# Response to Reviewer 1

The authors greatly appreciate the reviewer for taking the time to this review. We prepared 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). Thank you very much again for your review.

The authors present an offline simulation of dissolved and particulate Pa and Th, and sedimentary Pa/Th. The effect of bottom scavenging and scavenging efficiency depending on particle concentration are discussed through sensitivity experiments. The description of the modeling results are in great detail and the authors state that the model reproduces the observations reasonably well. However, there are still many places of model-data discrepancy. I understand that it is impossible to perfectly reproduce the observation, but the mismatch should at least be discussed. In addition, this study lacks novelty. It seems that it is confirming previous other modeling results and provides limited new results for improving our understanding of marine Pa and Th cycle. Also, a critical comparison of this modeling results of sedimentary Pa/Th with other modeling studies is missing.

We agree with the reviewer that there is room for improvement in our model. We discussed the model-data discrepancy (raised by the reviewer's detailed comments) in more detail in the revised manuscript. We believe that 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. On the other hand, the reviewer's comments about our insufficient comparison with previous studies are appreciated. Following the reviewer's comments, we discussed the differences between our study and previous modeling studies (especially, by referring to more recent modeling studies than Siddall et al., 2005) in the newly added section of "Comparison with previous modeling studies" of the revised manuscript.

Detailed comments are listed below:

In the introduction, the authors reviewed previous modeling studies. From the methods part, I see this study is offline modeling, which is a big difference from other modeling studies and should be emphasized.

Yes, our simulation is offline in that OGCM physical fields were calculated in advance and given as boundary conditions. We think that this is not a problem because the Pa/Th tracer distribution does not affect the physical fields at all (in other words, the simulated Pa/Th distribution does not depend on whether the model is offline or online).

Line 100, why not using observations for calcite and opal? Why calcite and opal export productions are calculated using ratio and POC observation?

There is no available observational global dataset for POC, calcite, and opal export flux. Therefore, we estimated them from chlorophyll concentrations and empirical rain ratios. The method is the same as that used in Siddall et al. (2005).

Line 154, sensitivity experiments in this study is using off-line model, it is not appropriate to call them "OGCM experiments", also later in the text.

As described above, the results of the Pa/Th simulation do not depend on whether the model is online or offline. Therefore, our simulation is equivalent to OGCM experiments.

What is the thickness of the nepheloid layer in the model?

Since we set the ocean deepest layer of each grid as the nepheloid layer, the thickness of the nepheloid layer depends on the thickness of the corresponding deepest ocean grid cell. The thickness increases with depth, from 5 m at the surface to 250 m at the deepest layer. This treatment is the same as Rempfer et al. (2017).

Rempfer et al., (2017) includes the bottom scavenging in their model, how this bottom scavenging implementation in this study differ from Rempfer et al., (2017)?

Our implementation is basically the same as Rempfer et al. (2017). More specifically, the concentration of lithogenic particles given in the deepest layer is the same as that used in Rempfer et al. (2017) (L.159). However, because the formulation of the partitioning coefficient (K) is not the same as Rempfer et al. (2017), the actual strength of bottom scavenging depends on its formulation and choice of their model parameters. Therefore, it was difficult for us to obtain appropriate parameters about K from Rempfer et al. (2017) and we needed to find appropriate parameters from our BTM-EXP (Fig.3). This point was more clearly described in the "Experimental design" section of the revised manuscript.

Line 170, C_total and C_ref, are they global average or values in each grid? Is C_total on each grid and C_ref a global mean?

Yes, the value of C_total is differently specified on each grid whereas C_ref is given as a globally uniform. We described the meaning of the variables more precisely in the "Experimental design" section of the revised manuscript.

Figure 3, although with bottom scavenging, the Pa_d concentration decreases below ~3,000m, from the vertical profile, the observation shows similar values below 3,000m with no decreasing trend with depth, but the model shows a clear decreasing trend (Figure 3d). How to explain this model-data discrepancy? It seems that the bottom scavenging is too strong.

The dissolved 231Pa is shown in Figure 2 (not Figure 3) and 230Th in Figure 3; therefore, it is not clear which the reviewer refers to. Here, we assume it is probably about 231Pa.

For dissolved 231Pa, introducing bottom scavenging helped to reproduce the concentrations seen in the data at depths below 3000m. However, as the reviewer pointed out, the model tends to simulate lower concentration than the observations below 3000m, which needs to be improved. The improvement was not possible simply by reducing the bottom scavenging (i.e., specifying the smaller K_bottom than Fig.2c/d), therefore more fundamental improvement appears to be required. One possibility is that our treatment of the nepheloid layer (i.e., the thickness of the ocean deepest layer) may be too simple and needs to be modified so that the thickness of the nepheloid layer is more realistically specified. This model-data discrepancy and its possible reason was discussed in the "Remaining issues" section of the revised manuscript.

Line 221, the authors pointed out the lower than observation Th_d, what could potentially cause this mismatch? Can it be improved in this model?

This point was a motivation of our PCE_EXP experiment and was already discussed in the manuscript. Please refer to the experiment introducing a dependence of scavenging efficiency on particle concentration (PCE_EXP), which improved this mismatch (Figure 5d).

Line 250-251 "Similar features are also found for dissolved 230Th". For Pa_d, the maximum around 3,000 is more or less reproduced. But for Th, the maximum in the model is much deeper than the observation. Near 30W, the simulated mid-depth Th_d is much lower than the observation. Also in Figure S6.

Thank you for pointing out this interesting feature. We are also curious about this difference between 230Th and 231Pa (i.e., successful simulation for 231Pa, but not for 230Th). The introduction of more realistic bottom scavenging and the consideration of the effects of particles from the continental shelf and hydrothermal vents may help to improve the model-data agreement for 230Th. We discussed this point in the "Reproducibility along GEOTRACES GA03 and GP16 transect" and "Remaining issues" sections of the revised manuscript.

Line 251-252, the authors state that the particulate Pa and Th are well reproduced. It is not obvious in Figure S5c and d due to the colormap scale. In Figure S5c, observations are all in dark blue (hard to tell the value from the color bar), but simulation has some green-yellow values. What processes in the model cause these high values in Pa_P? Also in Figure S5, Pa_P/Th_P should also be compared with observation.

Following the reviewer's advice, we modified the figures to visualize them more clearly. As shown in the response to the comments above, the high concentration of particulate 231Pa may be related to the fact that, as with dissolved 231Pa, strong boundary scavenging is not considered in the model. We also added figures of 231Pa/230Th ratios in the water columns in each GEOTRACES section.

Line 305, results about 1D_EXP. Sedimentary Pa/Th in the Atlantic is influenced by AMOC and particle flux effect in Siddall et al., (2005), probably AMOC is the first order factor (Gu & Liu, 2017). In this study, transport includes both of these effect. More experiments can be carried out to separate the effect of ocean currents and the diffusion caused by particle effects. In this off-line model, it is computationally achievable.

As we mentioned below (our response to your comment on Line 433-434), particle fields were not calculated in the model but specified as boundary conditions in our approach. Therefore, the particle fields were unchanged (i.e., there is no particle flux effect) in all our experiments. We feel that the reviewer misunderstood this point.

How is simulated sedimentary Pa/Th in CTRL and 3D_EXP compare with observation quantitatively? In Rempfer et al., (2017), the bottom scavenging is suggested to not affect Pa_P/Th_P to a small extent and also not affect the relationship between Pa_P/Th_P and AMOC. Does this study support the results in Rempfer et al., (2017)?

We examined the difference in the quantitative agreement of sedimentary 231Pa/230Th ratios between CTRL and 3D and added it to Table S1. For addressing the relationship to AMOC, the experiment for changing the strength of the AMOC is required, which is beyond the scope of the manuscript.

Also, how is the sedimentary Pa/Th in this study compare quantitatively with other previous models? This is important because sedimentary Pa/Th is an important paleo proxy.

We summarized the model-data agreement of sedimentary 231Pa/230Th ratios in Table S1. Also, we discussed a comparison of the agreement with other modeling studies in the "Comparison with previous modeling studies" section of the revised manuscript.

Line 334-336 is confusing. Please rephrase to make it more clear.

This sentence means that most of these radionuclides are present in the dissolved phase, so the change in the ocean transport of the dissolved (not particle) phase caused the change in the distribution of the particle phases. We modified the sentences more clearly.

Line 339-340, what is the "ocean transport" here mean? Southward transport by the lower limb of Atlantic Meridional Overturning Circulation or diffusion?

The "ocean transport" here means the total difference between 3D_EXP and 1D_EXP (see Table 3) including advection and diffusion. As the reviewer mentioned, we think that the southward transport by the lower limb of AMOC is one of the main processes controlling the "ocean transport" effect.

Line 340-342, "At the same time…; as a result…" I cannot follow the logic here.

Oceanic transport decreases dissolved 231Pa in the low latitudes and increases dissolved 231Pa in the Southern Ocean. As the increased dissolved 231Pa is adsorbed by particles in the Southern Ocean, particulate 231Pa increases (Figure 7a, 7c). The statement was modified to "*As a result of the change in the dissolved 231Pa (Fig. 7a), the changes in particulate 231Pa (Fig. 7c) also take place by satisfying Eq. (5b); this leads to lower sedimentary 231Pa/230Th ratios in lower latitudes and higher ratios in higher latitudes (Figs. 7e and 7f).*".

Line 379: Residence time is calculated in CTRL_EXP and Siddall_EXP, with bottom scavenging the residence time is significantly decreased. However, the residence time in CTRL_EXP is similar to the residence time in (Gu & Liu, 2017) which does not include bottom scavenging. Does this mean the correct residence time does not necessarily need bottom scavenging?

Yes, we think that residence time depends on "total" scavenging efficiency in the ocean. Although the incorporation of bottom scavenging leads to an increase in scavenging efficiency and tends to reduce the residence time, bottom scavenging is not the sole process that controls the residence time. Therefore, for example, the model which specified the relatively stronger affinity to the particle can lead to shorter residence time even if the model does not include the bottom scavenging.

Line 426 “Part of the error comes from the oceanic flow fields simulated in the ocean model”. How is the oceanic flow simulated in this model? It can be verified against products such as ECCO (Fukumori et al., 2018). Since Atlantic sedimentary Pa_P/Th_P is greatly influenced by AMOC, what does AMOC in this model look like? This information should be provided in the manuscript.

As described in section 2.1 of our manuscript, our physical flow fields were taken from the output of a pre-industrial simulation with MIROC and its details can be obtained in Kobayashi et al. 2015 and Oka et al. 2012. For the reader’s information, we showed the meridional overturning circulation used in this study in a new figure in the revised manuscript (Figure S11).

Line 433-434, why not in this study? Results presented in this study are mostly confirmation of previous studies. With the efficiency of this off-line model, more things can be done for example test this particle fields effect on Pa and Th.

Please note that particle fields were not calculated in the model but specified as boundary conditions in our approach. The specified distribution of biological particles is estimated by satellite-based observations. Since the bias of the particle field affects the distribution of 231Pa and 230Th, our approach has advantages over the other studies where the particles are explicitly simulated in the model. For the particle field, we use the most realistic distribution, so we do not conduct experiments to change it in our study.

The summary is too verbose. Line 435-480 is repeating things in the previous section.

We revised the “Summary and concluding remarks” section to avoid verbose statements.

Line 486-489 simulated sedimentary Pa/Th under glacial times are also discussed in a 2-D model (Lippold et al., 2012) and recently in a 3-D model (Gu et al., 2020).

We referred to previous glacial modeling studies by citing the references that the reviewer pointed out.

Quantitative model data agreement (Pa_D, Th_D, Pa_P, TH_P, and sedimentary Pa/Th) and also residence time can be summarized in a table for different experiments. In that way, the performance of different experiments can be clearly seen.

We showed the model-data agreement and the residence time of each variable in Table S2 in the revised manuscript.

Minor issues:

Colorbars only have the highest and lowest values, it is not easy to tell the values in the middle.

We modified the color bar to make its values easier to read.

Line 38, particulate organic carbon.

Thank you. We changed the word.

Line 47, Gu & Liu, (2017) also show AMOC change on sedimentary Pa/Th. Also, in Gu & Liu, (2017), the particle change due to freshwater and its impact on sedimentary Pa/Th is examined and should be cited in line 432.

We cited Gu & Liu (2017) also in this line.

Figure 6e, a lot of observations overlapped. It is hard to tell the color.

We tried to modify the figure to make it easier to read.

Figure 4 can be plotted in one figure as Figure 4 in Rempfer et al., (2017). In this way, the relative difference between different experiments is clearly shown.

Thank you for this suggestion. However, because we focused on two parameters (i.e., Kref and Kbottom), we think that dependency on these two parameters is easy to read in our Figure 4 rather than combing them all in one figure.

# Response to Reviewer 2

The authors greatly appreciate the reviewer for taking the time to this review. We prepared 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 231Pa and 230Th in the oceans and underlying sediments using COCO V4.0, an Ocean General Circulation Model (OGCM), from Hasumi 2006.

They implemented 231Pa and 230Th 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 cited the recent studies not only in the "Introduction" section but also in other sections; especially in the "Comparison with previous modeling studies" section which was newly added in this revision, we compared our results with previous studies in detail.

- Similarly, the choice of comparing the COCO model outputs with those of one of the earliest models used for 231Pa and 230Th, 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 at al. (2005) simulation.

As mentioned above, we compared 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 (Kref_230Th), and (3) inclusion of particle concentration effects to Kref_230Th. 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 230Th 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 Hulten et al., 2018 focused on consideration of different particle size). We also emphasize here that our simulation of 231Pa/230Th 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 discussed our model results by adding a comparison of our results with recent modeling studies, especially in the section of “Comparison with previous modeling studies”.

- In the same vein, there is a great lack of recent literature analysis on 231Pa and 230Th, 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 (238U/232Th) activity ratio on the calculation of 231Pa and 230Thxs are neither discussed or cited. A lot of the effects that the authors are discussing in their manuscript is actually discussed in details for 230Th 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 “230Th normalization”. 230Th 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 230Th 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 230Th on 230Th in sediments. These literatures were cited in the “Remaining issues” section of 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 described the influence of the particle field on sedimentary 231Pa/230Th ratios with appropriate citations of previous studies including Missiaen et al. (2020, QSR) which discussed that changes in the particle can affect 231Pa/230Th 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 is 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 cited Anderson et al. (1983) "Removal of 230Th and 231Pa 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 230Th and 231Pa. The partition coefficient cannot be the same for both radionuclides as they as have different behaviors. The value of the exponent used here (-0.42) has been given by Henderson et al. (1999) for 230Th and is indeed not valid for 231Pa. 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 230Th (not for 231Pa).

- L90: there is one class of settling velocity in the model presented here. There are two classes in van Hulten 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 Hulten 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 require a major upgrade of its model formulation. Therefore, its direct evaluation is difficult in our model. However, in the revised manuscript, we discussed the effect of specifying settling speed, for example, by comparing our results with previous studies introducing this effect such as van Hulten 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 explained more clearly how bottom scavenging was introduced in the revised manuscript.

- Amongst the conclusions, some are included in the equation. The fact that 231Pa 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 231Pa is the most important for sedimentary 231Pa/230Th 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 231Pa and 230Th 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.

Our original manuscript was already checked by a professional English proofreading service, but in the revised manuscript, we also checked the use of English 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 231Pa/230Th 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 appropriately cited the recent findings of models and observations. We then provided 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 added 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 230Th)

We added the information of residence time from the suggested literature (130 years for 231Pa and 20 years for 230Th 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 appropriately cited 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 cited and discussed literature covering all aspects of 231Pa/230Th modeling.

- L64: GEOTRACE database: cite

We added a reference to the GEOTRACES project.

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

We added a more detailed description of our model grid.

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

We specified 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 231Pa and 230Th 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.