's papers which discuss truly deep subsurface microbial life (e.g. Inagaki et al., 2015), which show microbial activity down to more than 2.5km (250000 cm).

I'm sorry to seem so negative about this. The study is certainly interesting and the combination of FeR-OC measurements with microbiology, and everything going on in the SMTZ is interesting. However, I think the novelty is not there for this journal – the scaling up is unsuitable and the regular discussion of geological timescales seems wrong to me.

References.

Faust, J. C., Tessin, A., Fisher, B. J., Zindorf, M., Papadaki, S., Hendry, K. R., Doyle, K. A., & März, C. (2021). Millennial scale persistence of organic carbon bound to iron in Arctic marine sediments. Nature Communications, 12(1), 1–9. https://doi.org/10.1038/s41467-020-20550-0

Inagaki, F., Hinrichs, K.-U., Kubo, Y., Bowles, M. W., Heuer, V. B., Hong, W.-L., Hoshino, T., Ijiri, A., Imachi, H., Ito, M., Kaneko, M., Lever, M. A., Lin, Y.-S., Methé, B. A., Morita, S., Morono, Y., Tanikawa, W., Bihan, M., Bowden, S. A., … Yamada, Y. (2015). Exploring deep microbial life in coal-bearing sediment down to ~2.5 km below the ocean floor. Science, 349(6246), 420–424. https://doi.org/10.1126/science.aaa6882

Longman, J., Faust, J. C., Bryce, C., Homoky, W. B., & März, C. (2022). Organic Carbon Burial With Reactive Iron Across Global Environments. Global Biogeochemical Cycles, 36(11), e2022GB007447. https://doi.org/10.1029/2022GB007447

Longman, J., Gernon, T. M., Palmer, M. R., & Manners, H. R. (2021). Tephra Deposition and Bonding With Reactive Oxides Enhances Burial of Organic Carbon in the Bering Sea. Global Biogeochemical Cycles, 35(11), e2021GB007140. https://doi.org/10.1029/2021GB007140

Longman, J., Manners, H. R., Gernon, T. M., McManus, J., Palmer, M. R., Rowland, S. J., & Sutton, P. A. (2024). Production and preservation of organic carbon in sub-seafloor tephra layers. Marine Chemistry, 258, 104334. https://doi.org/10.1016/j.marchem.2023.104334

Response to reviewers' comments on 'Reactive iron as an important reservoir of marine organic carbon over geological timescales.'

We would like to thank the editor and reviewers their helpful comments and suggestions, which helped improve the manuscript significantly. We have made considerable efforts to address all the concerns raised.

In summary, we have refocused our manuscript and emphasized the remobilization of Fe_R-OC in SMTZ sediments and its impact on subseafloor microorganisms with extra statistical analysis and calculations. The estimation of Fe_R-OC reservoir has been largely shortened, but the dataset used has been updated and extra statistical analysis has been done. Please find below our responses to the specific comments. Line numbers refer to lines in the clean version.

Reviewer #2 (Remarks to the Author):

This work explored the persistency of reactive-iron associated organic carbon and its behavior in continental slope sediments, then updated the global budget of FeR-OC. This work is significant. Some statements, however, need more explanations or calculation to support. See the comments below for details.
Response: Thank you for the overall positive evaluation of our manuscript. Please see below our responses to detailed comments.

Line 23: a comma is missing.
Response: Added.

Line 29: use the right format of scientific notation here and throughout the text.
Response: We have changed all the OC reservoir-related unit to Pg throughout the text.

Line 45: add "an" before "active electron acceptor".
Response: Added.

Line 58: "Nevertheless" is odd here and can be removed.
Response: Removed.

Line 109: Fig. 2a is not cited yet.
Response: Thank you for pointing this out. The description of Fe_R content record is now added to the revised manuscript and the Fig. 2a is cited. (Lines 124-126)

Line 117: provide a number or a range to show how low it is.
Response: The numbers are now provided in the revised manuscript. (Line 127)

Line 123: the average content of FeR-OC of 0.03% is contradictory to the statement on Line 119 where the minimum FeR-OC content is 0.04%.
Response: Thank you for noticing. The inconsistent data description has been corrected

throughout the text in the revised manuscript. (Lines 128-129)

Line 125: the average FeR-OC/DOC of 5.2% is is contradictory to the statement on Line 120 where the minimum FeR-OC/TOC is 10%.

Response: Thank you for noticing. The inconsistent data description has been corrected throughout the text in the revised manuscript. (Lines 128-129)

Line 141-146: I agree with the authors that microbial activities are the most likely reason for the phenomenon that in both cores, lower values are observed in SMTZ sediments. But I don't follow how a glacial-interglacial cycle has something to do with the reasons proposed for the lower values. More explanations are needed here.

Response: We have added more information to the introduction, which explains how glacial-interglacial cycles might influence sedimentary Fe_R-OC records:

> "Additionally, the supply of Fe_R-OC is expected to depend on the hydrological conditions through the influence on continental weathering and Fe_R formation as well as on sea level through the influence on shelf topography and thus transport of continental detritus into the deep sea[25-27]; both factors are expected to vary periodically on glacial-interglacial timescales." (Lines 65-69)

At the same time, we have de-emphasized the role of glacial-interglacial variations. Given that the chance is extremely small that the lower Fe_R-OC concentrations in the SMTZ are controlled by the depositional history and coincidentally located in both cores in the SMTZ horizon, we have added the following explanatory statement to the revised manuscript:

> "While we cannot entirely rule out that these distinct signals in SMTZs of both cores are related to the depositional history and coincidentally located in this horizon, we view this scenario as highly unlikely." (Lines 179-182)

Line 244: what is non-FeR-OC? Why is FeR-OC with a percentage of 10-20% in TOC preferentially remineralized? I don't get the point. More explanation is needed.

Response: We have added the explanation of "non-Fe_R-OC" as "OC that is not bound to Fe_R" in the revised manuscript (Line 269-270). We assume that the Fe_R-OC is preferentially remineralized over OC that is not associated with Fe_R in SMTZ, because Fe_R-OC only accounts for 10-20% of TOC but contributed 36.7% to the TOC mineralization flux.

We have rephrased the sentences to address this point:

> "Fe_R-OC remineralization contributes 36.7% to the TOC remineralization, which is disproportionally high compared to the percentage of Fe_R-OC in TOC (5.2±1.8%) in this zone. Therefore, Fe_R-OC is preferentially remineralized compared to OC that is not bound to Fe_R (non-Fe_R-OC) in the SMTZ of QDN-14B and results in the low percentage of Fe_R-OC in TOC." (Lines 267-271)

Line 269-272: using the numbers provided here, "a substantial fraction" the authors claim is less than 4%. Is this "substantial"?

Response: In previous manuscript, we compared the number of cells that can be supported by the Fe_R-OC remineralization flux in continental slope SMTZ sediments to the microbial cell abundance in global marine sediments, which underestimated the significance of Fe_R-OC remineralization flux in continental slope SMTZ sediments in supporting subsurface microorganisms. In the revised manuscript, the microbial cell abundance in continental slope SMTZ sediments is estimated and used instead:

> "Considering generally lower power demand of 10^{-20} to 10^{-16} W $cell^{-1}$ for microorganisms in marine sediments[51], this amount of energy could potentially support an even larger population of 10^{24} to 10^{28}cells. The average SMTZ depth in continental slope sediments is estimated to be 12.8±12.1 meters below seafloor[31], where the cell abundance typically ranges from 10^{6} to 10^{8} cells cm^{-3} [46]. Assuming the SMTZ depth interval of 1 m, the volume of global continental slope SMTZ sediments reaches 3×10^{13} m^3, which harbors about 3×10^{25} to 3×10^{27} cells. Therefore, the remineralization of Fe_R-OC in continental slope SMTZ sediments could support a substantial fraction of subsurface microbial life in this zone." (Lines 293-302)

Line 288: Based on the molar ratio of Fe_R-OC to Fe_R and the two mechanisms, the proportion can be estimated to support "a large proportion" the authors proposed. Otherwise, the proposal is weak.

Response: Yes, in principle the relative contribution of Fe_R-OC associated with Fe_R by adsorption and co-precipitation can be estimated by the end-member Fe_R-OC to Fe_R molar ratios of these two mechanisms and the observed Fe_R-OC to Fe_R molar ratios in sediment samples following a simple mass balance equation. However, as the end-member Fe_R-OC to Fe_R molar ratios of two mechanisms have large ranges, so it is not possible to give an accurate number on this proportion. We propose that "a major proportion of Fe_R-OC is formed at the redox interface in marine environments" not only because Fe_R-OC to Fe_R molar ratios higher than 1 indicating co-precipitation are observed in most layers of both sediment cores, but also because the carbon isotope ratios of Fe_R-OC indicate that marine OC consistently dominate over terrestrial OC.

Considering the Fe_R-OC pool is exchangeable during the early diagenesis. The sentence has been rephrased to avoid ambiguity:

> "All these clues suggest that a large proportion of terrestrial Fe_R-OC is replaced by marine OC when new Fe_R and OC associations are formed at the redox interface near the seafloor. This process could happen either autochthonously after deposition, or allochthonously in neighbouring surface sediments and transported to the core location as resuspended particulates[58, 59]." (Lines 315-320)

Line 329: "shelf" should be "slope"?

Response: Corrected.

Line 331: change "is" to "are".

Response: Corrected.

Line 331-334: a conjunction is needed for the two sentences.

Response: Corrected.

Line 354 & 357: provide the number of data to be averaged.

Response: Thank you for the suggestion. We have added the number of data to be averaged in Fig. 4 and Table S4 in the revised manuscript.

Reviewer #3 (Remarks to the Author):

The manuscript investigated the long-term variations in content and isotope of TOC and FeR-OC, emphasizing the impact of SMTZ in FeR-OC preservation. Based on the basically unaltered FeR-OC/TOC ratio long-buried sediments, this study provided an estimation of global budget of FeR-OC in Quaternary marine sediments. The topic is new and innovative, and experimental and analytical designs are convincing. The results provide an important experimental basis for understanding the role of sedimentary FeR-OC in regulating long-term carbon cycle.
Response: Thank you for your appreciation on our manuscript. Please see below our responses to detailed comments.

1. Title, the current title could not be completely reflect the topic and the present study, such as the items of digenesis and microbial community.
Response: We have changed the title to "Cycling and persistence of iron-bound organic carbon in subseafloor sediments" to fully reflect the dynamic cycling of Fe_R-OC we found in SMTZ sediments and its general stability in non-SMTZ sediments.

2. Before the process of SMTZ, how the FeR-OC change and/or response towards the reduction process of dissimilatory Fe and Mn reduction?
Response: Thank you for this comment. We added some information regarding the effect of dissimilatory iron reduction on Fe_R-OC in the manuscript:

> "The presence of low levels of dissolved iron throughout the core QDN-G1 (3-35 μM, Fig. 1b) is consistent with some background activity of dissimilatory iron reduction, independent of the biogeochemical zone. However, there is no discernable imprint on the content of Fe_R-OC, except in the SMTZ (Fig. 2b)." (Lines 131-134)

Mn reduction may affect the organic carbon that associated with Mn oxides in a similar way as iron reduction on Fe_R-OC, but it is beyond the scope of our study.

3. The impact of SMTZ in FeR-OC preservation was more significant in the sediment core QDN-14B nearby cold seeps due to the more abundant microbial occurrence; however, this degradation mechanism could not be suitable for the slope sedimentary setting, thus is the microbial evidence also be increased in the normal slope core with SMTZ section? In addition to the support from the remineralization of FeR-OC, what is the role of methane release for this specific microbial community structure?
Response: The modeled sulfate reduction rates, relative abundance of *Desulfobacterota* in bacteria and cell number of *Desulfobacterota* of core QDN-G1 (normal slope core) are shown in Supplementary Fig. 5. Although there is no evidence indicating elevated sulfate reduction rates and increased sulfate reducing bacteria in the SMTZ of QDN-G1 (Lines 222-226), the increase of the microbial activity is expected in the SMTZ of as methane could be utilized by methane oxidizing archaea in concert with sulfate reducing bacteria, which was supported by

the decrease of TOC and Fe_R-OC contents in the SMTZ (Fig. 2b).

The high methane flux in QDN-14B because of the nearby cold seep stimulates the sulfate reduction coupling with anaerobic oxidation of methane, which leads to the high relative abundance of sulfate-reducing bacteria in all bacteria and high relative abundance of methane metabolizing archaea among all archaea. (Lines 217-222)

4. Line 143-154. As the authors mentioned, the sediment provenance could alter the characteristics of FeR-OC and TOC. Although the impacts of provenance were not apparent in the studied two cores located in continental slopes, provenance was non-negligible factor in the shelfs due to exposure and submersion cycles on glacial/interglacial timescales. Therefore, is the extrapolation of the stable FeR-OC/TOC ratio to shelf and delta/estuary reasonable or rigorous?

Response: We have refocused our manuscript on the dynamic cycling of Fe_R-OC in SMTZ sediments, and largely shortened the part on Fe_R-OC reservoir estimation by providing only a rough estimation based on an updated dataset and extra statistical analyses:

> "Taken together, the Fe_R-OC records in these two cores suggest that a stable proportion of TOC survives early diagenesis as Fe_R-OC and is sequestered in marine sediments on at least 100-kyr timescales. This enables a rough estimation of the global Fe_R-OC reservoir by multiplying the TOC reservoir in global marine sediments from the Quaternary Period (0-2.59 Ma) storage[50] with published Fe_R-OC/TOC ratios in marine surface sediments. For deriving representative Fe_R-OC/TOC ratios, all published Fe_R-OC/TOC data in marine sediments using the CBD method were compiled and combined with our measured data and grouped into eight categories based on their study area information: delta and estuary, continental shelf, continental slope, deep sea, anoxic/sulfidic regions, wetland, mangrove and tephra (Fig. 4, Supplementary Table 3). One-way ANOVA and Tukey Honest Significant Difference (HSD) test revealed that significant difference ($P<0.05$) in Fe_R-OC/TOC values exists only between tephra-containing sediments and other environments (Supplementary Table 4). Considering TOC storage in the Quaternary sediments is estimated in the three domains shelf (water depth <200 m), margin (200 m < water depth <3500 m) and abyss (water depth >3500 m) [50], delta/estuary and continental shelf were combined to represent shelf, continental slope was used to represent margin and deep sea was used to represent abyss. Combining the published TOC reservoir and the corresponding surface Fe_R-OC/TOC ratios in these three domains[50], the global Fe_R-OC reservoir in Quaternary marine sediments was estimated to be 28,550 ± 12,160 Pg C (Supplementary Table 5), which is approximately 19-46 times the size of the atmospheric carbon pool." (Lines 335-356)

5. Lines 212-216. Re-adsorption of isotopically depleted DOC to Fe was considered to result in the negative δ13CFeR-OC values and low FeR-OC content. From the perspective of geological evolution, the sediments below the SMTZ have experienced the Fe reduction and DOC re-adsorption before burial in present depth, but the FeR-OC content and isotope characteristics are similar to those of sediments above SMTZ. Did some isotopically enriched OC adsorb to the FeR after passing through the SMTZ? If so, some of the FeR-OC maybe not only formed at

the redox interface as proposed in lines 288-289.

Response: Thank you for this comment. The re-adsorption of some isotopically enriched DOC to the Fe_R cannot be ruled out. We added following information to explain similar Fe_R-OC above and below the SMTZ:

> "The fact that sediments below the SMTZ resemble those above the SMTZ is at first surprising if we assume that the more deeply buried sediments were at some point in the geologic past also situated in the SMTZ. In this case, re-adsorption of DOM produced during cycling of particulate organic matter of marine origin could be the cause for the similarity of Fe_R-OC related signals above and below the SMTZ. However, there is also evidence that SMTZs do not "move" continuously through the sediment column with increasing sedimentation but rather "oscillate" from one horizon to another where they then remain stagnant in position for extended periods[37, 38]." (Lines 203-211)

We also acknowledge that what we originally proposed in Lines 288-289 "We propose that a large proportion of Fe_R-OC is formed at the redox interface in marine environments" was not accurate. Here we intended to describe the replacement of terrestrial OC to marine sourced OC near the seafloor. It has been rephrased to "All these clues suggested that a major proportion of terrestrial Fe_R-OC is replaced by marine-OC when new Fe_R and OC associations are formed at the redox interface near the seafloor." (Lines 315-317)

6. Line 293-319, 339-360, the global estimation of Fe-OC over a larger spatial and temporal scale should be important, however, much of these explanation and discussion as mentioned here could be shorten to make the draft more focusing.

Response: Thank you so much for this suggestion. We have revised this part of the manuscript substantially to make it more straightforward. (Lines 323-356)

7. Table 1 present clear comparisons of content and carbon isotope between non-SMTZ and SMTZ. Statistical analyses (e.g. one-way ANOVA) are recommended to guarantee the reliability of the comparison, just like you did in Figure 4.

Response: Thank you for the suggestion. The significance tests have been done and the results have been added to Table 1 and the main text (Lines 176-179). Detailed information has been added to methods (Lines 568-577).

Reviewer #4 (Remarks to the Author):

Chen et al. present a pair of detailed short-core datasets which they use to use to investigate the coupling of FeR and OC in marine sediments. This is a nice study, and looks to be scientifically sound. The findings are generally quite interesting, especially the suggestion that microbial remobilisation of FeR occurs in the SMTZ. However, I feel other than these data, the manuscript does not represent enough of a step forward to warrant publication in Nature Communications in its current form. My reasons for this are as follows:

1. The title, and the abstract discuss 'geological timescales', while the term 'long-buried sediments' is used in line 103. However, a study which investigates 100 kyr does not fit the bill

to me. Persistence of FeR -OC over much longer timescales (up to tens of millions of years) has now been demonstrated in numerous publications (Faust et al., 2021; Longman et al., 2021, 2024). Some of these studies have not been cited in this work, so emphasising this as an aspect of novelty seems wrong. I would adjust the title and the primary narrative of the manuscript to better reflect the more interesting data – what goes on through the SMTZ and below, and interactions of FeR and OC during burial.

Response: Thank you! We have revised the manuscript per your suggestions as follows:

(1) We have changed the title to "Cycling and persistence of iron-bound organic carbon in subseafloor sediments" to reflect the dynamic cycling of Fe_R-OC pool during early diagenesis. This new title also reflects the changed focus of the narrative.

(2) We have emphasized the novelty of our study as exploring the fate of Fe_R-OC in subseafloor sediments during microbially mediated diagenetic processes in the introduction:

> "A comprehensive picture regarding the fate of Fe_R-OC on geological timescales is still lacking but several studies of sediments of Pleistocene age suggest that the Fe_R-OC fraction remains relatively stable[11-14]. However, the question to what degree the Fe_R-OC reservoir interacts with sedimentary biogeochemical processes, in particular those involving redox reactions of iron and sulfur, remains unresolved." (Lines 44-49)

(3) The potential effect of diagenetic redox reactions and depositional history on sedimentary Fe_R-OC records has been added to the introduction as background information with literature review:

> "Fe_R has long been found to promote organic matter preservation in terrestrial soils as well, especially via adsorption[5, 15]. However, recent studies indicated the interactions between Fe_R and OC in soils are highly influenced by the redox oscillations during water-table fluctuatioins[16-18] in terrestrial environments. Under anoxic conditions, Fe_R reduction releases Fe_R-OC and increased the anaerobic mineralization of soil organic matter[17, 18]. While under oxic conditions, newly-formed Fe_R promotes OC retention on mineral surfaces[16]. In anoxic marine sediments, Fe_R is one electron acceptor actively involved in biogeochemical processes[19-21]. Both Fe_R reduction by microorganisms and biogenic sulfide produced during sulfate reduction could potentially weaken the association with OC. Indeed, recent lab incubations demonstrated that Fe_R-OC can be remobilized during microbial iron reduction and subsequently utilized as electron donor and/or carbon source for microbial communities[22-24]. Consequently, the remobilization of Fe_R-OC during early diagenesis may influence the relative size of the Fe_R-OC reservoir that preserved in sediments for long time. Additionally, the supply of Fe_R-OC is expected to depend on the hydrological conditions through the influence on continental weathering and Fe_R formation as well as on sea level through the influence on shelf topography and thus transport of continental detritus into the deep sea[25-27]; both factors are expected to vary periodically on glacial-interglacial timescales. To disentangle the effect of Fe_R-OC supply and early diagenetic reworking on sedimentary Fe_R-OC, downcore Fe_R-OC records need to be established and related to both geochemical zonation and sediment chronology." (Lines 51-72)

(4) We refocus the discussion on the dynamic cycling of Fe_R-OC in SMTZ sediments, providing more information on the most likely mechanism of Fe_R-OC remobilization and

remineralization, and the scenario of the interactions between Fe_R and OC beneath the SMTZ:

"Accordingly, two observations deserve attention: (i) the distinct depletions in ^{13}C content and contents of Fe_R-OC within the SMTZ, and (ii) sediments below the SMTZ show similar features with respect to ^{13}C content and contents of Fe_R-OC as sediments above the SMTZ. (i) The relatively low content of Fe_R-OC strongly suggests that a substantial fraction of it has been remobilized and possibly remineralized. Two mechanisms could account for the low $\delta^{13}C_{FeR\text{-}OC}$ of the residual fraction: (a) remobilization/degradation of a ^{13}C-enriched, weakly bound and/or more reactive fraction[34] with the residual fraction being 13C-depleted; since the ?13C of the residual fraction is broadly consistent with an origin from terrestrial plants, the residual fraction could be tightly adsorbed terrestrial organic matter supplied together with the detrital minerals from land. (b) The alternative involves again remobilization of a large fraction of Fe_R-OC combined with adsorption of 13C depleted dissolved organic matter (DOM) produced in the course of anaerobic oxidation of methane[35]; the extremely low molar ratios of Fe_R-OC to Fe_R below 1 in both SMTZs (Fig. 2e and j) indicate abundant binding sites available for DOM molecules[15, 36]. These two mechanisms are not mutually exclusive and could act in combination. In any case, both scenarios require remobilization and possibly degradation of a substantial fraction of Fe_R-OC. (ii) The fact that sediments below the SMTZ resemble those above the SMTZ is at first surprising if we assume that the more deeply buried sediments were at some point in the geologic past also situated in the SMTZ. In this case, re-adsorption of DOM produced during cycling of particulate organic matter of marine origin could be the cause for the similarity of Fe_R-OC related signals above and below the SMTZ. However, there is also evidence that SMTZs do not "move" continuously through the sediment column with increasing sedimentation but rather "oscillate" from one horizon to another where they then remain stagnant in position for extended periods[37, 38]" (Lines 186-211)

(5) We have largely shortened the last section on the persistence of Fe_R-OC on geological timescale, but only kept a rough estimation of Fe_R-OC reservoir in Quaternary sediments.

2. The compilation of data for the section discussing the scale of the marine sedimentary FeR-OC sink is lacking a large number of studies. There was a recently published updated dataset of FeR-OC in such environments (Longman et al., 2022), which cites a lot of work not considered here, and a lot of environments not considered. Furthermore, the existence of that study, which provides estimates of the amount of FeR-OC buried every year, means this study as presented does not represent something fully novel.

Response: Thank you so much for your comment and suggestions. We had not considered some data from the mentioned environments because of their limited distribution in the previous manuscript. However, we agree that all the published data should be used for compilation and statistical analyses. In the revised manuscript, the most up-to-date dataset was used and grouped into eight marine environments (Fig 4). The detailed information was updated in Supplementary Table 3. We have put more information in the footnote of Supplementary Table 3 for clarification:

"The most up-to date published dataset compiled by Longman et al., 2022 was used[21],

> with following exceptions: Ghaisas et al., 2021 was not included for different Fe_R-OC extraction method[22]; Tao et al. 2017 and Longman et al., 2024, which were not included in Longman et al., 2022, were included here[14, 20]."

3. What is novel is trying to estimate the total FeR-OC pool in quaternary sediments. However, I cannot see the reason behind making this calculation. The authors are aiming to estimate the amount of sediment which is microbially active, but the only microbial activity they demonstrate to have an impact on feR-OC is in the SMTZ. As they themselves show, this zone in limited, and does not represent the entirety of quaternary marine sediment. The cores studied here have SMTZ thicknesses of only up to a metre. Even in this study they only represent around 15% of the sediment. Outside of this there is no evidence for variability in FeR-OC driven by microbes. I really don't think any sort of extrapolation of FeR-OC based on the LaRowe estimate of Quaternary sediments makes much sense when trying to estimate what is impacted by microbes in the SMTZ. I would suggest a better way to investigate this would be to focus on the SMTZ – I am no expert on this, but are there estimates of thickness available globally?

Response: We estimated the Fe_R-OC reservoir in microbially active Quaternary sediments, but "microbially active" here is not related to the specific microbial processes remobilizing Fe_R-OC in SMTZs. Instead, it only means the sediments deposited since Pleistocene are not yet affected by severe high temperature and most of the microbial degradation of OC takes place in this portion of marine sediments. We put more information to clarify the different Fe_R-OC degradation patterns in general non-SMTZ and SMTZ sediments before we start with the estimation:

> "The average Fe_R-OC/TOC ratio in the two cores studied, except within the SMTZ in QDN-14B, is 13.3±3.2%. The relative stable Fe_R-OC/TOC ratios in the two cores indicates a similar degradation rate of Fe_R-OC and non-Fe_R-OC. The only exception is the especially active SMTZ in QDN-14B influenced by the methane-rich fluids, where iron reduction is enhanced significantly by processes likely induced by sulfate reducing bacteria, methanotrophic archaea and/or biogenic sulfide. The stimulated degradation of Fe_R-OC over non-Fe_R-OC leads to extremely low Fe_R-OC/TOC ratios in the SMTZ of QDN-14B. As these especially active SMTZs have limited distribution in global marine sediments and occupy narrow depth intervals in sediment columns, the overall impact on the Fe_R-OC reservoir in subseafloor sediments is small." (Lines 323-333)

In SMTZ sediments, the Fe_R-OC records we observed represents the characteristic of residual Fe_R-OC after the remobilization and remineralization. Therefore, instead of the Fe_R-OC reservoir in the SMTZ, the remineralization flux of Fe_R-OC in the SMTZ of global continental slope sediments was estimated. In the revised manuscript, the microbial cell abundance was estimated and compared with the biomass that can be sustained by the Fe_R-OC remineralization flux in this zone:

> "Considering generally lower power demand of 10^{20} to 10^{16} W $cell^{-1}$ for microorganisms in marine sediments[51], this amount of energy could potentially support an even larger population of 10^{24} to 10^{28}cells. The average SMTZ depth in continental slope sediments is estimated to be 12.8±12.1 meters below seafloor[31], where the cell abundance

typically ranges from 10^6 to 10^8 cells cm^{-3} [46]. Assuming the SMTZ depth interval of 1 m, the volume of global continental slope SMTZ sediments reaches 3×10^{13} m^3, which harbors about 3×10^{25} to 3×10^{27} cells. Therefore, the remineralization of Fe_R-OC in continental slope SMTZ sediments could support a substantial fraction of subsurface microbial life in this zone." (Lines 293-302)

4. However, even if the adjustment to reflect only the SMTZ is made, the fact that data show the FeR-OC values are similar above and below the SMTZ in one core (and likely in the other if deeper samples were available) suggests nothing of much consequence is ongoing. Yes, there may be some remineralisation, but overall if we are indeed concerned about the long term sink, it doesn't look like the SMTZ and microbes really affect this. This is definitely not an issue, but framing it as a finding of importance makes little sense to me.
Response: Similar to the last comment, with the information we added to clarify the different Fe_R-OC degradation patterns in general non-SMTZ and SMTZ sediments, this concern should also be addressed (Lines 323-333).

5. I'm not sure where the links to 'deep life' come from. Yes, FeR-OC can be a large labile reservoir of OC, but what suggests deep life? I assume the microbiology results? If so, make the link here clearer. However, if it is the microbiology, I see no evidence for this – you see some really interesting microbial activity in the SMTZ, but nothing to suggest this goes on in the 'deep' subsurface. Also, I don't think 900 cm is really 'deep'. There's papers which discuss truly deep subsurface microbial life (e.g. Inagaki et al., 2015), which show microbial activity down to more than 2.5km (250000 cm).
Response: Thank you for pointing this out. Yes, the 'deep life' mentioned here refers to microorganisms living in the subsurface sediments. We agree that comparing to the truly deep subsurface microbial life, we only discussed microbial life in relatively shallow sediments. We have rephrased "deep life" as "subseafloor microorganisms" to avoid the misunderstanding (Line 31).

I'm sorry to seem so negative about this. The study is certainly interesting and the combination of FeR-OC measurements with microbiology, and everything going on in the SMTZ is interesting. However, I think the novelty is not there for this journal – the scaling up is unsuitable and the regular discussion of geological timescales seems wrong to me.
Response: Thank you for your helpful comments and suggestions. Now we have substantially revised the manuscript by reemphasizing the novelty of our study in the introduction with an updated literature review, and reframed our discussion with more focus on dynamic cycling of Fe_R-OC in SMTZ sediments and its impact on subseafloor microorganisms. We have largely shortened the part on the estimation of Fe_R-OC reservoir in Quaternary sediments, but kept the rough estimation we made with updated dataset and extra statistical analyses.

References.
Faust, J. C., Tessin, A., Fisher, B. J., Zindorf, M., Papadaki, S., Hendry, K. R., Doyle, K. A., & März, C. (2021). Millennial scale persistence of organic carbon bound to iron in Arctic marine sediments. Nature Communications, 12(1), 1–9. https://doi.org/10.1038/s41467-020-20550-

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Inagaki, F., Hinrichs, K.-U., Kubo, Y., Bowles, M. W., Heuer, V. B., Hong, W.-L., Hoshino, T., Ijiri, A., Imachi, H., Ito, M., Kaneko, M., Lever, M. A., Lin, Y.-S., Methé, B. A., Morita, S., Morono, Y., Tanikawa, W., Bihan, M., Bowden, S. A., … Yamada, Y. (2015). Exploring deep microbial life in coal-bearing sediment down to ~2.5 km below the ocean floor. Science, 349(6246), 420–424. https://doi.org/10.1126/science.aaa6882

Longman, J., Faust, J. C., Bryce, C., Homoky, W. B., & März, C. (2022). Organic Carbon Burial With Reactive Iron Across Global Environments. Global Biogeochemical Cycles, 36(11), e2022GB007447. https://doi.org/10.1029/2022GB007447

Longman, J., Gernon, T. M., Palmer, M. R., & Manners, H. R. (2021). Tephra Deposition and Bonding With Reactive Oxides Enhances Burial of Organic Carbon in the Bering Sea. Global Biogeochemical Cycles, 35(11), e2021GB007140. https://doi.org/10.1029/2021GB007140

Longman, J., Manners, H. R., Gernon, T. M., McManus, J., Palmer, M. R., Rowland, S. J., & Sutton, P. A. (2024). Production and preservation of organic carbon in sub-seafloor tephra layers. Marine Chemistry, 258, 104334. https://doi.org/10.1016/j.marchem.2023.104334

Response: Thank you for the information. The papers studying the long-term preservation of Fe_R-OC have been cited in the introduction.

REVIEWER COMMENTS

Reviewer #2 (Remarks to the Author):

I have no more suggestions/comments.

Reviewer #3 (Remarks to the Author):

I have carefully reviewed your revised manuscript and am pleased to note that you have addressed the issues raised during the previous round of reviews comprehensively. The revised discussions have improved the coherence of the manuscript and greatly enhanced the quality of the paper. However, some of the newly added statements still need to be reconsidered.

You mentioned that the lower FeR-OC contents, lower FeR-OC/TOC values, and lower δ13CFeR-OC in SMTZ sediments are ascribed to the remobilization/degradation of FeR-OC (lines 201-203), which is caused by the biological and chemical reduction of FeR (lines 232-240). Further, the calculated FeR-OC remineralization fluxes in the SMTZ of core QDN-G1 and QDN-14B were similar (5.0 mol m-2 kyr-1 and 5.8 mol m-2 kyr-1, respectively). I am confused why sulfate reduction activity in the SMTZ of core QDN-G1 does not peak like that of core QDN-14B (lines 222-225). The cell number of Desulfobacterota in the SMTZ is even lower than that in non-SMTZ. Since I did not see any interpretation of the FeR reduction mechanism of core QDN-G1, it seems that the authors considered it is caused by the biological process as core QDN-14B. But why?

Lines 315-317. “All these clues suggest… near the seafloor.” I do not think this is an evidence-based statement, as even at the uppermost part of the core, the δ13CFeR-OC is highly enriched. Moreover, there exists the possibility that the terrestrial input FeR did not initially associate with terrestrial OC, but rather, it bonded with marine OC once entering the ocean.

Lines 324-325. “The relative stable FeR-OC/TOC ratios in the two cores indicates a similar degradation rate of FeR-OC and non-FeR-OC”. This statemen seems against the general concept that FeR would protect the OC from microbial degradation. If this is correct, one can suppose that the FeR did not have any effect on the preservation of OC on the glacial-interglacial timescales in the non-SMTZ sediments. I think this is not what the author intended to convey in this study.

Reviewer #4 (Remarks to the Author):

I am delighted to see Chen et al. have taken my comments on board, and the manuscript is greatly improved in its current form. Very nice work! I would suggest with a few more changes it will be suitable for publication. My small suggestions are below (and my line numbers refer to the clean new version of the manuscript):
L17 onwards: I think using TOC and OC in the same sentence is confusing. As the standard in the

field is to use the term FeR-OC for the coupling, I would suggest sticking with OC rather than using TOC when describing organic carbon throughout.

L22: Not sure how you come to the conclusion it's marine sourced – we don't know where the FeR comes from do we? Could be weathered material from the land. Same for the OC – some component of terrestrial OC is likely.

L40: I would rephrase to start the sentence with 'It is estimated ~20.5…'

L46: Just for reference, the Longman et al. (2024) study (ref.13) shows some sediments of Eocene age containing high FeR-OC.

L114-115: Can you include a reference for the marine sediment zonation?

Table 1: I wonder if some of these data, which are crucial to the ms, could be presented in box plot form? For example, the FeR-OC values and the d13CFeR-OC would be nicely presented as a box plot.

L238 'summary' rather than 'sum'

L257: 'The depth interval' is an odd way to say it. Maybe simply say 'The SMTZ…'

L337: This could be expanded as an explanation for the samples of even older age which show FeR-OC coupling to be important.

Jack Longman

Response to reviewers' comments on 'Cycling and persistence of iron-bound organic carbon in subseafloor sediments.'

We thank the editor and reviewers for re-considering our revised manuscript. We have followed the valuable advice and further revised the manuscript, including clarifying questionable statements that were newly added to the manuscript during the first revision. We have also improved our estimation on the Fe_R-OC remineralization flux in the SMTZ. Please find below our responses to the specific comments. Line numbers refer to lines in the clean version.

Reviewer #2 (Remarks to the Author):

I have no more suggestions/comments.
Response: We thank Reviewer 2 again for his/her efforts in reviewing our manuscript.

Reviewer #3 (Remarks to the Author):

I have carefully reviewed your revised manuscript and am pleased to note that you have addressed the issues raised during the previous round of reviews comprehensively. The revised discussions have improved the coherence of the manuscript and greatly enhanced the quality of the paper. However, some of the newly added statements still need to be reconsidered.
Response: We thank the reviewer for the supportive comments and recognition of our revision. We have further improved our manuscript based on his/her suggestions. Please see below our responses to detailed comments.

You mentioned that the lower FeR-OC contents, lower FeR-OC/TOC values, and lower δ13CFeR-OC in SMTZ sediments are ascribed to the remobilization/degradation of FeR-OC (lines 201-203), which is caused by the biological and chemical reduction of FeR (lines 232-240). Further, the calculated FeR-OC remineralization fluxes in the SMTZ of core QDN-G1 and QDN-14B were similar (5.0 mol m-2 kyr-1 and 5.8 mol m-2 kyr-1, respectively). I am confused why sulfate reduction activity in the SMTZ of core QDN-G1 does not peak like that of core QDN-14B (lines 222-225). The cell number of Desulfobacterota in the SMTZ is even lower than that in non-SMTZ. Since I did not see any interpretation of the FeR reduction mechanism of core QDN-G1, it seems that the authors considered it is caused by the biological process as core QDN-14B. But why?
Response: The reviewer has a good point: despite multiple lines of evidence for higher sulfate-reducing activity in core QDN-14B, the fluxes of Fe_R-OC remineralization, as approximated by our approach, appear rather similar.

In response to this valuable comment, we now more explicitly acknowledge that the estimate provided for the less active core QDN-G1 is associated with a higher uncertainty due to the steady decline in Fe_R-OC cand TOC ontent above the SMTZ. Consequently, we also used a slightly modified approach to estimate the lower boundary of the flux. The resulting flux is more consistent with the sulfate reduction rates. For global extrapolation, we now use the entire range of the two cores to express the uncertainty (see revised paragraph in lines 274 to 284).

We additionally added information to the methods on the estimation of TOC and Fe_R-OC remineralization fluxes in the SMTZ (Lines 515-541).

Lines 315-317. "All these clues suggest… near the seafloor." I do not think this is an evidence-based statement, as even at the uppermost part of the core, the δ13CFeR-OC is highly enriched. Moreover, there exists the possibility that the terrestrial input FeR did not initially associate with terrestrial OC, but rather, it bonded with marine OC once entering the ocean.
Response: We thank the reviewer for pointing this out. We agree that the previous statement may have been misleading. We have added the sentence "After entering the ocean, additional organic matter from the pool of marine dissolved organic carbon may be bound to Fe_R-bearing minerals by adsorption[59,60]." (Lines 316-318) and we clarified the text by deleting the statement regarding the replacement of terrestrial organic matter.

Lines 324-325. "The relative stable FeR-OC/TOC ratios in the two cores indicates a similar degradation rate of FeR-OC and non-FeR-OC". This statemen seems against the general concept that FeR would protect the OC from microbial degradation. If this is correct, one can suppose that the FeR did not have any effect on the preservation of OC on the glacial-interglacial timescales in the non-SMTZ sediments. I think this is not what the author intended to convey in this study.
Response: The reviewer is correct that we did not intend to make the theme of "preferential preservation of Fe_R-OC relative to non-Fe_R-OC on geological timescales" the major focus of the paper. A more comprehensive study would be required to emphasize this point and generalize this observation. Nevertheless, we feel that it is worth mentioning this observation. We slightly softened the statement by replacing "indicates" with "suggests" and by adding "on the timescales represented by these cores" (Line 331-333).

Reviewer #4 (Remarks to the Author):

I am delighted to see Chen et al. have taken my comments on board, and the manuscript is greatly improved in its current form. Very nice work! I would suggest with a few more changes it will be suitable for publication. My small suggestions are below (and my line numbers refer to the clean new version of the manuscript):
Response: We are pleased to hear that our efforts and the revised manuscript are well received. We would like to thank the reviewer again for all the constructive suggestions, which greatly improved our manuscript. We have further improved our manuscript based on the reviewer's suggestions. Please see below our responses to detailed comments.

L17 onwards: I think using TOC and OC in the same sentence is confusing. As the standard in the field is to use the term FeR-OC for the coupling, I would suggest sticking with OC rather than using TOC when describing organic carbon throughout.

Response: Thank the reviewer for this suggestion. We now use “fFe_R-OC” instead of “Fe_R-OC/TOC” to represent the fraction of Fe_R-OC in TOC throughout the text, which makes the terminology simpler and consistent with other recent publications in the field. In other cases, TOC is used when we refer to the bulk organic carbon in the sediments, which includes both Fe_R-OC and non-Fe_R-OC.

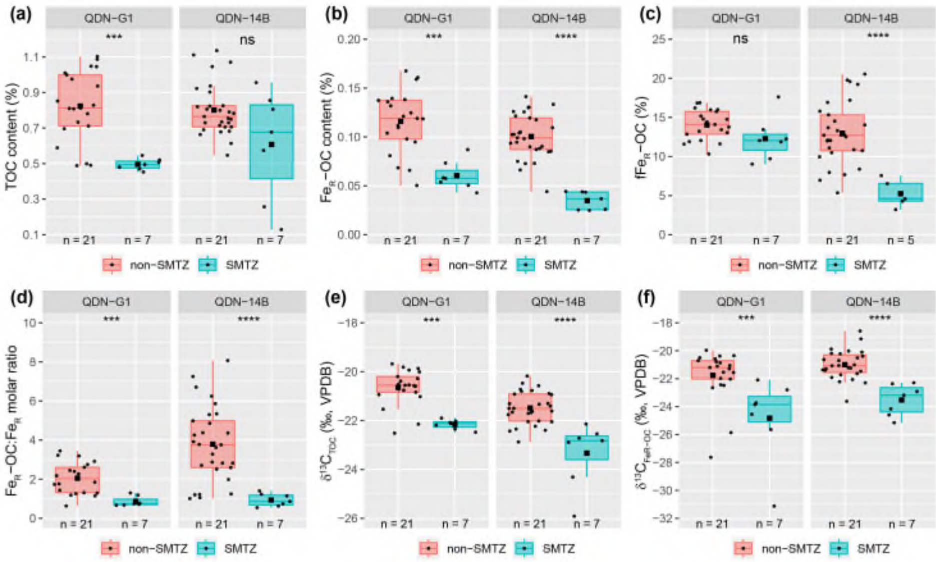
L22: Not sure how you come to the conclusion it’s marine sourced – we don’t know where the FeR comes from do we? Could be weathered material from the land. Same for the OC – some component of terrestrial OC is likely.
Response: Agreed. There is probably a fraction of Fe_R-OC coming from the land. The word “marine-sourced” has been removed from the sentence.

L40: I would rephrase to start the sentence with ‘It is estimated ~20.5…’
Response: Rephrased.

L46: Just for reference, the Longman et al. (2024) study (ref.13) shows some sediments of Eocene age containing high FeR-OC.
Response: The sentence has been changed to “A comprehensive picture regarding the fate of FeR-OC on geological timescales is still lacking but several studies of sediments of Pleistocene as well as late Paleocene to early Eocene age suggest that the fraction of Fe_R-OC in TOC (fFe_R-OC) remains relatively stable[11-14].” (Line 47-50).

L114-115: Can you include a reference for the marine sediment zonation?
Response: A classic paper from Canfield and Thamdrup (2009) discussing the geochemical zonation in marine sediments has been cited (Line 117).

Table 1: I wonder if some of these data, which are crucial to the ms, could be presented in box plot form? For example, the FeR-OC values and the d13CFeR-OC would be nicely presented as a box plot.
Response: We have put all the information presented in Table 1 into box plots as the new Supplementary Fig. 5.



Supplementary Figure S5 Comparisons of the TOC and Fe_R-OC records between non-SMTZ and SMTZ sediments in two cores, including TOC content (a), Fe_R-OC content (b), fFe_R-OC (c), Fe_R-OC to Fe_R molar ratio (d), carbon isotope ratio of TOC (e), and carbon isotope ratio of Fe_R-OC (f). fFe_R-OC at 410 and 420 cmbsf in QDN-14B are biased by extremely low TOC and are not included for analysis. Significant levels in Wilcoxon test are indicated (ns: $P > 0.05$、***: $P <= 0.001$、****: $P <= 0.0001$). Box plots indicate mean (solid square), median (middle line), 25th, 75th percentile (box) and 1.5 times interquartile range (whiskers) with data points (solid dots) overlapped on top.

L238 ‘summary’ rather than ‘sum’

Response: Done.

L257: ‘The depth interval’ is an odd way to say it. Maybe simply say ‘The SMTZ…’

Response: Done.

L337: This could be expanded as an explanation for the samples of even older age which show FeR-OC coupling to be important.

Jack Longman

Response: We thank the reviewer for this suggestion. We have added the following sentence to the manuscript to expand this statement a little bit to the samples of older ages: “Considering previous studies showing high fFe_R-OC in the samples even from late Paleocene to early Eocene[12], persistence of Fe_R-OC on even longer timescales is expected in marine sediments.” (Lines 343-345)

REVIEWERS' COMMENTS

Reviewer #3 (Remarks to the Author):

I am delighted to see Chen et al. have taken my comments, and the manuscript is greatly improved in its current form. I have no more suggestions/comments.