

## Authors' Response to Review Comments (Manuscript Number: 'essd-2021-259')

We are very thankful to all the reviewers for their thorough and constructive reviews. We have addressed all the issues raised by them and used all their suggestions and comments on the manuscript. We believe our revision improved the manuscript and we hope it pleases the three reviewers.

Specific replies to both general and specific comments for the three reviewers are given in 3 separated files in our reply to each reviewer's comment, and in a single file here.

We would like to generally address several remarks common among the three reviewers in relation to different aspects of new parameters to be included in individual data set.

It is worth mentioning that the amount of data that could be additionally reported in the compilation is very extensive due to the wide applicability that has been shown for  $^{234}\text{Th}$  along the years. However, for the sake of feasibility, we needed to set boundaries to the scope of the compilation and left some parameters out of this first version of the compilation. However, we would like to emphasize the dynamic spirit behind this effort, which will be periodically reviewed and extended. Both new data and information on the availability of sampling methods used complementarily to the  $^{234}\text{Th}$  technique will be included in future versions of the compilation, both evaluating new useful parameters arisen from novel research and following suggestions by researchers.

All the reviewers have provided very good suggestions about parameters that must definitely be implemented in the compilation. Especially considering that these parameters are now included in the data sets of nowadays cruises and programs.

We will definitely include these parameters as improvements in future datasets, as the idea of this compilation is to make annual reeditions. PANGAEA has designed a specific structure for the compilation that allows a customized updating and we have created the webpage ("Sea of Thorium", coming soon) to stimulate the reception of new contributions.

Furthermore, in the reviewed version of the manuscript we have modified Section 5 and it includes now a subsection that it is related with the several aspects of the compilation that will be upgraded in the next edition, following the reviewers' suggestion. We acknowledge the absence of some useful parameters, such as the CTD auxiliary sensors for fluorescence and PAR pointed out by the reviewer as gaps and discuss which parameters will be included in next versions of the compilation.

### **Reviewer #2**

**Review of "Revisiting five decades of  $^{234}\text{Th}$  data: a comprehensive global oceanic compilation" by Ceballos-Romero et al.**

#### **General comments**

This paper provides a global dataset on  $^{234}\text{Th}$  data in the ocean, accompanied by additional parameters used in the application of the  $^{234}\text{Th}$  approach to derive export fluxes, mainly particulate carbon export fluxes. This effort comes after the compilation of  $^{234}\text{Th}$  fluxes by Le Moigne et al. (2013) and the recent compilation of POC: $^{234}\text{Th}$  ratios by Puigcorbé et al. (2020), both published in ESSD, therefore I believe it is well suited within the scope of ESSD. This compilation will be very useful, as the authors mention, to better constrain the contemporary ocean carbon uptake by providing easy access to all the  $^{234}\text{Th}$  data collected over the last 50 years.

This is a major effort that has brought together a large number of datasets. The information provided regarding the global dataset, what parameters are included, how they are organized and how to access them is clear for the reader. Additionally, the authors have added a detailed history of  $^{234}\text{Th}$  and its use as particle tracer from its beginnings until today, and even looking into future programs.

I did enjoy reading the entire manuscript, but I also believe it should be streamlined because there is a bit of repetition between sections and within the same section. As a general comment, there were several typos and formatting errors along the manuscript, repeated words and sometimes sentences appeared out of the blue or were unfinished/misplaced. I had the feeling that the manuscript needed an additional proof reading by the authors prior to submission, but I tried to provide detailed comments when I spotted some of these issues.

We are very grateful to the reviewer for its thorough review, attending both formatting issues and the manuscript content itself. We apologize for all the formatting issues and typos that were overlooked in our previous version. We understand that such compilation should be perfectly edited. We employed time and effort to do that, including more than four people in the task. However, we see now, that our revision was far from perfect. That is why we specially appreciate the effort made by the reviewers to polish text, Figures and compilation itself. We had made an additional effort this time to improve the manuscript and the content.

We strongly believe the manuscript is greatly improved after implementing all the reviewer's suggestions.

Particular to Section 5, this section does not always discuss the gaps in the dataset, but it expands on issues that, although very important, most reader will be aware of them and they are not real gaps of the dataset, at least not in the way they are presented. To be honest, this section feels like it is a section to "advertise" the work of the first author (7 citations out of 8 in page 24 are Ceballos-Romero et al.), which it is actually really good, but I don't think it belongs to the "gaps of the dataset" section. Maybe move it to the overview of the dataset or to the discussion. If the authors decide to keep it in Section 5, as mentioned in my detailed comments, those particular paragraphs should be more to the point.

We agree with the reviewer and, as so, the entire section 5 has been rewritten given it a completely different approach more aligned with the take home message of the manuscript (see new section 5, from line 791 to 867).

The <sup>234</sup>Th timeline part (4. Discussion) would largely benefit from a figure showing a timeline and the key dates/milestones listed in the text. I think the readers and future researchers interested in <sup>234</sup>Th would appreciate having Section 4 summarized in a nice timeline figure.

We consider including this figure a very good idea and, as so, it has been implemented in the reviewed version of the manuscript (see new figure 3).

Also related to Section 4, I have provided specific comments below, but I believe that, even though this manuscript has a whole section of "storytelling", without providing additional data (which I enjoyed reading), the division by era's should provide some useful information for scientific purposes (e.g., period of years that the era lasted, using the name of the main program of the era, or stating what was done during such era). Although it is quite "artistic" to have named them by well-known novels linked to the ocean, those titles don't serve a clear purpose (they don't clearly describe the actual era) and changing their names might be more adequate when publishing a research paper.

We appreciate the reviewers' comment on this regard. Changes have been implemented, including the duration of each specific era and a description for the main theme that characterized it. We have kept name of the eras after titles from seminal ocean-related books, but included a quote from each one of them to evince each one of the choices made for the era, which follow an intention are not random.

Moreover, in that line, I think the manuscript, since it is a research publication, should provide some actual data, not just referring to the number of publications and number of datapoint, but actual data on <sup>234</sup>Th, similar to what was done in the two previous compilations related to <sup>234</sup>Th (see Fig. 1, 3, 7 and Table 2 from Le Moigne et al. (2013) or Fig. 2, 3 and 5 from Puigcorbé et al. (2002)). This manuscript does not provide data on <sup>234</sup>Th, just on the number of studies/cruises/stations/parameters. The authors could include a table with <sup>234</sup>Th concentrations (tot/part/diss) similar to Table 2 from Le Moigne et al. (2013) or in a figure similar to Fig. 4 from Puigcorbé et al (2020) or something completely different, but they should report actual <sup>234</sup>Th

data/values, not just data points. A description of the number of datasets, studies and locations is already provided in the abstract of the dataset in the PANGAEA repository. In this manuscript, the authors should provide something additional to it, that goes beyond explaining the structure of the dataset, the percentage of studies that do one thing or another, or the history of  $^{234}\text{Th}$ .

We agree with the reviewer that the previous version of the manuscript lack data. Accordingly, we have included 2 new figures with data in section 6 (see line868).

Despite agreeing on the fact that this dataset is very necessary, and it will be very useful, I have two (relatively major) issues with it:

- Not all the studies reported analyzed POC, some analyzed PIC+POC. There is no reference made regarding this "problem". Either in the manuscript or in the dataset (or both), a comment should be made to indicate if the data reported is truly POC, or if it is not, why it is still considered as POC (e.g., negligible amount of PIC).

Only data reported as POC in the sources consulted have been reported as POC data. And equivalent for PIC data.

- The authors have compiled additional data, when available, on water temperature, salinity,  $^{238}\text{U}$ , and particulate nitrogen, and if they can, they also report the bloom stage. However, the authors have omitted a critical parameter for the application of the  $^{234}\text{Th}$  method. This is particularly surprising since one of the authors has recently (2020) published a PNAS paper entitled "*Metrics that matter for assessing the ocean biological carbon pump*". It has been shown that the integration depth has an impact on the magnitude of the fluxes (the strength of the biological carbon pump) and therefore, we should be using reference depths related to the euphotic zone (or primary production zone (PPZ); Owens et al. (2015)). I understand that not all the studies, particularly in earlier days, provided this information, but recently, the integration depth is no longer fixed and authors explain what kind of reference depth they use to integrate the fluxes. I believe fluorescence and/or PAR data to define the euphotic zone or the PPZ (or providing those depths directly) should be added in the dataset, the same way that temperature and salinity are. With that addition, in the manuscript it should also discuss the importance of the integration depth and cite the Buesseler et al. (2020b) paper in a more appropriate line than where it is currently cited (L778). If this dataset is expected to be "alive" and authors want to be contacted to update it by adding new data, having this kind of input is crucial for future applications of the  $^{234}\text{Th}$  method.

The reviewer is right about this gap in the compilation, integration depth is proved to be a crucial parameter for an accurate use of  $^{234}\text{Th}$  disequilibrium to evaluate POC export. However, only very recent papers acknowledge this issue and include details of the integration depth. On the other hand, as it is stated in the PNAS paper "*Metrics that matter for assessing the ocean biological carbon pump*", the choice of the integration depth is not straightforward. This way we do not think we should select one specific integration depth for each data set, as that will imply that we are interfering in the interpretation of the data. To this regard, the inclusion of the fluorescence and/or PAR data, as suggested by the reviewers will allow future users of the compilation to estimate the integration depth. We mention in Section 5 that CTD auxiliary sensors for fluorescence and PAR will be some of the parameters to be included in next versions of the compilation. This way, we expect that this will also serve the users of the compilation as a reminder of one important parameter to include together with the  $^{234}\text{Th}$  results.

Finally, regarding the datasets available in PANGAEA, I found that <https://doi.pangaea.de/10.1594/PANGAEA.918125> provides the metadata information and there is a table with parameters shown in that file. However, <https://doi.pangaea.de/10.1594/PANGAEA.937414> contains the actual data but there is no table of parameters there. Please consider including such table for clarity (even if it is a long one), since not all the "parameters" might be clear to the reader/user, e.g., "integrated\_depth\_real\_particle (m)". Notice my comment below for Table 1, where the starting and end date

for the sampling period have been reversed in some cases. The dates are also incorrect in the dataset. I noticed some from Table 1 but please double check them all again in the dataset as well.

We thank the reviewer for his comments. We have already made this request to PANAGEA for the Table referred to by the reviewer also be included in <https://doi.pangaea.de/10.1594/PANGAEA.937414>.

#### **Detailed comments:**

L11-16: “However, it was.... the biological pump.” This is a very long sentence. I don’t think that level of detail is necessary for the abstract. The information detailed here is already discussed in detail in the manuscript. Please, simplify it.

In the reviewed version of the manuscript this sentence has been simplified (see abstract).

L 20-21: “...that include two size classes;...”. By reading the rest of the paper more por sizes (e.g., <70 µm) are mentioned and reported. Maybe just finish that sentence after “filtration method” in the abstract.

Done.

L23: “...included, including...”. Try to find another word to avoid repetition

Done.

L38-39: I understand where that is coming from but production rates of anthropogenic radionuclides might not always be that well characterized or can vary over time.

We agree with the reviewer that this clarification is important and in the reviewed version of the manuscript we have solved this issue by specifying that it is the case for naturally occurring radioactive elements (see line 35).

L41: I would even say more than just years (paleo tracers).

In the reviewed version of the manuscript, we have replaced years by millenniums (see line 38).

L44: “use” instead of “used”

Corrected.

L45: Delete “a” from “a <sup>234</sup>Th”

Done.

L53: “when high scavenging by particles”. Correct, there is something missing.

Corrected.

L55-56: Instead of citing that many papers without a clear reason behind their selection, maybe cite Le Moigne et al. 2013, which provides a compilation of studies using the <sup>234</sup>Th approach to derive estimates of POC export.

Done.

L57-58: PIC and BSi abbreviations are not needed. PIC is only used one more time, in L 474, where it is also defined, and BSi is not used anywhere else in the manuscript.

PIC and BSi abbreviations have been removed.

L61: "...is in part..." seems to imply that there is another part, but it is never mentioned.

Corrected.

L63-64: Not sure how necessary is to provide the editors' names for the special issue.

Editors named have been erased (see line 59).

L86: Maybe delete the "only"

Deleted.

L88: Missing Th after <sup>234</sup>

Corrected.

L93: "...and useful metadata to be used". Change words to avoid repetition of "use..."

Done.

L114: Add the GEOTRACES website

Done.

L124: "...spanning all oceanic regions" maybe add "across": "spanning across all oceanic regions"

Added.

L129: Do you mean when reported by the original authors? There could be issues on the way the different authors, over the different years, have decided if the conditions where non-bloom, pre-bloom, bloom or post-bloom. Please discuss it a bit more.

We have compiled all data as reported by the original authors, except for conversion of units when necessary for the sake of consistency between all datasets. We did not assess the bloom stage but instead included the information as indicated by the original authors when provided. This information has been stressed out in the reviewed version of the manuscript. We recognise the way we assigned the four bloom stages we distinguish (i.e., "non-bloom", "pre-bloom", "bloom" and "post-bloom" conditions) was not clear in the previous version of the manuscript and has been clarified in the reviewed version (see lines 119-129).

Additionally, we have acknowledged the lack of evaluation of the bloom stage at the sampling moment for each of the studies compiled following a standard criterion as a gap in the compilation and proposed such an improvement for future version of the compilation (see new section 5).

L145: 210 should be a superscript. Same for L435, L537, L610, L665 and L666

Corrected throughout the manuscript.

L158-159: "...the authors were directly contacted for data is indicated in the localization." Please rephrase for clarity.

Done.

L169: It should be  $\mu\text{mol}$  instead of  $\mu\text{m}$ . Same for L178

Corrected.

L169-170: "...and include... in which we included". Try to find synonyms to avoid repetition of words

Modified.

L174: Correct "The particles' sampling..."

Corrected.

L180: "Measurement uncertainties in the data points as provided by the data measurer...". Please rephrase for clarity.

Corrected.

L200-203: Delete or move to the section "General overview", rephrase or delete "here" since you refer to "here" for the entire manuscript, for a particular section that you are discussing and for a particular section that you are about to discuss, which makes it confusing.

Done.

L203: "on in" correct it

Corrected.

L208: "larger" instead of "lager"

Corrected.

L209: "During these 50 years of study, we have....". Maybe rephrase so it doesn't sound like you took 50 year to write this manuscript. Something like "During these 50 years of  $^{234}\text{Th}$  data" similar to the title.

Corrected.

L209 and L211: "14 major ocean programs" vs "as part of one of these 13 programs"

14 has been corrected to 13 (see line ).

L211: "as reported by the authors". I'm not sure which authors are you referring to, the ones of that major program? Yourselves? If it is yourselves there is no need to have that sentence. If it is referring to the authors of those datasets, I don't think it is correct, because they could not tell that their dataset represented 30% of the total datasets.

The reviewer is right, that sentence wasn't correct and has been erased (see line 217).

L212: "reported" instead of "compiled"

Corrected.

L214: delete the - after TSS

Done.

L218: “time-series stations” should be TSS as defined in L214.

Corrected.

L222: “least” instead of “last”

Corrected.

L222-223: “re-occupied” should be “reoccupied”, as in L223

Corrected.

L225-231: Missing the degree (°) symbol for the coordinates of all the time series stations

Corrected throughout the manuscript.

L237: POC(PON) to  $^{234}\text{Th}$  ratios have been defined previously (e.g., 144) as POC(PON): $^{234}\text{Th}$ .

Corrected.

L247: Correct “datasets”

Corrected.

L253: Correct “expeditions to the”

Corrected.

L258: “is not significant of the number”. Rephrase for clarity.

Done.

L263: Correct “experiments”

Corrected.

L264: Delete “started in 1969” or add “which”

Added.

L271: it should be “of” instead of “on”

Corrected.

L276: Delete #

Done.

L288: Same as previous comment from L237

Corrected.

L301: it should be “sample” instead of “sampled”

Corrected.

L304: “yet maximize...” add “the number of locations sampled”

Added.

L305: “were” instead of “was”

Corrected.

L308: Correct for consistency “POC(PON):<sup>234</sup>Th”

Corrected.

L317: “...in situ pumps, bottles and...” remove the “and” and change for a comma.

Corrected.

L324-326: Missing reference to “Not et al (2012). Conservative behavior of uranium vs. salinity in Arctic sea ice and brine. *Marine Chemistry* 130-131; pp. 33–39. doi:10.1016/j.marchem.2011.12.005

Reference added and text modified accordingly (see lines 344).

L337: “...in an oceanographic expedition...”, delete “field” is redundant.

Done.

L340: I would recommend changing the names of the eras in order to actually provide some information about each era. Naming era 1 “The Old Man and the Sea” doesn’t say much about what happened during this era. I would provide the periods of each era and, if possible, something more such “JGOFS”, “GEOTRACES” or something related to the main advances of each era, e.g., “the beginning of <sup>234</sup>Th as tracer of POC export” or in that line.

We appreciate the reviewers’ comment on this regard. Changes have been implemented, including the duration of each specific era and a description for the main theme that characterized it. In the reviewed version of the manuscript, we have kept name of the eras after titles from seminal ocean-related books, but included a quote from each one of them to evince each one of the choices made for the era, which follow an intention are not random.

L344: Bhat had co-authors in that first <sup>234</sup>Th publication.

Corrected.

L345-347: “Bhat et al. (1969)... part of the total <sup>234</sup>Th.” What’s the purpose of this sentence? It is interesting but it appears out of the blue and it is not discussed any further in a way that it is linked to the following sentences. Maybe rephrase between L345 and L354 to facilitate the flow.

Rephrased (see lines 370-380).



L353: It sounds strange to say that you use “this radionuclide” (referring to  $^{234}\text{Th}$ ) to study the scavenging of thorium. All the thorium isotopes are particle-reactive.

Radionuclide replaced by radioisotope.)

L358: Rephrase “realizing the relevance to the downward  $^{234}\text{Th}$  flux” for clarity.

Rephrase.

L367: Correct “increase” instead of “increased”

Corrected.

L371: Correct “prolong” instead of “prologue”

Corrected.

L376: It should be “ $\text{MnO}_2$ ” (the 2 should be a subscript)

Corrected.

L380: Linked to my comment from L340, here the authors refer to JGOFS era. Avoid confusions.

All references on the manuscript to the JGOFS era (which were 3 in total in the previous version of the manuscript) has been erased in the reviewed version of the manuscript for the sake of avoiding confusion with the eras that we have defined in this paper for the  $^{234}\text{Th}$  timeline.

L382-385: Missing the degree ( $^\circ$ ) symbol for the coordinates of all the time series stations

This issue has been corrected throughout the manuscript.

L388-389: Avoid repeating “introduced”

Changed.

L391: “...a precipitate that serves for removing Th...”. Change to “...a precipitate that removes Th...”

Changed.

L393: Maybe refer to it as “For the  $\text{MnO}_2$  cartridges technique”, since  $\text{MnO}_2$  is the method being used currently, although mainly without the cartridges.

Done.

L394: Same comment as L376

Corrected.

L394: Delete “(MnA and MnB)”. There is no graph and, therefore, there is no need to specify the name of the cartridges (which could also be “named” Mn-I and Mn-II or Mn-1 and Mn-2 or many other ways).

Deleted.

L394: Did you mean “and gamma count them”? Not sure it is needed here since gamma counting is mentioned in L398.

Text erased.

L397: Why the differentiation between MnO<sub>2</sub> and Mn cartridges? We have only been talking about cartridges with fibers impregnated with MnO<sub>2</sub>.

The reviewer is right, such a differentiation between MnO<sub>2</sub> and Mn cartridges is pointless. Text has been modified to always refer to MnO<sub>2</sub> cartridges for consistency (see lines 424-436).

L398: Refer to it as MnO<sub>2</sub> cartridges for consistency. Otherwise, modify the text above and refer to Mn cartridges the whole time, after specifying that those are cartridges with fibers impregnated with MnO<sub>2</sub>.

Text has been modified to always refer to MnO<sub>2</sub> cartridges for consistency (see lines 424-436).

L403: The reference to the volumes sampled doesn't seem to have a purpose. It is not discussed or linked in any way to the rest of the paragraph. It provides information but I don't think it is relevant as it is presented right now, plus the sample volume reference is not done for the studies cited between L404-410.

The sentence referred to volumes sampled has been erased.

L407: Correct “where” instead of “with” or say, “with a predominant particulate size class cut-off of 0.45 μm”. If it is just for one study, can you say it is the predominant size class? Were there others in that same study?

0.45 μm was the predominant pore size cut-off for the majority of the studies that carried out particulate analyses. This sentence has been rephrased to clarify this point following reviewer comment on L407 and L408-410 (see line 444).

L408-410: “Note that... McKee et al., 1986.” This sentence seems not to match the size class cut-off of 0.45 μm mentioned just before. Is this operational distinction applying to the previous studies cited between L403 and L405? If that's the case, move it there. If not, remove or integrate better to the paragraph.

See response to comment on L407.

L435: Same comment as L145: 210 should be a superscript

This issue has been corrected throughout the manuscript.

L436: Same comment as L340

Same response as before. We appreciate the reviewers' comment on this regard. Changes have been implemented, including the duration of each specific era and a description for the main theme that characterized it. In the reviewed version of the manuscript, we have kept name of the eras after titles from seminal ocean-related books but included a quote from each one of them to evince each one of the choices made for the era, which follow an intention are not random.

L437: The authors keep referring to the JGOFS era: “...what we are calling the “JGOFS era” (1989-2003)” but this is not an actual era in their manuscript, not by period of time nor by name. Please avoid confusion by naming the eras appropriately to refer to something that has a meaning linked to the history of <sup>234</sup>Th.

All references on the manuscript to the JGOFS era (which were 3 in total in the previous version of the manuscript) has been erased in the reviewed version of the manuscript for the sake of avoiding confusion with the eras that we have defined on this paper for the  $^{234}\text{Th}$  timeline.

L443-444: It is odd to cite a reference from 2008 in the 2<sup>nd</sup> era (1992-2001). Moreover, the following sentence starts with “Accordingly, ...”. Please cite the first paper discussing this topic, not one from 2008.

Done.

L446 or 447-448: Adapt the format of equation 1 to match the format of the different parameters in the description (e.g, add 234 as a superscript, keep consistency with italics and non-italics, etc.)

Done.

L477: Add the units of the  $^{234}\text{Th}_{\text{flux}}$  as done for the ratio.

Done.

L452: It should be “Element:  $^{234}\text{Th}$ ” instead of “POC:element”. I don’t think the italics are needed.

Corrected and italics removed.

L453: “its” should be “their”

Corrected.

L453: Delete “dependent” since it already says that they vary with both depth and particle size. Or say that they are “depth and particle size dependent”

Done.

L454-456: “...can be calculated from the disequilibrium between the  $^{238}\text{U}$ - $^{234}\text{Th}$  resulted from particle sinking...” This has already been said a couple of times before. Simplify: “ $^{234}\text{Th}$  flux can be calculated by evaluating...”

Done.

L456: Why “of the dissolved  $^{234}\text{Th}$ ”? It has been done with total  $^{234}\text{Th}$  (see Savoye et al. 2006-Marine Chemistry and references in there).

The reviewer is right. This clarification to dissolved  $^{234}\text{Th}$  is wrong and has been removed.

L458: Add “a”: “only a single  $^{234}\text{Th}$  profile”

Done.

L458: Remove the comma after “NSS model”

Done.

L458-460: Simplify it. Too many sentences within one sentence. “...when activity gradients can be assessed in time thanks to repeated sampling, ideally over the course of 2 to 4 weeks.”

I think the authors should refer to Resplandy et al. (2012)-DSR-I work (<http://dx.doi.org/10.1016/j.dsr.2012.05.015> )

Done.

L473: PIC abbreviation is not required since it isn't used anymore. Also, notice that PIC and POC fluxes are elemental fluxes of the same element, C.

Abbreviation erased and text corrected.

L476: It should be “MnO<sub>2</sub>” (the 2 should be a subscript)

Corrected.

L481: Missing the degree (°) symbol for the coordinates of the time series station

This issue has been corrected throughout the manuscript.

L487: “with large volumes (2500-3500 L) passing...” instead of “passed”

Corrected.

L498: Why “we initiated”? Delete

Deleted.

L489: Why “and filtered”? Delete

Deleted.

L492-494: There are 7 studies that sampled 2 size fractions but there are only 4 papers cited, 2 for 53 µm screens and 2 for 70 µm screens. Either cite the 3 remaining studies or add “e.g.,” when citing the other studies.

Corrected: all the references are indicated now (see lines 535-542).

L494: How many studies used sediment traps? 7 studies did size fractionation, it would be good to mention those that used sediment traps, for consistency.

This information and references have been added (see line 540).

L503: Another reference to the “JGOFS era”. Maybe consider this as one of the eras then?

All references on the manuscript to the JGOFS era (which were 3 in total in the previous version of the manuscript) has been erased in the reviewed version of the manuscript for the sake of avoiding confusion with the eras that we have defined in this paper for the <sup>234</sup>Th timeline.

Note that we do not consider JGOFS as one of the eras in the manuscript because we are discussing the history of <sup>234</sup>Th, not the history of oceanography, and JGOFS was not conceived at all to focus on the study of <sup>234</sup>Th. It simply happened as part of one JGOFS cruise.

L505: “at” instead of “in”

Corrected.

L506: Based the era period described by the authors (1992-2001), the NABE study should not be included here. It's not part of the overlap, it's overlapping with era 1, but not with era 2.

We have chosen as a criterion to distinguish eras seminal publications related to 234<sup>Th</sup> that changed the course of the technique. In this part of the text, we are presenting the papers published during era 2, which is based on the criteria just mentioned, to which the year in which sampling took place is indifferent. We agree with the reviewer that this point was not clearly explained in the previous version of the manuscript, and it seemed we were mixing events from different eras. In the reviewed version of the manuscript, we have modified the text in all the paragraphs were presenting publications versus year of sampling to make clear this point and avoid any confusion in this regard.

L506-520: There is no consistency in the amount and type of information provided for each process study. For example, why the objective of the EqPac study is described but not for the other studies, for which mainly the dates and locations are provided?

Information provided for each process study has been modified (shortened or extended depending on the case) for the sake of consistency process studies.

L523: "initiatives" should be singular (only OMEX included here).

Done.

L525: "carried out" instead of "made"

Done.

L532-536: "Time-series.....Murray, (1992))". Cut the sentence, it's too long.

Done.

L535-536: 10 studies used sediment trap and also <sup>210</sup>Pb-<sup>210</sup>Po at the same time (same studies for both)? If not, rephrase for clarity

Rephrased.

L536: Same comment as L145: 210 should be a superscript.

This issue has been corrected throughout the manuscript

L538: Same comment as L340

Same response as before. We appreciate the reviewers' comment on this regard. Changes have been implemented, including the duration of each specific era and a description for the main theme that characterized it. In the reviewed version of the manuscript, we have kept name of the eras after titles from seminal ocean-related books but included a quote from each one of them to evince each one of the choices made for the era, which follow an intention are not random.

L543-544 and L551-552: Same sentences. Delete one to avoid repetition or change them to say different things.

Deleted.

L556-557: Missing the degree (°) symbol for the coordinates of the PAP station

Corrected.

L557-558: Why there is so much detail provided for the PAP station and not for the others? Why it is not explained what sensors the other TSS have? Provide similar levels of detail for all.

The reviewer is right, all TSS should be treated equal. We do not consider describing all the TSS in such a high level of details falls within the scope of the manuscript and there, the detailed information for the PAP site station has been removed in the reviewed version of the manuscript been removed (see line 618).

L560-564: I'm not sure the level of detail provided here is necessary. Simplify.

Done.

L569-571: "An additional improvement... method." This is out of place, this should be moved and discussed in the following era since it was published in 2021.

This text has been moved to era 4 (see lines 716-719).

L573: I guess "contraptions" is supposed to be "concentrations"

Corrected.

L573: Delete "and" after "POC:<sup>234</sup>Th"

Corrected.

L578: What do you mean by "surveys"? Cruises, programs? It can't be studies because those 4 "surveys" are differentiated from the 87 studies. Please clarify

We meant cruises. This part of the text has been deleted in the reviewed version of the manuscript for the sake of clarity.

L580: Are HOT, BATS and DYFAMED considered programs? In table 2 they are referred as TSS. The program that initiated these time series was JGOFS. Maybe refer to them as TSS?

Done.

L584: Missing the degree (°) symbol for the coordinates of the K stations

Corrected.

L586: Correct "JGOFS"

Done.

L587: U.S. JGOFS-Japan refers to a collaborative station between US and Japan?

U.S. shouldn't be there and has been erased.

L587: Missing the degree (°) symbol for the coordinates of the KNOT station

Corrected.

L590: Rephrase: “And a pilot cruise was reported under the GEOTRACES program” or similar

Rephrased.

L590: Correct “...discussed in the next section”

Corrected.

L592: “Finally, a great number of projects and experiments belong to this era (see Table S3 for a chronological summary).” Remove, starting the sentence with “finally” creates great expectations for the reader, but there nothing after that.

Done.

L593: “Sampling”

Done.

L601: “while and” is incorrect. Please choose which one to use.

Done.

L608: Change “used” for “use”

Done.

L610: 210 should be a superscript

Corrected.

L612: Same comment as for L340

Same response as before. We appreciate the reviewers’ comment on this regard. Changes have been implemented, including the duration of each specific era and a description for the main theme that characterized it. In the reviewed version of the manuscript, we have kept name of the eras after titles from seminal ocean-related books but included a quote from each one of them to evince each one of the choices made for the era, which follow an intention are not random.

L619: It should be “GEOSECS”

Corrected.

L624: “carried out two years later”. Later from which reference year? Just say the year that it took place.

Done.

L626: Correct “cruises”

Corrected.

L629-632: Maybe some of the studies could be cited, including Clevenger et al., (2021) cited in the previous era, or the GEOTRACES intermediate data products from 2014 and 2017 (Mawji et al., (2015) and Schlitzer et al. (2018)) or the trace metal fluxes studies (Black et al., 2018; maybe add some more studies here since you refer to a shift from POC to trace metal fluxes).

New references have been added to this part of the text (see lines 696).

L638: “and known” should be “are known”

Corrected.

L643-645: The 8 cruises that sampled in the NH and the SH are included in the 27 that sampled in the SH or are considered a separate group. If so, maybe mention them apart from the NH cruises.

The text has been rephrased to clarify this issue (see lines 724-727).

L646: Refer to GEOTRACES instead of “a major ocean program”.

GEOTRACES is not the only major program under which expeditions took place within era 4 (as shown in Table 2 to which we refer the reader to after our statement). Therefore, this change suggested by the reviewer has not been implemented. Nonetheless, the text has been corrected to avoid confusion.

L676: Add “and” in between “2<sup>nd</sup> 3<sup>rd</sup>”

Done (see line 730).

L646-651: Why are this projects/experiments/cruises not cited in Table 2? What criteria was used to include them or not in that table?

The compilation of program included in Table 2 represent a selection of selected programs, chosen in terms of number of cruises achieved, duration of the program, relevance of results throughout time, etc. And overall, a global representation of programs from all countries have been followed. HilATS have been deleted from the Table and from the “selected ocean program” in the reviewed version of the manuscript.

L659: Change to “...whether a SS, a NSS or both models were used”

Done.

L660-661: Are those citations referring to what? The use of SS, NSS or both? Why so many citations? Just write “e.g.,” and cite max 3.

References now added for each approach including a maximum of 3 (see lines 743-753).

L665 and L666: Same comment as L145: 210 should be a superscript.

Done.

L666: Correct “compared them”

Corrected.

L668: Same comment as L340



Same response as before. We appreciate the reviewers' comment on this regard. Changes have been implemented, including the duration of each specific era and a description for the main theme that characterized it. In the reviewed version of the manuscript, we have kept name of the eras after titles from seminal ocean-related books but included a quote from each one of them to evince each one of the choices made for the era, which follow an intention are not random.

L675: the abbreviation for net primary production is not needed since it is not used anymore

Abbreviation removed.

L677-678: "availability for phytoplankton at a key junction of the global ocean circulation". Rephrase for clarity

Rephrased.

L684: BCP has not been defined yet. It is defined in L697. Abbreviation probably not needed, used a few more times later on but there is a large number of abbreviations in this manuscript already.

We agree with the reviewer that there are many abbreviations in the manuscript, but we consider the target audience of this manuscript are well familiarised with the BCP abbreviation for the Biological Carbon Pump and have decided to keep it. Nonetheless, abbreviation has been defined when it corresponds in the first use of the concept (see line 30), not where it was in the previous version of the manuscript.

L685-686: Why APERO and PICCOLO are not included in the previous group of projects? What is the criteria to differentiate them from COMICS, EXPORTS, CUSTARD, etc.

APERO is in a list apart because it is a planned initiative to be started in future years, as indicated in the manuscript. This difference has been stressed out in the reviewed version of the manuscript to avoid confusion. PICCOLO was misplaced in the previous version of the manuscript and has been moved to the list with the rest of projects, as corresponds to an initiative already started.

L689-691: Please clarify what JETZON does. "...international coordinating umbrella to serve as a focal point..." means that they organize or fund cruises, or meetings? Are any of the recent programs cited before (COMICS, EXPORTS, CUSTARD, OTZ, SOLACE, APERO, PICCOLO) included or linked to JETZON in a formal way?

JETZON is a UN Ocean Decade Programme, acting as an international coordinator and focal point for Twilight Zone studies. International projects such as COMICS, EXPORTS, CUSTARD, OTZ, SOLACE, APERO, PICCOLO, etc. are linked to JETZON, that acts a meeting point to share data, results or discussions and organize seminars, among other things. The link is robust although it is not formal. JETZON description is improved in the manuscript (see lines 781-785).

L697: See my comment for L684

We consider the target audience of this manuscript are well familiarised with the BCP abbreviation for the Biological Carbon Pump and have decided to keep it. Nonetheless, abbreviation has been defined when it corresponds in the first use of the concept (see line 30), not where it was in the previous version of the manuscript.

L697: Delete "of"

Deleted.

L700: “time-series site” should be TSS

Corrected.

L707: Use “< 1%” instead of “less than 1%” (if not add a space after “than”)

Space added (see line 805).

L718: Correct “for the application”

Corrected.

L726: Change “...since most cruises do not allow repeated measurements...”

Corrected.

L727: Delete “i.e., neglecting changes...”. It has already been explained what a SS model implies.

Corrected.

L718-731: This paragraph seems repetitive with previous sections of the manuscript, when presenting SS and NSS models. This section should describe how many studies applied SS and NSS or both and in which cases the application of one model or the other had a significant impact. Maybe citing the work by Resplandy et al. (2012). <http://dx.doi.org/10.1016/j.dsr.2012.05.015> can help to provide the overview on this particular gap.

We agree with the review that the approach given to this entire section in the previous version of the manuscript was not the most right one. Accordingly, Section 5 (affecting these paragraphs referred to by the reviewer) has been completely rewritten in the reviewed version of the manuscript to address this and some other comments by both this reviewer and reviewer 3 (see new section 5, lines 791-866).

L732-733: Too broad. Not all the radionuclides present deficits in the water column and not just because of their half-lives, also because of their physicochemical behavior. Please rephrase referring only to <sup>234</sup>Th, since it is the radionuclide of interest for this manuscript.

Done.

L736: Add “e.g.” in front of the citation to Ceballos-Romero et al. (2018), there are other studies that have reported similar results.

Added (see lines 817).

L736-744: This paragraph is not referring to a gap in the dataset. This is an explanation about the importance of the bloom period and the reliability of the SS and NSS approaches (issues that the readers accessing this dataset are most probably already aware of). I don't think it is needed, plus it seems a bit repetitive and too extensive considering that an explanation about SS and NSS already started in L718. Please simplify from L718 to L744 in order to highlight the importance of this considerations and their implications in a much briefly way.

We agree with the review that the approach given to this entire section in the previous version of the manuscript was not the most right one. Accordingly, Section 5 (affecting these paragraphs referred to by the reviewer) has been completely rewritten in the reviewed version of the manuscript to address this and some other comments by both this reviewer and reviewer 3 (see new section 5, lines 791-866).

L745-751 are great, they report the actual data and highlighting the importance of reporting bloom conditions.

Section 6 has been fully re-written following the spirit of this paragraph mentioned by the reviewer.

L755: Correct “occurs”

Corrected.

L758: Delete “sampled” since it has been stated several times that all the oceans were sampled.

Deleted.

L760-766: Again, this is not a discussion about a gap in the dataset, it is explaining again what data is reported, how important it is to combine  $^{234}\text{Th}$  with other tracers or devices to quantify export, etc. This can be deleted since it has already been presented and discussed along the manuscript.

This paragraph has been deleted in the new version of the manuscript.

L761: Correct “provides”, since it is the “combined interpretation” that provides us with the tools.

Corrected.

L763-764: The “Ceballos-Romero et al. 2016” citation is not needed or add other studies, this is not the only one that has combined Th-234 with other tools.

Citation removed.

L764-765: “For that reason..... might be missing.” These two sentences are important, but they should be moved to the section describing the dataset, section 2. Data.

The text has been moved to section 2 (see lines 146-151).

L777: Change “us” by “as”

Changed.

L778: Please rephrase “Together with particle sinking velocities disequilibrium derived”, I do not follow it.

The part of the text affecting this sentence has been entire rewritten (see section 6 of the reviewed version of the manuscript, lines 867-915).

L781: It should be BCP

Corrected.

L783: all the ways to re-evaluate the  $^{234}\text{Th}$ -derived POC fluxes can be done using the dataset of this compilation, but it does not allow to evaluate those fluxes in relation to the particle sinking velocity or the synthesizing modelled particle remineralization, so please rephrase to clarify what can be done with it alone and what could be done if combined with other datasets.

This sentence has been clarified as requested by the reviewer. In fact, the entire section 6 has been rewritten with a new approach that address comment about the lack of actual data in the manuscript from this reviewer.

**Table 1:**

Please check sampling periods. In several instances the dates are placed reversed i.e., first the end of the sampling followed by the starting date. See: Matsumoto, 1975; Minagawa and Tsunogai 1980; Wei and Murray, 1991; Porcelli et al. 2001 (different format used: yyyy/mm/dd); Lepore et al. 2009

All the sampling periods have been checked and corrected when found an issue (done for a total of 5 studies). (see table 2).

This problem is also found in the dataset available in PANGAEA. Please correct it there too. I noticed these ones but please review them all again.

We thank the reviewer for checking this issue in PANGAEA. All the spreadsheets underwent several rounds of quality control, both internal and external, but some formatting issues like the one pointed out by the reviewer were undetected. All the spreadsheets in PANGAEA have been reviewed and corrected. This mistake was detected in a total of 5 studies of the 223 compiled. These studies are the ones indicated by the reviewer plus Speicher et al., 2009 and Lepore et al., 2009.

Also, some of the studies have only one sampling date, therefore, that term could be simplified by reporting just that date instead of reporting it twice. See: Moran and Buesseler, 1992; Buesseler et al. 2000; Cai et al 2002; Coppola et al 2002; Guo et al 2002; Hung and Gong 2007; Buesseler et al 2010 and Pabortsava, 2014

Dates for the studies indicated by the reviewer (and some others with similar start and end dates of sampling) have been simplified as suggested (see table 2).

It would be good to increase the width of the “Reference” column because some of them take more than one row. No data repository is reported in that column, so the title of that column can be simplified.

We appreciate the reviewer’s comment on this issue. We consider this table will benefit from being depicted in a page with landscape orientation. However, the versions of the manuscript submitted for reviewer depicted the tables in the best way to be read following the format request by the journal (i.e., portrait orientation). We will take into consideration the reviewer’s suggestion regarding formatting issues during the proof-reading phase with the journal.

Nonetheless, some changes have been made in the Table in the reviewed version of the manuscript to ease its reading.

**Table 2:**

As mentioned in my detailed comments, I would like to understand the criteria used to include programs or in this table or not, since some mentioned in the manuscript are not included. On the other hand, OSP- Station PAPA Ocean Weather Station “P” is not mentioned in the manuscript but is included in the table.

The reviewer is right, we missed reporting OSP- Station PAPA Ocean Weather Station “P” in the manuscript. It has been included in the reviewed version as a milestone of era 4 (see lines 420-423).

Please format the table so that the “Acronym” column as well as the “Activity” column are wide enough to have the entire name in one row. Right now, it is too big and it makes it hard to follow.

Same response as for the comment on the formatting of table 1. We consider this table will benefit from being depicted in a page with landscape orientation. We will take into consideration the reviewer’s suggestion regarding formatting issues during the proof-reading phase with the journal.

Nonetheless, some changes have been made in the Tale in the reviewed version of the manuscript to ease its reading.

**Figure 1:**

Panel c) LTTS has not been used anywhere in the manuscript, the acronym used is TSS. If for some reason you prefer to use long-term, high-frequency time-series stations, do not use the acronym.

Corrected (see Figure 1).

Same for L831, TTS station is the wrong acronym.

Corrected (see Figure 1).

Panels a) and c) have already a lot of symbols and colours reporting data. I don't see the necessity to provide the topography data. A scale of greys for the background might be less shocking and allow to better visualize the data reported in these panels.

We agree with the reviewer that maps contained too much information in the previous version of the manuscript. Accordingly, all maps have been redone in the reviewed version of the manuscript to ease their interpretation.

**Figure 2:**

L842: vi) remove the “” from the category Others in the caption.

Corrected (see Figure 2).

For panel b) the lines for 2001 and 2010 are very out of place. Please add the legend in this graph again if this panel is placed right below panel a) for publication.

This issue with the dotted lines has been corrected in all the plots. Legend has been added for panel b).

For panels a) and c) the dotted lines indicating the tipping points are also not centred in relation to the specific year.

This issue with the dotted lines has been corrected in all the plots.

**Figure 3:**

L873: Delete one of the “by the”

Corrected (see Figure 3).

Same as per Figure 1, I think a grey background might allow to better visualize the datapoint reported in the different panels.

All maps have been redone in the reviewed version of the manuscript to ease their interpretation.

The caption does not match the symbols/colours of the map:

Panel a) ii)  $^{238}\text{U}$  + total  $^{234}\text{Th}$  the caption should be dark blue squares; iii) no  $^{238}\text{U}$  the caption should be magenta dots

Corrected (see Figure 3).

Panels b, c and d) iii) the caption should be no total  $^{234}\text{Th}$  sampled (magenta dots); iv) the caption should be no POC:  $^{234}\text{Th}$  ratios (light blue triangles)

Corrected (see Figure 3).

Notice that the legend for panels b), c) and d) the Th in the light blue triangles shouldn't be a superscript.

Corrected (see Figure 2).