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 file 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 ²³⁴Th 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 ²³⁴Th 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 #3

This study presents a compilation of oceanic ²³⁴Th measurements made at global scale over the past 50 years (1967-2018). The dataset is composed of several parameters including total, dissolved, and particulate ²³⁴Th activity, POC:²³⁴Th and PON:²³⁴Th ratios, along with ²³⁸U activity, POC and PON concentrations when available. This set of parameters constitutes the basis for the use of ²³⁴Th as a proxy of carbon and nitrogen export fluxes from the oceanic upper water column. The data were obtained from several sources, the vast majority from peer-reviewed articles (214) and to a lesser extent from PhD manuscripts (4), public data repositories (8), and unpublished datasets (9). The database is composed of 223 excel spreadsheets along with a compilation file hosted in PANGAEA repository. For each data set, relevant metadata (geographic location, sampling date, project, sampling and processing methodology, bloom stage, etc.) have been systematically included. The associated paper introduces the dataset, presents a global overview, and then discusses the timeline of ²³⁴Th measurements according to four periods covering the last 50 years. Finally, the authors discuss gaps in the dataset and present some perspectives on its future uses.

General comments

First, I would like to acknowledge the extensive work that has gone into this very comprehensive and clear compilation of over 50 years of research on the short-lived radionuclide ²³⁴Th. Such a compilation was lacking until now and represents a new important step in the use of ²³⁴Th as a proxy for the export of C and other elements (N, BSi, CaCO₃, and trace elements) from the upper water column. The database is well structured and clearly described with detailed metadata of significant importance. It also appears very exhaustive, and I could only identify a few minor omissions or errors (see detailed comments below).

We appreciate the comments of the reviewer. We specially appreciate the very thorough and elaborated revision. We strongly believe its meticulous remarks have greatly improved the text and the whole compilation

My first general concern is with the form, as the manuscript contains a significant number of typographical and editorial errors that would have benefited from careful review before its submission. This concerns in particular the list of bibliographic references which contains a significant number of errors (authors list, authors name, type of reference such PhD manuscript, book chapter, research article). Still on the form, there is a surprising confusion between concentration and activity made throughout the manuscript. The ²³⁴Th and ²³⁸U data you report are activities not concentrations.

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.

As for the activities versus concentrations issue, we agree with the reviewer that these terms were used indistinctly in the previous version of the manuscript which generated confusion.

What we report in the compilation for total, dissolved and particulate ²³⁴Th and for ²³⁸U are "activity concentrations" as corresponds to activities per unit volume (measured in units of dpm L⁻¹), as defined by ISO 921 of this 4 of (chrome-(see comment on page the IAEA glossarv extension://efaidnbmnnnibpcajpcglclefindmkaj/viewer.html?pdfurl=https%3A%2F%2Fwwwns.iaea.org%2Fdownloads%2Fstandards%2Fglossary%2Fiaea-safety-glossary-draft-

2016.pdf&clen=1708013&chunk=true). For the sake of simplicity, we have referred to these data as simply "concentration". as acknowledged in the reviewed version of the manuscript (see lines 85-86).

On the content, I was pleased to read the timeline of the Th studies which summarizes the major stages that contributed to the development of the method. For clarity to the reader, I would recommend to indicate for each period the corresponding years both in the manuscript in the subsection headings and in the figures.

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.

This information has been added to the text, tables, and figures.

About the different eras, I might have subdivided era 1 and 2 a little differently by considering the JGOFS program as the beginning of era 2. In fact, it seems more logical to me to take into account the sampling periods corresponding to different programs rather than the date of publication of the resulting studies. This is illustrated further in Table 2, where most of the studies belonging to era 2 were performed in the framework of JGOFS.

We agree with the referee that the arrangement of the eras could be logical considering the sampling, not the publications. However, to do a correct and consistent analysis of the data we need to be extremely coherent with the dates associated to the datasets, so in that case, we should use that classification for the whole dataset. And the problem of a classification by sampling is that it cannot be applied to many of the publications analyzed, as they include several samplings, and it is difficult to designate a single sampling date. Furthermore, other publications present annual results of a single station, thus including many years. In those cases, we can only use the date of publication. And for consistency, publication date is applied to all the analysis of the eras, otherwise, consistent statistics could not be obtained.

On the other hand, the eras are chosen by a differential univocal event that can be considered the kick-off of the era. The reviewers were right that the criteria to choose this univocal event was not clear in the previous manuscript. Therefore, we have decided to define four relevant papers for each era as kick-off event. Those papers are the following: First published paper detection ²³⁴Th disequilibrium in the ocean (Bhat et al, 1969), first paper where POC export is quantified from 234Th disequilibrium (Buesseler et al., 1992), publication of the first small volume technique for ²³⁴Th measurement (Buesseler et al., (2001) and Benitez-Nelson et al., 2001) and finally first ²³⁴Th paper published within the GEOTRACES program (Cai et al., 2010).

This way, the eras, and the analysis of their data, are finally distributed as follows:

- 1969-1991: The beginning of ²³⁴Th as tracer of POC export.
- 1992-2000: Introduction of the empirical determination of POC export from ²³⁴Th profiles.
- 2001-2006: Consolidation of the ²³⁴Th technique.
- 2010-present: GEOTRACES program and a new way to study the ocean.

Therefore, the eras are arranged as in the previous version of the manuscript, but we believe are reason for the criteria chosen are well justified in the reviewed version of the manuscript.

References:

Bhat, S. G. G., Krishnaswamy, S., Lal, D., Rama, & Moore, W. S. S. (1969). 234Th/238U ratios in the ocean. Earth and Planetary Science Letters, 5, 483–491. <u>https://doi.org/10.1016/S0012-821X(68)80083-4</u>.

Buesseler, K. O., Bacon, M. P., Kirk Cochran, J., & Livingston, H. D. (1992). Carbon and nitrogen export during the JGOFS North Atlantic Bloom experiment estimated from 234Th: 238U disequilibria. Deep Sea Research Part A. Oceanographic Research Papers, 39(7–8), 1115–1137. <u>https://doi.org/10.1016/0198-0149(92)90060-7</u>.

Buesseler, K. O., Benitez-Nelson, C., Rutgers van der Loeff, M., Andrews, J., Ball, L., Crossin, G., & Charette, M. A. (2001). An intercomparison of small- and large-volume techniques for thorium-234 in seawater. Marine Chemistry, 74(1), 15–28. https://doi.org/10.1016/S0304-4203(00)00092-X.

Benitez-Nelson, C., Buesseler, K. O., van der Loeff, M. R., J. Andrews, L. B., Crossin, G., Charette, M. A., Loeff, M. R. van der, J. Andrews, L. B., Crossin, G., & Charette, M. A. (2001). Testing a new small-volume technique for determining 234Th in seawater. - Journal of Radioanalytical and Nuclear Chemistry, 248(3), 795-799. https://doi.org/- 10.1023/a:1010621618652.

Buesseler, K. O., Bacon, M. P., Kirk Cochran, J., & Livingston, H. D. (1992). Carbon and nitrogen export during the JGOFS North Atlantic Bloom experiment estimated from 234Th: 238U disequilibria. Deep Sea Research Part A. Oceanographic Research Papers, 39(7–8), 1115–1137. <u>https://doi.org/10.1016/0198-0149(92)90060-7</u>.

Regarding the first era, you may think to introduce the GEOSECS program earlier than L365 after the description of the Coale and Bruland papers. I think it could be relevant to mention that it was in the framework of GEOSECS that the concept of scavenging by particles has really emerged, especially for the open ocean. You may cite the seminal work of Turekian (1977) who introduced the concept of the "great particle conspiracy". Furthermore, it might be interesting to mention that during GEOSECS little attention

was given to ²³⁴Th in comparison to ²²⁸Th and ²³⁰Th for studying scavenging processes in the open ocean (Broecker and Peng, 1982). It was only later, with the papers of Coale and Bruland, that the role of ²³⁴Th as a tracer of short-term particle dynamics was really highlighted.

We appreciate the review's comment regarding GEOSECS, but we consider there is no need to mention it earlier than done, when starting to discuss ²³⁴Th timeline in era 1 (i.e., section 4.1).

The discussion suggested by the reviewer regarding particle scavenging and the great particles conspiracy has been included in the text, as suggested by the reviewer (see lines 364-369).

Regarding era 4 related to the GEOTRACES program, I would set the starting year to 2008 or even 2007, which corresponds to the International Polar Year (2008) and the start of the GEOTRACES sampling program.

Wee agree with the reviewer here regarding the starting date that should be associated to the GEATRACES program, corresponding to the IPY. However, as explained above, this is not relevant anymore since, in order to follow a strict, straightforward criterion to designate the eras, we have chosen influential ²³⁴Th-related papers as starting point to frame the eras. In this case we have chosen Cai et al. (2010) (<u>https://doi.org/10.1029/2009JC005595</u>) as the first Th-234 results published in the framework of the GEOTRACES program.

Regarding this program, it would be relevant to include in table 2 all cruises that have been sampled for ²³⁴Th so far.

Table 2 include the description of the programs, but the cruises are not included in any of the different programs. Therefore, we find that it will not be consistent to include GEOTRACES cruises and not for the others. But in those cases, if all the cruises were included for all the programs, Table 2 would be immensurable. We hope the reviewer understand our concern about including the cruises here.

All programs under which sampling have taken placed are indicated in the summary table available in PANGAEA (see <u>https://doi.pangaea.de/10.1594/PANGAEA.918125?format=html#download</u>).

For consistency, I would recommend using the name of the section or process study considered as defined by GEOTRACES, you can then indicate which country has been involved in the sampling (US/UK/Netherland/Germany/India/France). This remark is also valid for the Th_database file, the projects performed within the framework of GEOTRACES should be named in a more consistent way such as for instance GEOTRACES (section or process study number, country, and eventually project acronym).

This information is included in each of the individual data files and in the text (see for example lines 290-296).

Still on the GEOTRACES program, I think it is important to mention that not all sections analyzed for ²³⁴Th are available in the 2014 and 2017 Intermediate Data Products (L367). Even if data obtained as part of GEOTRACES are published in peer-reviewed journals, their inclusion in the IDP requires some additional steps (submission and acceptance by the GDAC). Also, you may indicate that the last IDP (2021) was released very recently.

This remark is now included in the text. IDP2021 has been added to the manuscript (see line 563).

I have also some concern regarding the section 5. Significant gaps in the global dataset. It is not clear to me what the gaps you want to discuss are. Reading the first paragraph (L696-701), it seems the gaps you want to consider are related to the current understanding of the Biological Carbon Pump. On this topic, I would recommend to include some more recent reviews (Henson et al., 2019; Boyd et al., 2019; Siegel et al., 2016), which detail some of the processes that require further consideration. Reading the following, I notice you discuss two main points, the first one is related to the spatio-temporal distribution of 234Th data and the second one, too long considered from L711 until the end of the section L768, is related to the modeling

approach (SS vs NSS) used for estimating the export fluxes of ²³⁴Th. It is surprising to note that this entire section is mostly discussed using only two references and written by the first author of this review. In my view, this section needs to be reconsidered, first by giving more attention to the existing literature on the SS/NSS approach and the different oceanic contexts to which it has been applied (not only the North Atlantic), and second to the other numerous issues related to the ²³⁴Th method. Among these, it is important to point out the role of physics (lateral and vertical advection and diffusion) (Buesseler et al., 1995; Savoye et al., 2006; Resplandy, 2012; Le Gland et al., 2019; Roca-Marti et al., 2017), the importance of the depth of integration (Buesseler et al, 2020), and finally all the other issues related to the conversion to carbon fluxes using the POC to Th ratio of sinking particles (the choice of the relevant size fraction, the interpolation methods, etc.). By following these guidelines and considering that export fluxes have not been calculated or compiled in this review, you may be able to give recommendations to future users of this database.

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 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).

Finally, and still about the gaps in this dataset, there is one point that could be considered that concerns the quality of available data. I understand that it is difficult to answer this point but if we take into account the successive evolutions of the methods used for the determination of ²³⁴Th, all the data are probably not of the same quality. I think this should be at least mentioned or even taken into account in the form of a quality flag assigned to each dataset.

See specific comment at the beginning of the response to reviewers. To take into account in the discussion of the dataset that there are certain parameters that are missing in the compilation we have modified Section 5. As discussed above, we agree with the reviewer that this section was not well focused, and we have rewritten the whole section accordingly. Section 5 now includes a subsection related to the parameters that are missed in the compilation, including the necessity of a flag to estimate the quality of the data.

For the particular case of the reviewer's comment, we believe that addressing the quality of the data compiled is out of the scope of the compilation, at this stage. It would imply ruling out quality of the data provided by the researchers without a reaching a previous consensus with the authors.

We acknowledge however, that the point made by the reviewer is an important one to be considered, and it would improve the quality of the compilation. For that reason, for next editions of the compilation we will consider including a Flag to the new data included. That flag will be added after agreement with the authors whose data are included the data set.

Detailed comments

L15: the 234Th-238U pair is primarily used for assessing export fluxes, to look at export efficiency you need to compare with the net primary production, which is actually not included in the dataset. Please clarify

The reviewer is right, we do not provide the primary production, parameter required to evaluate the export efficiency and therefore, the sentence pointed out by the reviewer is confusing. In the reviewer version of the manuscript, we have rephrased this part of the text (see abstract).

L20: replace concentrations by activities and at all other occurrences in the manuscript for both 234Th and 238U.

Done throughout the entire manuscript.

L29: the term uptake is a bit ambiguous and not directly related to the 234Th method and to export fluxes. Uptake can be used either for air-sea exchanges or biological assimilation, please clarify.

L35: correct the reference (Cochran and Masqué, 2003)

Corrected (see line 32).

L45: remove "a" in front of 234Th

Corrected.

L50: mean life time

Corrected (see line 47).

L52: check the end of sentence "high scavenging"

Checked and corrected.

L55: correct reference for Cochran, 1993 (Kirch is the first name)

Corrected (although note this reference has been erased from this part of the manuscript in the reviewed version).

L57-60: you may cite also the works of Lemaitre et al. (2016) for N, BSi, Fe export in the Kerguelen area (KEOPS2 project) and Lemaitre et al. (2020) for BSi, CaCO3, lithogenic material and trace elements along the GA01 section (GEOVIDE) in the North Atlantic.

This reference has been cited in the text (see line 54), and also included in the Excel files. We thank the reviewer for this reference since we were not aware of this data set.

L63-64: remove the editors list of the special issue

Removed.

L88: add Th after 234

234 removed, that was the mistake.

L104: 238U activity

Corrected.

L105: you mean compiled and not "complied"

Corrected.

L114-115: as mentioned earlier, data obtained in the frame of GEOTRACES are not necessarily in the IDP

This sentence has been modified to take this into account (see lines 103-105).

L124: not necessary to define supplementary material with an acronym (figure or table number is sufficient).

Acronym removed.

L129: could you provide more details on how the bloom stages were distinguished?

Done (see lines 119-129).

L145: please clarify what you mean with CHN data and how this differ from POC and PON concentrations reported in the dataset (as mentioned L104). Furthermore, CHN is not the only analytical method that can be used to determine POC and PON, EA-IRMS is also widely used for this purpose.

We have used the CHN label to indicate studies that provide POC and/or PON concentrations (in μ mol L⁻¹). This has been clarified din the reviewed version of the manuscript. Other analytical methods that exit to measure CHN data in addition to an elemental analyzer have also been mentioned, as suggested by the reviewer (see lines 89-92).

L151: Check sentence "Where data..."

Checked and corrected.

L168: remove concentration for 234Th

Removed.

L169: correct units µmol dpm⁻¹ for POC and PON to 234Th ratios

Corrected.

L172: correct >53 μ m and not <53 μ m

Corrected.

L173: was responsible instead of is

Corrected.

L178: check units µmol L⁻¹

Checked and corrected.

L181: correct one sigma

Corrected (see line 190).

L190: remove the _ after authors

Removed.

L193: check sentence and correct "used for"?

Removed.

L198: the previous compilation by Le Moigne et al. (2013) include 234Th fluxes, POC:Th ratios, POC fluxes and NPP estimates.

We have included this information in the reviewed version of the manuscript (see lines 205-208).

L200: correct aspects

Removed.

L201-202: check the end of the sentence

This text has been rephrased.

L205: remove SM

Removed.

L208: correct larger

Corrected.

L214: put the acronym in brackets

Done.

L222: correct "on at least.."

Corrected.

L235: repeated occupation?

Corrected.

L242: Correct "the way ocean is currently..."

Corrected.

L275: you may include the KEOPS project (sampling in 2005) with the study of Savoye et al. (2008) and also the great Calcite Belt expedition with the study of Rosengard et al. (2015) carried out both in the Atlantic and Indian sectors of the SO.

We thank the reviewer for the suggestion; however, we have only identified a total of 210 and 399 ²³⁴Th data points for the studies of Savoye et al., (2008) and Rosengard et al., (2015) in comparison to the e.g., 1610 and 1040 data points reported by Owens et al., (2015) and Buesseler et al., (2020) sampled in 2010 and 2018 respectively and referred to in this part of the text. Is for this reason that we do not consider these 2 studies suggested by the reviewer make a significant peak in the annual distribution of ²³⁴Th data points relative to the years selected, this is i) 1992 ii) 1997 iii) 2004, iv) 2010 and v) 2018), and we have decided not to include these 2 studies in the reviewed version of the manuscript. References:

Owens, S. A., Pike, S., & Buesseler, K. O. (2015). Thorium-234 as a tracer of particle dynamics and upper ocean export in the Atlantic Ocean. *Deep Sea Research Part II: Topical Studies in Oceanography*, *116*(0), 42–59. https://doi.org/http://dx.doi.org/10.1016/j.dsr2.2014.11.010

Buesseler, K. O., Benitez-Nelson, C. R., Roca-Martí, M., Wyatt, A. M., Resplandy, L., Clevenger, S. J., Drysdale, J. A., Estapa, M. L., Pike, S., & Umhau, B. P. (2020). High-resolution spatial and temporal

measurements of particulate organic carbon flux using thorium-234 in the northeast Pacific Ocean during the EXport Processes in the Ocean from RemoTe Sensing field campaign. *Elementa: Science of the Anthropocene*, 8(1). https://doi.org/10.1525/elementa.030

L276: remove the # and correct "in terms"

Removed and corrected.

L283: correct "field"

Corrected.

L284-285: what do you mean with "chemical scavenging"

Corrected to simply "scavenging".

L290: correct "through years"

Corrected.

L292: check the beginning of the sentence

Checked and corrected.

L305: correct "were"

Corrected.

L329-333: the end of the paragraph is not related to 238U measurements but to additional metadata information (model assumption and bloom stages)

The end of this paragraph has been removed as included repeated information.

L357-358: check sentence

Checked and corrected.

L364-366: I would consider the GEOSECS program earlier (see my general comment)

We appreciate the reviewer's comment on this regard, but as previously mentioned, we consider there is no need to mention the GEOSECS program (or any other program) earlier than in Section 4, where we discuss the timeline of the ²³⁴Th technique, to which these and other programs are relevant.

L387: Clarify the beginning of the sentence

Clarified.

L397: correct "do not"

Corrected.

L404: remove concentration with activity

Not removed, see reply to general comment regarding the use of activity versus concentration in the manuscript.

L409: Nucleopore

This sentence has been rephrased and "Nucleopore" does not appear anymore.

L420-423: I would also mention that physical terms were not considered for solving the 234Th activity balance.

The mention to the physical term has been included (see lines 500-503).

L423: remove of or rephrase

The entire paragraph has been rephrased (see line 501-505).

L433: Clarify what kind of models were used (one or two boxes) and the corresponding fluxes (scavenging fluxes J and export fluxes P). Also check the end of the sentence.

The kind of models used has been specified in the reviewed version of the manuscript (lines 497-509).

L437: JGOFS started in 1987 according to Table 2

After consulting more sources, we have marked the beginning of JGOFS program in 1984 and corrected this date throughout the entire manuscript (see table 2).

L441-442: correct relative to its parent nuclide or 238U

Corrected.

L443: clarify what you mean with "response time" and also to what corresponds the second equation

It was an error, it should say mean residence time, the second equation has been deleted from the text, as it was a very specific one, and it did not fit in the text (line 485).

L455: correct "with time"

Corrected.

L456: here you keep focusing on the SS vs NSS approach. You need to better account for the physical terms (according to Buesseler et al., 1995 in specific ocean settings such as upwelling regions). Furthermore, vertical diffusion can be quantified from a single 234Th profile if the diffusivity coefficient is known or assumed.

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.

L458: check sentence, you may change to "when temporal fluctuations in 234Th activity can be assessed". In addition, and as mentioned by Savoye et al. (2006), NSS approach requires the same water mass to be sampled. This is another difficulty of the NSS approach that can be difficult to meet in dynamical settings.

The whole paragraph has been rewritten including all the suggestions of the reviewer and the discussion on the physical terms (see lines 501-507)

L466: correct "0.001<colloids..."

Corrected (see line 512).

L471: There is a confusion, the 3-D model was not built to estimate the gradients of 234Th activities but to estimate the physical components to the flux (V terms). Without these components, the sinking flux would have been largely underestimated in the equatorial upwelling region.

This is corrected in the text (see lines 517-519).

L482: I don't think the SEATS time-series was operated in the frame of the French JGOFS program, most probably by Taïwan.

The reviewer is right, SEATS time-series station is operated by Taiwan, established din the framework of JGOFS. This fact has been corrected in the reviewed version of the manuscript (see lines 522-523).

L489: Clarify the end of the sentence "filtered"

Clarified.

L494: sediment traps collect sinking particles not bulk.

Corrected.

L505: correct "at the BATS..."

Corrected.

L506: replace "boxes" with areas

Corrected.

L508: check coordinates for the NABE experiment

We've identified no mistake with these coordinates (see line 557).

L518: correct "field work"

Corrected.

L520: Australian

Corrected.

L523: correct "other major initiatives..."

Corrected.

L534: remove "during sampling"

Corrected.

L537: clarify what are the CHN data

Clarified.

L569: the reference Clevenger et al. (2021) is apparently missing in the reference list.

Corrected.

L573: clarify "234Th contraptions"

Corrected.

L626: Correct "Portland (USA, Oregon)" and "Numerous cruises"

Corrected.

L638: correct "are known..."

Corrected.

L664: remove "of the studies"

Corrected.

L666-667: check the sentence

Checked and corrected.

L680: correct "emphasizes"

Corrected (see line 771).

In the section, you may include the MOBYDICK project (Marine Ecosystem Biodiversity and Dynamics of Carbon around Kerguelen: an integrated view, 2018-2022) during which the 234Th method has been implemented.

We thank the reviewer for this useful information which has included in the reviewed version of the manuscript (see lines 776-777).

For the APERO project, it will start in 2022 to 2026 and the cruise is planned for 2023 in the western North Atlantic.

We thank the reviewer for this useful information which has included in the reviewed version of the manuscript (see line 777-780).

L706: correct "gaps"

Corrected.

L722: correct "with time"

Corrected.

L777-778: correct "such as" and clarify the whole sentence

Corrected and sentence clarified.

Table 1. Check start and end date (sometimes inverted). The design needs to be improved as it is very difficult to identify which parameters has been measured for each studies.

Checked and corrected (see table 2).

We appreciate the reviewer's comment on the design of the table issue, and we agree with it. We consider this table will beneficiate 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 Tale in the reviewed version of the manuscript to ease its reading.

I noted a few mistakes for the studies that I know (I didn't check all entries in this table)

The study of Stewart et al. (2007) reports POC:Th ratio from in-situ pumps and sediment traps but not PON:Th ratios (check the crosses)

We thank the reviewer for this comment. In the previous version of the manuscript the information detailed in the caption did not match the information as included in the table. We have checked and corrected the entire table, include the studies highlighted by the reviewer.

Thomalla studies in the Atlantic ocean was published in 2008

In addition to the study in 2008, Thomalla also published additional data in the North Atlantic in her paper of 2006 and her PhD thesis in 2007. This author kindly sent us the PhD dissertation to include data from it in the compilation.

References:

- Thomalla, S., Turnewitsch, R., Lucas, M., & Poulton, A. (2006). Particulate organic carbon export from the North and South Atlantic gyres: The 234Th/238U disequilibrium approach. *Deep Sea Research Part II: Topical Studies in Oceanography*, *53*(14–16), 1629–1648. <u>https://doi.org/10.1016/j.dsr2.2006.05.018</u>
- Sandy Thomalla. (2007). Particulate organic carbon and mineral export from the North and South Atlantic gyres: the 234Th/238U disequilibrium approach. University of Cape Town.

For Jacquet et al. (2011), PON:Th ratio were not provided but POC:Th ratio from sediments traps yes

Corrected and full table checked.

For the Rutgers van der Loeff et al. (2011) study, underway sampling were performed and reported

We thank the reviewer for providing this detail. This underway information has been corrected in the excel file (Reference_USE 0142) and in table 2.

Planchon et al. (2015) study does not report PN:Th ratio but C:Th ratio from sediment traps. Also dissolved Th was plotted in Figure 2. The PN:Th ratios for this cruise (KEOPS2) can be found in Lemaitre et al. (2016).

We thank the reviewer for this comment. Table 2 has been entirely reviewed and corrected when necessary for the information provided by each study. Additionally, PN:234Th ratios from KEOPS2 reported by Lemaitre et al. (2016) have been included in the

Additionally, PN:2341n ratios from KEOPS2 reported by Lemaître et al. (2016) have been included in the excel spreadsheet corresponding to this field expedition (i.e., Planchon et al., 2015, Reference_USE 0129).

For the Lemaitre et al. (2018) study, please correct the reference as follow:

Lemaitre, N., Planchon, F., Planquette, H., Dehairs, F., Fonseca-Batista, D., Roukaerts, A., Deman, F., Tang, Y., Mariez, C., and Sarthou, G.: High variability of particulate organic carbon export along the North Atlantic GEOTRACES section GA01 as deduced from 234Th fluxes, Biogeosciences, 15, 6417–6437, https://doi.org/10.5194/bg-15-6417-2018, 2018.

Corrected and all references checked.

The study of Maïti et al. (2016) reports total Th profiles

Corrected and full table checked.

Table 2.

As for Table 1, the design needs some improvements. Column headings needs to be clarified and further details on the other programs than JGOFS could be included. This could be especially the case for GEOTRACES as well as EXPORTS. Furthermore, I do not see the reason why a given program is considered a major program. For instance, the HiLATS program is indicated only for 2001

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.

Check the start date for DYFAMED

Checked and corrected to 1987, when the first ²³⁴Th sampling was reported by Schmidt et la., (1990).

Note that the compilation published by Coppola et al., (2021) was also consulted to this end, but 2 different dates are stated there (1988 and 1991) and no conclusion could be drawn from it.

References:

- Schmidt, S., Reyss, J. L., Nguyen, H. V., & Buat-Ménard, P. (1990). 234Th cycling in the upper water column of the northwestern Mediterranean Sea. *Global and Planetary Change*, *3*(1–2), 25–33. https://doi.org/10.1016/0921-8181(90)90053-F
- Coppola, L., Diamond Riquier, E., & Carval, T. (2021). Dyfamed observatory data. In *SEANOE*. https://doi.org/https://doi.org/10.17882/43749

Figure 1: use the same acronym as in the text for long-term time series (TTS)

Corrected (see figure 1).

Figure 3: check the legend "by the" written twice

Corrected (see figure 3).

Regarding the dataset, it would be worth checking if the data of Stuckel et al. (2015) and Ducklow et al. (2018) have been included.

We thank the reviewer for these references provided. We confirm that they are not included in the current version of the compilation but will be included in the next version along with other two studies (see below for references) that we have identified as missing during this review process.

- Ma, H., Zeng, Z., He, J., Han, Z., Lin, W., Chen, L., Cheng, J., & Zeng, S. (2014). 234Th-derived particulate organic carbon export in the Prydz Bay, Antarctica. *Journal of Radioanalytical and Nuclear Chemistry*, 299(1), 621–630. https://doi.org/10.1007/s10967-013-2842-y
- Turnewitsch, R., Reyss, J.-L., Nycander, J., Waniek, J. J., & Lampitt, R. S. (2008). Internal tides and sediment dynamics in the deep sea—Evidence from radioactive 234Th/238U disequilibria. *Deep Sea Research Part I: Oceanographic Research Papers*, 55(12), 1727–1747. https://doi.org/10.1016/j.dsr.2008.07.008

References:

Boyd, P.W., Claustre, H., Levy, M., Siegel, D.A., Weber, T., 2019. Multi-faceted particle pumps drive carbon sequestration in the ocean. Nature 568, 327–335. https://doi.org/10.1038/s41586-019-1098-2.

Buesseler, K.O., Andrews, J.A., Hartman, M.C., Belastock, R., Chai, F., 1995. Regional estimates of the export flux of particulate organic carbon derived from thorium-234 during the JGOFS EqPac program. Deep Sea Research Part II: Topical Studies in Oceanography 42, 777–804. https://doi.org/10.1016/0967-0645(95)00043-P.

Buesseler, K.O., Boyd, P.W., Black, E.E., Siegel, D.A., 2020. Metrics that matter for assessing the ocean biological carbon pump. Proc Natl Acad Sci USA 117, 9679. https://doi.org/10.1073/pnas.1918114117.

Ducklow, H.W., Stukel, M.R., Eveleth, R., Doney, S.C., Jickells, T., Schofield, O., Baker, A.R., Brindle, J., Chance, R., Cassar, N., 2018. Spring–summer net community production, new production, particle export and related water column biogeochemical processes in the marginal sea ice zone of the Western Antarctic Peninsula 2012–2014. Philosophical Transactions of the Royal Society A: Mathematical, Physical and Engineering Sciences 376, 20170177. https://doi.org/10.1098/rsta.2017.0177.

Henson, S., Le Moigne, F., Giering, S., 2019. Drivers of Carbon Export Efficiency in the Global Ocean. Global Biogeochemical Cycles 33, 891–903. https://doi.org/10.1029/2018gb006158.

Le Gland, G., Aumont, O., Mémery, L., 2019. An Estimate of Thorium 234 Partition Coefficients Through Global Inverse Modeling. Journal of Geophysical Research: Oceans 124, 3575–3606. https://doi.org/10.1029/2018JC014668.

Lemaitre, N., Planquette, H., Dehairs, F., Planchon, F., Sarthou, G., Gallinari, M., Roig, S., Jeandel, C., Castrillejo, M., 2020. Particulate Trace Element Export in the North Atlantic (GEOTRACES GA01 Transect, GEOVIDE Cruise). ACS Earth Space Chem. 4, 2185–2204. https://doi.org/10.1021/acsearthspacechem.0c00045.

Lemaitre, N., Planquette, H., Dehairs, F., van der Merwe, P., Bowie, A.R., Trull, T.W., Laurenceau-Cornec, E.C., Davies, D., Bollinger, C., Le Goff, M., Grossteffan, E., Planchon, F., 2016. Impact of the natural Fefertilization on the magnitude, stoichiometry and efficiency of particulate biogenic silica, nitrogen and iron export fluxes. Deep Sea Research Part I: Oceanographic Research Papers 117, 11–27. https://doi.org/10.1016/j.dsr.2016.09.002.

Resplandy, L., Martin, A.P., Le Moigne, F., Martin, P., Aquilina, A., Mémery, L., Lévy, M., Sanders, R., 2012. How does dynamical spatial variability impact 234Th-derived estimates of organic export? Deep Sea Research Part I: Oceanographic Research Papers 68, 24–45. https://doi.org/10.1016/j.dsr.2012.05.015.

Roca-Martí, M., Puigcorbé, V., Iversen, M.H., van der Loeff, M.R., Klaas, C., Cheah, W., Bracher, A., Masqué, P., 2017. High particulate organic carbon export during the decline of a vast diatom bloom in the Atlantic sector of the Southern Ocean. Deep Sea Research Part II: Topical Studies in Oceanography 138, 102–115. https://doi.org/10.1016/j.dsr2.2015.12.007.

Savoye, N., Benitez-Nelson, C., Burd, A.B., Cochran, J.K., Charette, M., Buesseler, K.O., Jackson, G.A., Roy-Barman, M., Schmidt, S., Elskens, M., 2006. 234Th sorption and export models in the water column: A review. Marine Chemistry 100, 234–249.

Siegel, D.A., Buesseler, K.O., Behrenfeld, M.J., Benitez-Nelson, C.R., Boss, E., Brzezinski, M.A., Burd, A., Carlson, C.A., D'Asaro, E.A., Doney, S.C., 2016. Prediction of the export and fate of global ocean net primary production: the EXPORTS science plan. Frontiers in Marine Science 3, 22.

Stukel, M.R., Asher, E., Couto, N., Schofield, O., Strebel, S., Tortell, P., Ducklow, H.W., 2015. The imbalance of new and export production in the western Antarctic Peninsula, a potentially "leaky" ecosystem. Global Biogeochemical Cycles 29, 1400–1420. https://doi.org/10.1002/2015GB005211.