

Response to Reviewer 2

The authors greatly appreciate the reviewer for taking the time to this review. We will prepare a revised manuscript by taking into account all comments raised by the reviewer. Below, please find our responses (black) shown after the reviewer comments (blue).

This study explores the processes that control the distribution of ^{231}Pa and ^{230}Th in the oceans and underlying sediments using COCO V4.0, an Ocean General Circulation Model (OGCM), from Hasumi 2006.

They implemented ^{231}Pa and ^{230}Th in the model using offline tracer simulations based on physical fields from COCO. They implemented bottom scavenging as well as a “dependence of scavenging efficiency on particle concentration” in the model.

General comments

- The most puzzling aspect of this manuscript is the lack of use of recent modeling results and the almost total lack of comparison with these model simulations (e.g. Missiaen et al., 2020a and 2020b; van Hulten et al., 2018; Rempfer et al., 2017; Lippold et al., 2012; Luo et al., 2010; Dutay et al., 2009; Roy-Barman, 2009). It is all the more surprising that most of these papers are cited by the authors although mostly as examples of recent publications instead of being analyzed in depth and compared to the COCO model outputs. A more thorough assessment of these new model simulations and how / why they agree / differ from the simulations presented in the manuscript must be done before publication.

The authors are appreciated for the reviewer's comments. In the original manuscript, we intended to provide a closed-form description of our model results, but we agreed with the reviewer that comparisons with the other modeling studies were also important and needed to be included in the revised manuscript. Therefore, we will cite the recent studies presented not only in the Introduction section but also in other sections and compare our results with them in the revised manuscript.

- Similarly, the choice of comparing the COCO model outputs with those of one of the earliest models used for ^{231}Pa and ^{230}Th , namely the model from the Siddall et al., 2005 study, is very disappointing as it misses out all the improvements made by the newer modeling studies and most of the conclusions drawn from these more recent simulations, most of which representing a significant improvement from the Siddall et al. (2005) model. The authors need to carefully and thoroughly justify their choice. Nevertheless, an in-depth discussion to compare their model outputs and conclusions with that of the more recent modeling studies is needed and should not be limited, as it

is the case in the present manuscript, to a comparison with the Siddall et al. (2005) simulation.

As mentioned above, we will compare our results with not only Siddall et al. (2005) but also other recent modeling studies in the revised manuscript. We think that our choice of reference to Siddall et al. (2005) is useful for demonstrating what kinds of new model treatment/parameters are required from this classic model for reproducing the recent observations from the GEOTRACES database. Our improvement comes from (1) incorporation of bottom scavenging, (2) choice of larger partitioning coefficient for ^{230}Th ($K_{ref_230\text{Th}}$), and (3) inclusion of particle concentration effects to $K_{ref_230\text{Th}}$. The first point (i.e., bottom scavenging) was already discussed in the previous study (Rempfer et al., 2017); our model confirmed its importance, and this point itself is not new. However, we showed that the performance of ^{230}Th modeling is not enough to be improved simply by introducing the bottom scavenging and points (2) and (3) are required for its improvement. As the reviewers pointed out, the other recent modeling studies also showed improvement from Siddall et al. (2005) but are not necessarily the same way as the direction of improvement of our model results (e.g., Dutay et al. 2009 and van Hulst et al., 2018 focused on consideration of different particle size). We also emphasize here that our simulation of $^{231}\text{Pa}/^{230}\text{Th}$ is based on the ocean general circulation model (which is not a simplified model such as a 2D model or reduced complexity model). In the revised manuscript, we will discuss our model results by adding a comparison of our results with recent modeling studies.

- In the same vein, there is a great lack of recent literature analysis on ^{231}Pa and ^{230}Th , e.g. the recent review by Costa et al. (2020) or the recent findings of Missiaen et al. (2018) on the effect of the detrital ($^{238}\text{U}/^{232}\text{Th}$) activity ratio on the calculation of ^{231}Pa and ^{230}Th s are neither discussed or cited. A lot of the effects that the authors are discussing in their manuscript is actually discussed in details for ^{230}Th in the review paper by Costa et al. (2020).

Thank you for providing the references. We know that Costa et al. (2020) is a very nice review paper about “ ^{230}Th normalization”. ^{230}Th normalization (which is a tool for reconstruction the sediment flux) is not a topic of our study but we found that this paper also includes some helpful information on ^{230}Th modeling (section 5). We also thank you for providing paper information on Missiaen et al. (2018) about recent finding on the influence of lithogenic and authigenic ^{230}Th on ^{230}Th in sediments. These literatures will be cited in the revised manuscript.

- The literature used to discuss the effect of particles type and distribution is neither the first/pioneering papers on the topics nor the latest. The authors should read the review by Costa et al. (2020) and look at the modeling results of Missiaen et al. (2020b) and references therein. These results should be both mentioned in the state-of-the-art section

of the Introduction and later discussed.

We will describe the influence of the particle field on sedimentary $^{231}\text{Pa}/^{230}\text{Th}$ ratios with appropriate citations of previous studies including Missiaen et al. (2020, QSR) which discussed that changes in the particle can affect $^{231}\text{Pa}/^{230}\text{Th}$ ratios.

- Similarly, the older literature is fundamentally overlooked. The term “boundary scavenging” has been defined and used by Anderson et al. (1983b). Part of what the authors seem to define as a discovery on the effect of particle concentration on scavenging is actually perfectly defined and modeled by Anderson and co-authors in this paper and subsequent papers. This leads to a conceptual problem L356-369 (see also comment on L194 below).

In our understanding, although this paper does not use the term “boundary scavenging” (they used “intensified scavenging” or “near-bottom scavenging”), the concept of “boundary scavenging” was actually introduced in this paper as the reviewer pointed out. We will cite Anderson et al. (1983) “Removal of ^{230}Th and ^{231}Pa at ocean margins” as a pioneering study about boundary scavenging in the revised manuscript.

- Several sentences or model presentation are very vague, e.g. in equation 4a, there is a term “Transport” (L116-120) defined as representing transport by advection, diffusion and convection. These are 3 very distinct physical processes in their formulation, why is the term “Transport” not explicitly given? What does the term “convection” represent in the oceans. There is no bottom heating so I have great troubles understanding what the authors mean here.

We think that the equation 4a is a very standard expression for representing ocean tracer concentration (e.g., this is equivalent to equation 9 of Siddall et al. 2005). As the reviewer pointed out, the transport term includes oceanic advection, diffusion, and convection. The convection term is represented in the model by so-called “convective adjustment” (e.g., Yin and Sarachik, 1994) where unstable stratification leads to very large vertical mixing. The notation of “transport” is also used in previous similar modeling studies (Rempfer et al., 2017; Gu and Liu, 2017; Missiaen et al., 2020, CP).

- I am very puzzled by the use of equation (10) (L169) for both ^{230}Th and ^{231}Pa . The partition coefficient cannot be the same for both radionuclides as they have different behaviors. The value of the exponent used here (-0.42) has been given by Henderson et al. (1999) for ^{230}Th and is indeed not valid for ^{231}Pa . I do not see what can be achieved by using the same reference partition coefficient for both isotopes.

There is a misunderstanding in this reviewer’s comment. We introduced the dependence of particle concentration only for ^{230}Th (not for ^{231}Pa).

- L90: there is one class of settling velocity in the model presented here. There are two classes in van Hulst et al. (2018). Since the authors discuss the effect of the concentration of particles on scavenging, they should discuss the effect of having one vs. more classes of settling speed on their conclusions

van Hulst et al. (2018) introduced multiple size classes of particles, and we understand that specifying the different settling velocity depending on size classes is one of the important aspects in their study. On the other hand, specifying the different settling speeds is unavailable in the framework of our scavenging model and will require a major upgrade of its model formulation. Therefore, its direct evaluation is difficult in our model. However, in the revised manuscript, we will discuss the effect of specifying settling speed, for example, by comparing our results with previous studies introducing this effect such as van Hulst et al. (2018) and Dutay et al. (2009).

- L194: The authors say they included bottom scavenging in benthic nepheloid layers. This is a very important aspect of the model. However, how this is done is not explained. More explanations of this very important aspect are necessary, especially considering the objective of the journal.

We have already described bottom scavenging in the section of experimental design (see L156-162) but will explain more clearly how bottom scavenging was introduced in the revised manuscript.

- Amongst the conclusions, some are included in the equation. The fact that ^{231}Pa is more affected by advection is 1) the basis for using Pa/Th as a proxy for ocean circulation and has already been verified by several models, and 2) is somehow imbedded in the equations of scavenging.

Although the conclusion that the advection of ^{231}Pa is the most important for sedimentary $^{231}\text{Pa}/^{230}\text{Th}$ ratios is the same as in previous studies, it is notable that the contributions of the transport and bottom scavenging of each element are evaluated separately. We also believe that our successful more result about both ^{231}Pa and ^{230}Th along with GEOTRACES sections is also worth to be reported in GMD.

- English should be proofread. The meaning of several sentences remains very ambiguous or unclear.

The manuscript was already checked by a professional English proofreading service, but the revised manuscript will also go through English editing again.

To conclude on these general comments: the model and its interpretations seems

detached from what is already known on Pa/Th both in the water column and the sediment from both modeling and data studies. This manuscript shows a lack of thorough reading (state-of-the-art) of the most recent (last 10 years) literature on the subject and lacks discussion of these recent findings / conclusions. The choice of using one of the oldest model to compare these new simulation results is very odd and thus lacks a great part of the novelty added by more recent studies. There are also several conceptual problems that need to be addressed.

As mentioned above, the other recent modeling studies also showed improvement from Siddall et al. (2005) but are not necessarily the same way as the direction of improvement of our model results. We also emphasize here that our simulation of $^{231}\text{Pa}/^{230}\text{Th}$ is based on the ocean general circulation model (which is not a simplified model such as a 2D model or reduced complexity model). Our simulation which successfully reproduced the overall pattern of both the sedimentary Pa/Th ratio and dissolved Pa and Th of GEOTRACES data together with a detailed description and analysis about this model result is worth to be reported in GMD.

Following the reviewer's comments, in the revised manuscript, we will appropriately cite the recent findings of models and observations. We will then provide a more detailed discussion of the comparison between our work and recent modeling studies.

Specific comments

- L24-25: if one wants to cover the all date range, there are more recent papers than Bohm et al. (2015), e.g. Sufke et al., 2020 or Waelbroeck et al. 2018

We will add the suggested literature.

- L30: there is also Henderson and Anderson 2003 review that gives a large range of residence times (see also Costa et al., 2020 for ^{230}Th)

We will add the information of residence time from the suggested literature (130 years for ^{231}Pa and 20 years for ^{230}Th in Henderson and Anderson 2003).

- L36: for the LGM/Holocene comparison, there are more appropriate references, such as Lippold et al., 2014 which is a modeling and compilation of Atlantic data for the LGM vs. Holocene.

We will appropriately cite the suggested previous studies focusing on the LGM.

- L44 and after: several references missing or not cited appropriately. Many of the references cited cover several aspects of the Pa/Th modeling rather than only a specific aspect as the citation format made by the authors suggests.

We will cite and discuss literature covering all aspects of $^{231}\text{Pa}/^{230}\text{Th}$ modeling.

- L64: GEOTRACE database: cite

We will add a reference to the GEOTRACES project.

- L76: 43 vertical layers: are these of uniform or different heights. Be more precise.

We will add a more detailed description of our model grid.

- L81: how do you assess that you reached a steady state? explain

We will specify the criteria for determining the steady state.

- L81: Explain why you choose 100 years average rather than another number

In fact, since the residence time of ^{231}Pa and ^{230}Th is at most a few hundred years, they reach a steady state in about a thousand years, and almost no change in the average concentration of the entire ocean occurs. To remove short-term fluctuations and analyze the ocean mean state, the model is integrated over 3,000 years and the last 100-year average is used in the analysis.

- L171: “reference concentration”. It is very unclear to me, based on the information given here what is the reference concentration. More details should be given.

This reference concentration is the standard concentration for introducing the dependence of scavenging efficiency on particle concentration.