Response to the reviewers' comments on "racing the

contributions of dust sources on deposition and phytoplankton

carbon uptake in global oceans"

Yaxin Liu a, Yunting Xiao a, Lehui Cui a, Qinghao Guo a, Yiyang Suna, Pingqing Fua,

Cong-qiang Liu^a, Jialei Zhu ^{a,*}

^a Institute of Surface-Earth System Science, School of Earth System Science, Tianjin

University, Tianjin 300072, China

* Corresponding author.

Email address: zhujialei@tju.edu.cn

Dear Editors and Reviewers:

Thanks for your letter and for the valuable comments. We carefully studied all comments and revised the manuscript accordingly. The updates in the manuscript are marked in red, which is quoted in *blue italics* in this response letter. The main revisions in the paper and replies to comments point by point are as following.

In response to the reviewers' comments, we have made the following key revisions:

1. We have expanded the Methods section to provide a more detailed expression of the limitations of our approach. 2. We have added a detailed expression of the uncertainties associated with applying the linear relationship in the non-HNLC regions and clarified their potential implications in the manuscript.

Response to the comments from <u>reviewer 1</u>

The current version of the manuscript includes substantial text additions that help understand the methodology and its limitations. Yet, some of this information is a bit "hidden" at the end of the manuscript. In my opinion, it should also appear somewhere in the methods and maybe even in the abstract. For instance, the fact that the C: Fe approach does not take into consideration other limiting (such as light) or co-limiting (such as the combination of micronutrients) factors, or the lack of data in large regions of the ocean, in which conclusions are based on heavily interpolated maps.

Response: We sincerely appreciate your suggestions, as well as your appreciation of our improved work. We have added "However, using the Fe: C ratio in phytoplankton cells to estimate the response of phytoplankton carbon uptake to dust deposition may introduce some uncertainty, because this method does not fully account for potential co-limiting factors such as light availability and the interactive effects of multiple micronutrients. The results, especially in non-HNLC regions, is only a hypothetical research results, and the uncertainty needs to be further reduced after being enriched with experimental and observational data." and "To prevent data from a single site from affecting oceanic regions on both sides of a landmass, we delineated land—sea boundaries during interpolation. However, the uneven global distribution and limited number of observations and complex land-sea boundaries could lead to interpolation uncertainty. Refining interpolation methods may reduce the uncertainty and improve estimates the impact of Fe on phytoplankton carbon uptake." in the Methods section (Lines 303-309 and 275-280) to highlight potential limitations and uncertainties.

Response to the additional comments in the reviewer's response document:

"these approaches produced less satisfactory results." --- In fact, during our research process, we tried many interpolation methods. However, other interpolation

methods couldn't yield more reasonable interpolation results. A discussion of the test results will follow below. Due to the uneven spatial distribution of observational data and the relatively small time coverage, we don't have sufficient evidence to prove that there are large spatial differences in the solubility of iron in the ocean. Therefore, we tried our best to obtain an interpolation result with relatively gentle changes. The inverse distance weighting method used in our current manuscript best meets our requirements. Although interpolation inevitably leads to uncertainties and errors, ultimately, we hope that there will be more comprehensive ocean observational data in the future to reduce the errors in our understanding of the global distribution of iron solubility in the ocean.

"most reliable performance achievable given the existing data constraints." --Lagrange interpolation is indeed an elegant and widely used method; however, for our
dataset with hundreds of unevenly distributed points across the globe, it may not be the
suitable choice. It relies on constructing a single global polynomial that can become
numerically unstable and highly oscillatory when the data distribution is irregular, and
it does not fully account for the spherical geometry of the latitude-longitude grid. We
have tested kriging, natural neighbor and spline interpolation methods, but found that
they tend to allow a single sampling point to influence areas on both sides of a land
boundary or a global uneven spatial distribution beyond our trusted range. (Fig. R1,
where a multicolor colormap was used to highlight this issue). To address this problem,
we ultimately adopted inverse distance weighting method specifically designed for
irregular data on the sphere, incorporating land-sea boundaries to ensure that land
masses properly block the influence of observations across them.

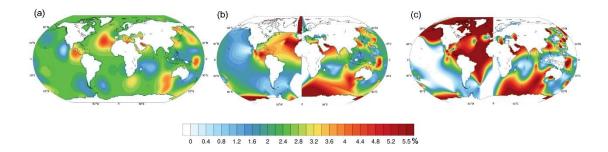


Fig. R1 The interpolated iron solubility data by using (a) kriging (b) natural neighbor (c) spline interpolation

"dust deposition is 0.7 Pg C in summer" --- This is the total amount for the global ocean in summer, not the global average, and the expression was change to "the total amount of global marine phytoplankton carbon uptake driven by dust deposition is 0.7 Pg C in summer" in the manuscript.

Response to the comments from <u>reviewer 2</u>

I want to thank the authors for taking the time to revise their manuscript and answer my questions. I think the manuscript has generally improved and I think it is almost ready for publication. I particularly thank the authors for their efforts on improving the clarity of the Figures. I have a few minor comments and one more general comment on the scope of the article.

Overall, I think the authors responded to most of my questions and comments and the manuscript is now clearer. However, I still believe that the most important and interesting contribution of this article is the dust source apportionment and quantification of each source region's contribution to the global Fe ocean supply. The additional calculations regarding C uptake are highly uncertain and rely on hypotheses that I find too strong to be convincing. If not removed, at least the results regarding other regions than the HNLC regions should be properly discussed (i.e. how does the consideration of a linear relationship between dFe and Qfe influence the results? What about luxury uptake of Fe? How to decipher the effects of Fe with those from other limiting nutrients?...).

Response: Thank you for your thoughtful and constructive comments, as well as your appreciation of our improved work. For the high nutrient, low chlorophyll (HNLC) regions where iron (Fe) is the primary limiting factor, we have added some results to emphasize the results of these regions with a high degree of credibility in Lines 533-540: "In addition, phytoplankton carbon uptake driven by dust deposition in HNLC regions is the highest in spring $(9.6 \times 10^{-2} \text{ Pg C season}^{-1})$, compared with summer $(6.5 \times 10^{-2} \text{ Pg C season}^{-1})$, autumn $(6.3 \times 10^{-2} \text{ Pg C season}^{-1})$, and winter $(2.1 \times 10^{-2} \text{ Pg C season}^{-1})$. Dust-driven phytoplankton carbon uptake is the highest in HNLC regions of the NP across all seasons, accounting for 86.1% in spring, except in winter. In winter, phytoplankton in the SO contribute the most to dust-driven marine carbon uptake in HNLC regions, with $1.1 \times 10^{-2} \text{ Pg C } (\sim 50.2\%)$, while the NP accounts for $\sim 45.6\%$."

Meanwhile, we additionally emphasized the uncertainty of the results in non-HNLC regions to indicate that this result is only a hypothetical research results, and the uncertainty needs to be further reduced after being enriched with experimental and observational data. We have added "However, using the Fe: C ratio in phytoplankton cells to estimate the response of phytoplankton carbon uptake to dust deposition may introduce some uncertainty, because this method does not fully account for potential co-limiting factors such as light availability and the interactive effects of multiple micronutrients. The results, especially in non-HNLC regions, is only a hypothetical research results, and the uncertainty needs to be further reduced after being enriched with experimental and observational data." in Lines 303-309 to acknowledge the limitation.

Although some non-HNLC regions may be co-limited by multiple micronutrients, Fe can also alleviate nutrient limitation, and dust deposition can stimulate nitrogen fixation, thereby promoting phytoplankton growth and influencing the carbon cycle. Therefore, despite the relatively larger uncertainties compared to HNLC regions, we conducted this estimation to provide a potential global range of phytoplankton carbon uptake responses to dust deposition as a reference for future research: "The estimation of global marine phytoplankton carbon uptake attributed to dust deposition is 5.6 Pg C yr⁻¹, which may be overestimated due to the assumption that every grid where dust deposition occurs over the ocean responds to its Fe supply. Therefore, the actual annual marine phytoplankton carbon uptake due to dust deposition worldwide is likely between 0.2 Pg C yr⁻¹ and 5.6 Pg C yr⁻¹".

Moreover, the linear relationship between cellular Fe quota (Qfe) and dissolved Fe (dFe) may not necessarily apply to all phytoplankton types across the global ocean. As the experiments in Sunda et al. (1995) were conducted on selected coastal and oceanic phytoplankton clones, there are inevitably some spatial and taxonomic limitations, which could introduce uncertainties into our assessment. To clarify this uncertainty, we have added the statement "We adopt a parameterization scheme from previous studies, assuming a linear relationship between cellular Fe: C ratios and dFe

concentrations and using a piecewise linear formula to describe this dependency. However, using this approach to assess global marine phytoplankton carbon uptake in response to Fe supplied by dust deposition has certain limitations. The linear relationship reported in the original experiments was derived from a limited number of phytoplankton species under controlled conditions, and it is uncertain whether it applies universally to all phytoplankton groups across diverse oceanic regions, given the physiological and ecological differences among species." in Lines 790-798.

Using Fe: C ratio to estimate phytoplankton carbon uptake influenced by dust-borne Fe does not explicitly account for potential luxury uptake of Fe. However, we think that the omitting this process might not cause a substantial bias into our large-scale estimates, as luxury uptake typically occurs in estuaries, nearshore areas, or regions with intense upwelling, where such Fe-replete conditions are generally short-lived and cover only a small fraction of the global ocean. To better acknowledge this limitation, we have added "Moreover, the approach we used does not explicitly account for luxury uptake of Fe, in which cells may continue to accumulate intracellular Fe beyond what is required for immediate growth. Ignoring this process introduces uncertainty in the assessment of phytoplankton carbon uptake, particularly during transient high-iron events such as dust deposition, riverine input, or upwelling." in the manuscript of Lines 798-802.

Reference

Sunda W. G. & Huntsman, S. A.: Iron uptake and growth limitation in oceanic and coastal phytoplankton, Mar Chem, 50, 189-206, 10.1016/0304-4203(95)00035-P, 1995.

Minor comments:

Abstract

Line 30, remove "which is an"

Response: Thanks for the suggestion. The phase "which is an" in Line 30 was removed.

Line 33 remove "phytoplankton"

Response: Thank you for your suggestion. The word "phytoplankton" in Line 33 was removed.

Line 39 please add "11.1 Tg yr-1 of total iron ..." to clarify.

Response: We appreciate your suggestion. "11.1 Tg yr⁻¹ of **total** iron ..." is added in Line 39 to clarify.

Lines 39-40: remove "promoting 5.6 Pg C yr⁻¹ of carbon uptake by marine phytoplankton."

Response: Thank you for your suggestion. The expression "promoting 5.6 Pg C yr⁻¹ of carbon uptake by marine phytoplankton" in Lines 39-40 was removed.

Methods:

Sentence lines 226 and 238 are the same, remove one.

<u>Response:</u> Thanks for the suggestion. The sentence "Consequently, we filtered the Fe solubility data to retain only values below 6.0%." in Line 238 was removed.

Line 286: I cannot find the Sunda (1995) reference in the reference list. Is the linear relationship between Qfe and dFe valid everywhere and for all phytoplankton groups? I think this assumption needs to be justified.

Response: Thank you for your suggestion. We sincerely apologize for the omission of the reference Sunda (1995):

Sunda W. G. & Huntsman, S. A.: Iron uptake and growth limitation in oceanic and coastal phytoplankton, Mar Chem, 50, 189-206, 10.1016/0304-4203(95)00035-P, 1995.

Sunda et al. (1995) provided key evidence for a linear relationship between cellular Fe: C ratios and dFe concentrations based on experiments with selected coastal and oceanic phytoplankton clones. However, their study cannot fully confirm whether this relationship applies to all phytoplankton groups and oceanic regions. We have therefore added the related expression to note the limitation (Lines 790-798).

Results:

Lines 442-443: "A small Fe: C ratio in phytoplankton cells indicates large marine phytoplankton carbon uptake driven by the same amount of Fe supply." Replace 'small Fe: C ratio' by something like 'An Fe: C ratio lower than the optimal value'. It's not the absolute value that counts but the distance to optimal Fe: C ratio.

Response: Thank you for your suggestion. The sentence was change to "An Fe: C ratio in phytoplankton cells lower than the optimal value indicates large marine phytoplankton carbon uptake driven by the same amount of Fe supply."

In the next sentence: "Increased Fe supply usually can enhance carbon uptake by phytoplankton, but only soluble Fe is bioavailable," Add "and Fe: C is lower the optimal value".

Response: Thank you for your suggestion. The sentence was changed to "Increased Fe supply usually can enhance carbon uptake by phytoplankton, but only soluble Fe is bioavailable and Fe: C is lower the optimal value".

Lines 450-454: The 2 sentences seem a bit contradictory. I think you can remove the 1st one.

Response: We appreciate your valuable suggestion. We sincerely apologize for any unintended ambiguity in our original wording. Our intention was to convey that we provide not only the global assessment results but also the results specifically for the HNLC regions. We have revised these two sentences to clarify our intended meaning. "We estimated global phytoplankton carbon uptake induced by dust deposition using the Fe: C ratio in phytoplankton cells. Because Fe is the primary limiting nutrient in HNLC regions, we additionally provided a separate estimate for these regions." was updated in Lines 461-464.

Line 456: add "in HNLC regions at the end of the sentence".

Response: Thank you for your suggestion. However, this result refers to the global ocean rather than the HNLC regions. We sincerely apologize for the confusion caused by our unclear wording. We have now revised the sentence as follows: *Our simulations indicate that annual dust deposition supplies 11.1 Tg of Fe to the global ocean, of which 0.4 Tg is dFe, driving a carbon uptake of 5.6 Pg C yr⁻¹ by phytoplankton.*

In methods, you say that the 3 HNLC regions are SO, EP and NP. And then you say that you only calculate the C uptake by phytoplankton driven by dust Fe in HNLC regions (because in other regions, the linear relation between Qfe and dFe doesn't hold). But lines 458-459, you mention Fe: C ratio in phytoplankton cells in the EA region. According to the methods, the calculation of the ratio shouldn't hold for this region (which receives very large dust fluxes and is not Felimited). I think the interpretation of results for regions other than the HNLC region is not justified and should be removed (lines 455-485). Maybe this can be discussed in the paper. Same for the presentation of results on the seasonal uptake (lines 486-512).

Response: Thank you for your constructive comments. We fully agree that assuming a linear relationship between Qfe and dFe across all regions introduces uncertainty into our assessment. In our study, we present the impacts of dust deposition on phytoplankton carbon uptake not only in HNLC regions but also across the global ocean. Although some non-HNLC regions may be limited by multiple micronutrients (e.g., Fe, nitrogen, phosphorus), dust deposition can stimulate nitrogen fixation, thereby alleviating nutrient limitation and potentially promoting phytoplankton growth. As a result, the linear relationship between Qfe and dFe may also be applicable in certain non-HNLC regions. Our intention is primarily to provide a global range of phytoplankton carbon uptake responses to dust deposition, acknowledging that more detailed considerations could yield more precise estimates. We have incorporated this source of uncertainty into the manuscript (Lines 303-309) and provided additional results for the HNLC regions (Lines 533-540).

Lines 513-517 are exactly the same as lines 450-454.

Response: Thank you for your kind reminder. To avoid redundancy and ensure conciseness, we have replaced the content in Lines 523-524 with the following sentence: "Recognizing Fe as the primary limiting nutrient in HNLC regions, we provided a separate estimate for these regions.".

Again, phytoplankton may respond to dust addition in LNLC regions, but such areas are not Fe-limited and the linear relationship assumed between Qfe and dFe may not be verified in such regions. I cannot find the Sunda, 1995 referenced in the methods to justify this link in the reference list.

Response: We sincerely apologize for the omission of the reference Sunda (1995) again, and have added it in the reference list.

Discussion:

The hypotheses on around Equation X to calculate phytoplankton C uptake should be discussed. Is the relationship between Qfe and dFe always linear? How does Fe phytoplankton uptake change with luxury uptake? Are there other hypotheses and works than the rather old Sunda 1995 paper on Qfe?

Response: We appreciate your suggestion. Sunda et al (1995) proposed the linear relationship between cellular Fe: C ratios and dFe for a limited set of phytoplankton clones in controlled experiments. However, their study was necessarily limited in terms of spatial coverage and species diversity, and thus cannot fully demonstrate whether this linear relationship holds universally across all phytoplankton groups and oceanic regions. We have noted the limitation in Lines 790-798.

Using Fe: C ratio to estimate phytoplankton carbon uptake influenced by dustborne Fe does not account for potential luxury uptake of Fe. However, we think that the omission of luxury uptake might not cause a substantial bias in our large-scale estimates of phytoplankton carbon uptake. Because luxury uptake of Fe usually temporarily occurred in estuaries, nearshore areas, or regions with intense upwelling, and such Fe-replete conditions occupy only a small fraction of the global ocean and are often transient. We have added a corresponding statement in Lines 798-802 of the manuscript to highlight this limitation.

The parameter settings in the equation of calculating Fe: C ratio is following Wiseman et al. (2023), which is a recent study focusing on Qfe.

Reference

Sunda W. G. & Huntsman, S. A.: Iron uptake and growth limitation in oceanic and coastal phytoplankton, Mar Chem, 50, 189-206, 10.1016/0304-4203(95)00035-P, 1995.

Wiseman, N. A., Moore, J. K., Twining, B. S., Hamilton, D. S., and Mahowald, N. M.: Acclimation of phytoplankton Fe: C ratios dampens the biogeochemical response to varying atmospheric deposition of soluble iron, Global Biogeochem. Cycles, 37, 10.1029/2022GB007491, 2023.

Fig4. If possible, please put boxes around each ocean region for clarity

<u>Response:</u> Thank you for your suggestion. We have added blue lines to the figure, in combination with the land-sea boundaries, to indicate different ocean regions.

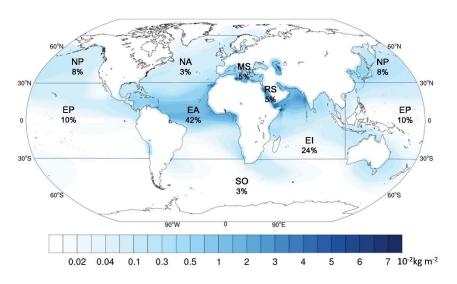


Fig. 4 The spatial distribution and proportion of the global five-year average dust deposition. Blue lines together with land-sea boundaries indicate different ocean regions. The percentages express the proportions of annual dust deposition in each ocean to global ocean (Oceans: NP - North Pacific Ocean; NA - North Atlantic Ocean; MS - Mediterranean Sea; RS - Red Sea; SO - Southern Ocean; EP - Equatorial Pacific Ocean; EA - Equatorial Atlantic Ocean; EI - Equatorial Indian Ocean.)

Once again, thank you very much for all the comments and suggestions.