

Revised title: Living and nonliving particulate iron in the subtropical North Pacific Ocean

Manuscript # egusphere-2025-6068

CC1: 'Comment on egusphere-2025-6068', Yang Xiang, 15 Jan 2026

Comments to the authors:

Bates and Hawco present a well-written manuscript demonstrating the partition between different particulate iron (pFe) pools, especially biogenic and nonliving pFe, during seasonal cruises at Station ALOHA. Such work is of great significance given the increasing attention on the role of authigenic mineral phases in the overall surface Fe cycling. The authors have done a good job presenting and interpreting the seasonal variations of particulate Fe data. The use of ATP to estimate biogenic is novel and quite interesting. The manuscript is clear, thorough, and nearly free of typos. However, I do have some substantive comments and editorial remarks as listed below. Overall, I recommend publication with major revisions.

We appreciate Dr. Xiang taking the time to provide feedback and have incorporated this feedback where possible.

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Major interpretation points

I have two major comments.

Firstly, the authors made many assumptions in their estimations of biogenic pFe, many of which were not mentioned in the text. For example, the Fe:C uptake ratio used in Equations 3-5 does not necessarily equal the cellular Fe:C quotas (Fe:C) used in Sofen et al. (2023). The authors need to acknowledge this and be clear about their assumptions.

This difference is discussed in lines 328-331, when comparing our data with that of Sofen et al. (2023). We have clarified this to:

“Note that the methods of estimating pFe_{Bio} differ between the two sites. Sofen et al. (2023) estimated pFe_{Bio} using Eq. 4 and cellular Fe quotas of eukaryotes determined by synchrotron X-ray fluorescence. This approach, using labile particulate phosphorus, includes detrital organic material as part of pFe_{Bio} and assumes Fe quotas from living, eukaryotic phytoplankton cells are representative of the entire microbial pool (Sofen et al., 2023).”

Additionally, Sofen et al. (2023) used PP_{Labile} in their calculations, while the authors used bulk PP in Equation 4. The authors should have the leachable PP data since they did the Berger leach. Al-Hashem et al. (2022; 10.1029/2022GB007453) found up to 60% of PP that cannot be accessed with the Berger leach, for example. Since the authors are comparing their results with Sofen's, it's important to make sure the calculations have been conducted similarly. Otherwise, those results may not be directly comparable.

We will clarify in section 3.5 Comparison to the North Atlantic Subtropical Gyre that our approach does not allow for a perfect comparison to the Sofen et al. data, due to differences in methodological approach. We still see important value in comparing the data with the caveat of these methodological differences may have a small effect on the absolute values that we derive.

We disagree that the use of $pPO4_{\text{labile}}$ is the only valid measurement. The Al-Hashem et al. paper is based on a coastal transect in a very high dust region (directly off the coast of the Namib Desert), where detrital $pPO4$ is worth considering. In contrast, in open ocean conditions with low dust deposition like Station ALOHA, $pPO4$ overwhelmingly represents autochthonous 'biogenic' materials (living cells + organic detritus). Indeed, at the station closest to Station ALOHA on the GEOTRACES GP15 transect, >80% of small (0.2-51 μm) particulate P was labile (Lam et al., 2024; lability data not available for large particles, but large particulate P comprised ~10% or less of total particulate P).

Lam, P. J., Lee, J., Amaral, V. J., Laubach, A., Carracino, N., Rojas, S., Mateos, K. (2024). Size-fractionated major, minor, & trace particle composition and concentration from Leg 1 (Seattle, WA to Hilo, HI) of the US GEOTRACES Pacific Meridional Transect (PMT) cruise (GP15, RR1814) on R/V Roger Revelle from Sept to Oct 2018. Biological and Chemical Oceanography Data Management Office (BCO-DMO). (Version 1) Version Date 2024-01-30 doi:10.26008/1912/bco-dmo.918811.1

Secondly, some of the calculations are flawed. The use of PC in Equation 3 to estimate biogenic Fe is likely an overestimation.

With respect, this is the intention of the calculation. PC or POC is the primary particulate carbon measurement by HOT, BATS, and GEOTRACES, and thus is a natural starting point for estimation of biogenic Fe.

While phytoplankton production (uptake of dissolved inorganic carbon) is the dominant source of new organic matter in the open ocean, the standing stock of POC (here the authors use PC since those are the only data available) includes significant

contributions from other sources. What about heterotrophic and allochthonous sources, for example?

We are not able to locate data arguing for allochthonous POC at Station ALOHA, which is generally representative of North Pacific subtropical gyre conditions and has a turnover time with respect to photosynthesis on the order of a few days. Our recent work describing a coastal source of Fe from the Hawaiian Islands only requires a mixture of approximately 0.1-1% coastal seawater (Bates & Hawco, 2025) so we do not consider the islands to be a major source of carbon to Station ALOHA.

Bates, E. S. and Hawco, N. J.: Dissolved Iron Seasonal Cycle and Residence Time in the North Pacific Subtropical Gyre, *Geophysical Research Letters*, 52, <https://doi.org/10.1029/2025GL118095>, 2025.

Based on the authors' definition of biogenic pFe, the authors should possibly use phytoplankton carbon rather than total POC in Equation 3. Please refer to Graff et al. (2015; 10.1016/j.dsr.2015.04.006) for the nuances, with the former values much smaller.

While we appreciate this recommendation, measurement of phytoplankton carbon derived from fluorescence activated flow cytometry is nontrivial and come with its own sets of caveats and uncertainties. We are currently onboarding these techniques, but even when implemented successfully, they are low throughput, and therefore will inevitably require an extrapolation to compare with other data. Ultimately, our APT-based approach accomplishes the same goal.

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Interpretation points, by line #

Lines 14-16: Do the authors have data or literature to show how much of a fraction is labile pFe in dust deposition at Station ALOHA? I assume that it's a relatively small fraction, right?

During dust deposition events at Station ALOHA, we observed labile pFe export from midwater sediment traps notably increased along with the increased lithogenic pFe export, resulting in an estimated 12-17% lability of the pFe export (Bates et al., 2025). Additionally, initial data from the Hawaii Aerosol Time-series suggests approximately 12-24% of aerosol Fe is labile (Kollman et al., 2024). Regardless, this would not change the conclusions from this study, as we are just

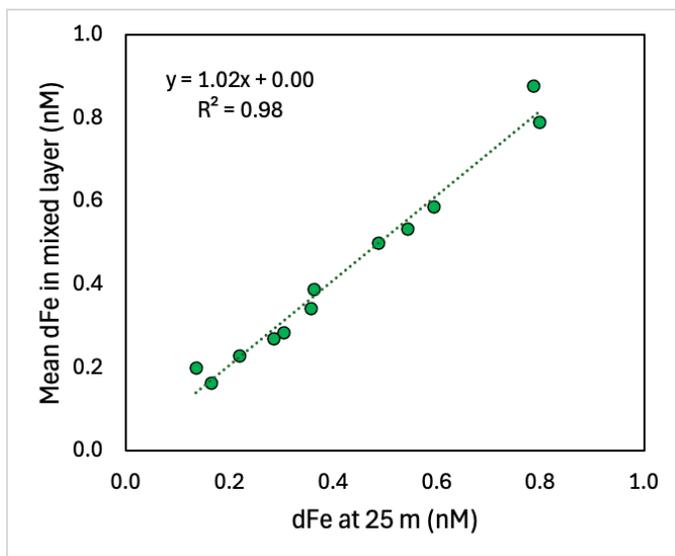
trying to recognize that some portion of the “authigenic” pFe pool may be instead derived from dust, not formed *in situ* (lines 252-256).

Bates, E. S., White, A. E., and Hawco, N. J.: Variability and Export Timescales of Upper Ocean Particulate Trace Metals in the North Pacific Subtropical Gyre, *Global Biogeochemical Cycles*, 39, <https://doi.org/10.1029/2025GB008657>, 2025.

Kollman, C., Marsay, C. M., Ohnemus, D., Stephens, M. P., Bates, E. S., and Buck, C. S.: Results from the Hawaii Aerosol Time-Series reveal seasonally contrasting aerosol solubility in the North Pacific Oligotrophic Gyre, driven by source diversity in spring and fall dust pulses, Abstract OS21H-0665, 2024 AGU Fall Meeting, 9-13 Dec, ADS Bibcode: 2024AGUFMOS21H0665K, 2024.

Lines 103-104: Are there any specific reasonings for using dFe at 25 m rather than the mean mixed layer concentrations? How different are dFe concentrations at 25 m and within the mixed layer?

The incubations were performed using seawater collected from 25 m; thus, the dFe concentration at 25 m is the most direct comparison for calculating an uptake rate. DFe concentrations at 25 m are well correlated with mean dFe concentrations in the mixed layer ($R^2 = 0.98$, $m = 1.02$) throughout our study period.



Line 117: Are sinking particulate Fe data from sediment traps at Station ALOHA? I assume that data were also extracted from the HOT-DOGS. If so, please add the data source to section 2.3.

We have clarified that these data are from sediment traps at Station ALOHA, which were generated as a result of our timeseries project. However, these data are not available on the HOT-DOGS platform, but from Bates et al. (2025), as cited. This dataset is publicly available on BCO-DMO.

Lines 174-179: If carbon uptake rates (GPP) are underestimated by the ^{14}C incubation experiments (values between GPP and NPP), the Fe:C uptake ratios will be overestimated. This could help decrease the $p\text{Fe}_{\text{Bio}}$.

The commenter points out a real issue with the use ^{14}C productivity measurements, but, with respect, we believe the issue is likely reversed. The best C-based denominator, in our opinion, would be net primary production, not gross primary production as implied in the comment above. This is because some fraction of the ^{14}C -determined productivity will be respired over the day while Fe would be only accumulate. If anything, the Fe:C stoichiometry would increase if we were able to precisely measure NPP, subsequently increasing the derived biogenic $p\text{Fe}$.

Lines 189-191: Assuming the rest of POC are phytoplankton carbon for simplicity, the actual $p\text{Fe}_{\text{Bio_PC}}$ will decrease by 58-74%, which will make $p\text{Fe}_{\text{Bio_PC}}$ much more similar to $p\text{Fe}_{\text{labile}}$ and $p\text{Fe}_{\text{Bio_ATP}}$.

We appreciate the commenter noting this, as this is why we included the ATP approach in addition to the PC approach. The 58-74% nonliving organic matter number comes from ATP measurements.

Lines 191-193: The way the Fe:C ratio is calculated is more like an average ratio with respect to the bulk pool of particles though. I don't think a lower Fe:C stoichiometry in detritus, a specific pool of particles, makes any difference to the overall calculation.

The Fe:C ratio is calculated as the uptake rate of Fe divided by the ^{14}C primary production, suggesting it represents the Fe:C ratio of bulk living primary producers. Using the PC approach, we are applying this bulk living Fe:C ratio to the entire PC pool. In these lines, we were explaining that this could be a cause of the overestimation if the PC pool contains particles that have a much lower Fe:C ratio than the live primary producers. Based on the mass balance constraints associated with $p\text{Fe}_{\text{labile}}$, a lower Fe:C in organic detrital material appears likely.

Lines 194-198: The authors could possibly quantify the effects from variations in phytoplankton community composition, since the C:P of living cells should range between 109 and 195 at Station ALOHA. With the minimum $\text{C:P}_{\text{phyto}}$ we have, the

pFe_{bio_pp} is likely overestimated by up to 50%, which will result in values more similar to pFe_{labile} , but not for all the data.

Based on the commenter's feedback, we estimated the variability expected in C:P based on 1) the variability in C:P for each group and 2) the variability in community composition. Using the interquartile range of observed C:P ratios for each group by Lomas et al. (2021), $C:P_{phyto}$ varies between 155-179 mol:mol. Based on the variability in composition over an annual cycle reported by Rii et al. (2016), $C:P_{phyto}$ would vary between 158-172 mol:mol. While there is variability in the C:P expected, it is still far from the 80 ± 36 mol:mol that would be needed to produce pFe_{Bio} values that would actually be in agreement with pFe_{Labile} .

Lomas, M. W., Baer, S. E., Mougnot, C., Terpis, K. X., Lomas, D. A., Altabet, M. A., and Martiny, A. C.: Varying influence of phytoplankton biodiversity and stoichiometric plasticity on bulk particulate stoichiometry across ocean basins, *Commun Earth Environ*, 2, 1–10, <https://doi.org/10.1038/s43247-021-00212-9>, 2021.

Rii, Y., Karl, D. M., and Church, M.: Temporal and vertical variability in picophytoplankton primary productivity in the North Pacific Subtropical Gyre, *Mar. Ecol. Prog. Ser.*, 562, 1–18, <https://doi.org/10.3354/meps11954>, 2016.

Lines 198-199: The exact reason will make equation 3 an overestimation of pFe_{bio} .

Based on this feedback and comments from a reviewer, we have clarified that including heterotrophic bacteria may lead to an overestimation of C:P.

"Additionally, the calculated $C:P_{phyto}$ used in Eq. 4 omits a major contribution from heterotrophic bacteria to bulk biomass (Karl et al., 2022) and therefore may overestimate the true C:P of living biomass (Fagerbakke et al., 1996; White et al., 2019)."

Fagerbakke, K., Heldal, M., and Norland, S.: Content of carbon, nitrogen, oxygen, sulfur and phosphorus in native aquatic and cultured bacteria, *Aquat. Microb. Ecol.*, 10, 15–27, <https://doi.org/10.3354/ame010015>, 1996.

White, A. E., Giovannoni, S. J., Zhao, Y., Vergin, K., and Carlson, C. A.: Elemental content and stoichiometry of SAR11 chemoheterotrophic marine bacteria, *Limnol Oceanogr Letters*, 4, 44–51, <https://doi.org/10.1002/lol2.10103>, 2019.

Line 242: If the authors name the remaining pool as nonliving, this implies what has been subtracted is "living". The pFe_{bio} is not necessarily pFe_{living} , right? Naming it as "non biogenic" is also not perfect, but it may be a better term. I agree that a more accurate term to name this pool of pFe is difficult...

Based on feedback from the reviewers, we have changed the terms to “biotic” and “authigenic + detrital”.

Lines 247-258: The authors used a lot of text to explain how different their nonliving pFe is from the more common term used in the field, authigenic pFe. Their explanations rely on the fact that the operationally-defined Berger leach gets the pFe pool that is fully labile. What if the Berger leach also accesses some of the relatively reactive lithogenic pFe? Is that a possibility that could account for the differences and the similarity between pFe_{nonliving} and pFe_{litho}? Please clarify.

Yes, the Berger leach appears to access some portion of dust-derived pFe, as evidenced by Berger leach-accessible Fe found in aerosol samples (e.g., Shelley et al., 2018). This is discussed in lines 252-254 as a possible contribution to what has been termed the “authigenic” pFe pool. While these minerals could be chemically similar to Fe oxyhydroxides that form readily in seawater, can they be considered truly authigenic if they are formed in the atmosphere instead of *in situ*?

Lines 250-251: If dFe adsorb onto or somehow get incorporated into nonliving organic particles, they likely exist as authigenic Fe minerals, right?

While this is true in some cases, there are other instances such as fecal pellets or biological detrital exudates where Fe may not be in mineral form (e.g. if chelated by organic moieties). We have clarified:

“First, this approach may include non-mineral Fe in or adsorbed to nonliving organic particles, such as in detrital organic matter, shown above to be a significant portion of the particulate C and P pools.”

Lines 272-274: This conclusion is likely not to hold. What about adsorption, disaggregation, and redox?

Lines 291-293: This could also account for the similarity between pFe_{Bio} and pFe_{litho} at Lines 272-274.

We appreciate the commenter’s feedback and have changed the lines 272-274 to:

“Refractory pFe is chemically inert and should only be affected by particle aggregation, disaggregation, and export processes. Based on the similarity between pFe_{Auth+Det} and pFe_{Lith}, we suggest that the pFe_{Auth+Det} profile is also primarily driven by these processes.”

Lines 311-313: The summer-time dFe concentrations at BATS are much higher. I am not sure why such features were not discussed in the range of dFe.

The summertime dFe concentrations at BATS (mean in upper 100 m: 0.67 nM) are similar to those in winter at Station ALOHA (mean in upper 100 m: 0.64 nM) highlighting differences in seasonal Fe cycling between the two sites. While this is discussed in lines 311-313, we chose not to plot winter dFe concentration at Station ALOHA in Figure 5 as comparable data from BATS in winter from the BAIT project are not available and because we were not able to perform Fe uptake experiments at ALOHA during winter.

Lines 316-320: Why not present the comparisons of labile pFe between BATS and Station ALOHA? It's more meaningful than the derived parameters, which are prone to large uncertainties.

We will add the labile pFe profile comparisons. However, we are choosing to keep the comparison of the derived parameters because the primary focus of the paper is exploring different approaches to understanding what's happening within the labile particulate Fe pool and how these parameters vary, not necessarily the labile pFe pool as a whole.

Lines 329-331: What if the authors conduct similar calculations as Sofen et al. (2023) by using their average Fe:C quota at Station ALOHA and labile PP, how will the values of biogenic and nonliving pFe change? Will this affect the conclusions? From what I can tell, Station ALOHA will possibly still have higher biogenic pFe within the mixed layer, but the values below 125 m will be potentially more comparable between these two sites.

This can be approximated following our PP approach, which uses the Fe:C quota and particulate P measurements from the HOT program. As discussed in section 3.2, this estimates the biogenic pFe pool as ~200% of the labile pFe pool, meaning nonliving pFe would be ~ zero for 0-150 m on average. While we would love to provide an exact comparison to Sofen et al.'s data, different choices were necessary based on the nature of the study sites and the data available to us.

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Editorial remarks (by line #):

Lines 138-142: Figure 1a- It seems like the data point from August 2022 is the only one without any error bars. Why is that?

The error bars are smaller than the data marker. We have amended the figure caption to include:

“Data points without visible error bars have uncertainties smaller than the marker size.”

Line 260: I assume that the pFe_{Bio} hereafter is $pFe_{\text{Bio_ATP}}$. The authors should consider either pointing it out or just using $pFe_{\text{Bio_ATP}}$ to avoid confusion.

We thank the commenter for pointing this out, it has been clarified.

Line 275: Figure 3a- How variable are the labile and lithogenic pFe ? It may be helpful to plot the standard deviation at each depth with error bars.

We have added data showing the variability of labile pFe to Figure 1, and of lithogenic pFe to a supplemental figure. For further information on the variability of labile and lithogenic pFe and their drivers, we point the reader to Bates et al. (2025), where it is discussed in detail.

Lines 282-284: The authors should consider plotting this data point on Figure 4c. Additionally, to exclude a data point as an outlier, a statistical method will be preferred.

This data point is plotted on Figure 4c. We have changed the wording from “an outlier in October 2021” to “data from October 2021” to avoid confusion in our intention.

Line 285: Figure 4b- Is this data point at (0,0) real or of bad quality? I can only see one bar with nonliving pFe as 0 in Figure 4a. When is the other zero nonliving pFe observed?

We thank the commenter for pointing this out. The mean mixed layer concentration was plotted in Fig. 4a compared to the concentration at 25 m in Fig. 4b, which is why there was a difference. We have changed panel b to show the comparison between mean mixed layer concentrations of $pFe_{\text{Auth+Det}}$ and lithogenic pTi , which has a very similar correlation, to avoid confusion.

Line 285- Figure 4d- Is this correlation significant?

The correlation is not significant ($p > 0.05$), but given the role of picoeukaryotes in explaining Fe uptake rates (e.g. Fig. 1), we think this comparison is worthwhile. Indeed, if we were to exclude the anomalous October 2021 data point from the regression, the correlation would be significant ($p = 0.05$, $R^2 = 0.56$). We hope the change in lines 282-284 help to better clarify this figure. We do not wish to remove the Oct 2021 data point, merely point out that, aside from this cruise where *Prochlorococcus* numbers were significantly higher and appeared to drive a

peak pFe_{Bio} , picoeukaryotes probably play a role in determining the concentration of pFe_{Bio} .

Lines 316: Figure 5g-i: The way biogenic pFe was calculated is different between this study (ATP method) and Sofen's method. The authors should consider labeling such differences clearly in the legend or captions.

We have changed our terminology to “biotic” and “authigenic + detrital” to make this more clear, and have clarified further in the caption.

Lines 355-357: For some reason, I cannot open any of these links. However, I can find the data page on BCO-DMO. It's weird, but the authors should make sure that all of these links work during revision.

We thank the commenter for noticing this, the dash mark between BCO and DMO was causing issues when copied into a web browser. This has been fixed.

Citation: <https://doi.org/10.5194/egusphere-2025-6068-CC1>