

Response to Reviewer 1

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). 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 will discuss the model-data discrepancy (raised by the reviewer's detailed comments) in more detail in the revised manuscript. However, 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 will discuss the differences between our study and previous modeling studies (especially, by referring to more recent modeling studies than Siddall et al., 2005) in 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 estimate 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 will be more clearly described in 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 will describe the meaning of the variables more precisely in 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 ^{231}Pa is shown in Figure 2 (not Figure 3) and ^{230}Th in Figure 3; therefore, it is not clear which the reviewer refers to. Here, we assume it is probably about ^{231}Pa .

For dissolved ^{231}Pa , 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 will be discussed in 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 ^{230}Th ”. 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 ^{230}Th and ^{231}Pa (i.e., successful simulation for ^{231}Pa , but not for ^{230}Th). 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 ^{230}Th . We will discuss this point in 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 will modify 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 will also add 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 are 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 will examine the difference in the quantitative agreement of sedimentary ²³¹Pa/²³⁰Th ratios between CTRL and 3D and add 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 will summarize the model-data agreement of sedimentary ²³¹Pa/²³⁰Th ratios in Table S1. Also, we will discuss a comparison of the agreement with other modeling studies.

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 will modify 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.

This statement is related to Line 344-366. Oceanic transport decreases dissolved ^{231}Pa in the low latitudes and increases dissolved ^{231}Pa in the Southern Ocean. As the increased dissolved ^{231}Pa is adsorbed by particles in the Southern Ocean, particulate ^{231}Pa increases (Figure 7a, 7c). The statement will be modified to “As described above, this ocean transport effect is also acting to particulate ^{231}Pa via changes in dissolved ^{231}Pa ; as a result...”.

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 will consider showing a new Figure of the meridional circulation used in this study and information about its volume transport in the revised manuscript.

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 ^{231}Pa and ^{230}Th , 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 will revise the Summary 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 will refer 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 will show 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 will modify the color bar to make its values easier to read.

Line 38, particulate organic carbon.

Thank you. We will change 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 will cite Gu & Liu (2017) also in line 432.

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

We will try 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 focus 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.