Response to Reviewer 1

The authors are greatly indebted to the reviewers who took the time to write this review. We revised manuscript by considering all comments raised by the reviewer. Below, please find our responses (black) shown after the reviewer's comments (blue).

I appreciate the authors' efforts in revising the manuscript by adding a section of the comparison with other modelling studies. However, the comparison is too descriptive without quantitative assessment and in-depth discussion. The revised manuscript still lacks "novelty" and new insights into the marine Pa and Th cycle. As the authors state in the text, bottom scavenging and dependence of scavenging efficiency on particle concentration are all confirming previous studies. Limited new insights are provided in the current manuscript.

Because our manuscript is submitted as a model description paper, we believe that it is worth to report our effort about simulating the global distribution of 231Pa and 230Th in seawater and sediment by using our 231Pa/230Th model with OGCM COCO ver4.0. Although the individual processes are already reported in previous studies as pointed out by the reviewer, it is valuable to demonstrate that combination of these processes can reproduce the overall structure of observed Pa and Th distribution from our model. Considering the reviewer's comment and the editor's suggestion, we changed the title of the manuscript in this revision: old tile " An investigation into the processes controlling the global distribution of dissolved 231Pa and 230Th in the ocean and the sedimentary 231Pa/230Th ratios by using an ocean general circulation model COCO ver4.0"→ new title "The global simulation of dissolved 231Pa and 230Th in the ocean and the sedimentary 231Pa/230Th ratios by using an ocean general circulation model COCO ver4.0".

Major:

1. "Novelty". With a computational efficient offline model, more ideas can be tested which will help the future improvement of Pa and Th modelling in those 3-D online models. For example, in section 4.5, the thickness of the nepheloid layer is proposed. Why not carry out some sensitivity experiments to see how the nepheloid layer thickness affect Pa and Th? Also, the other reviewer suggested one vs more classes of settling velocity, which is also something "new" to test. But the authors' response is "specifying the different settling speeds is unavailable

in the framework of our scavenging model and require a major upgrade of its model formulation", which is not acceptable. The authors should take the advantage of the offline framework and think carefully about experimental design to really advance our understanding of marine Pa and Th cycle.

As described above, main aim of our manuscript is to describe our 231Pa and 230Th model and report detail results of our simulation. Following the reviewers' previous comments, description about the comparison with previous modeling results (section 4.1) were also added in the previous revision. Therefore, we believe that the content of our present manuscript meets the objectives and standards of the GMD journal. As for the reviewer's comment about the thickness of nepheloid layer, we set it as the thickness of the deepest grid of each ocean grid as in Rempfer et al. (2017). Therefore, it is not possible now to freely change its thickness (also see our reply to your comment 2). As for the comment about the settling velocity, its dependency was already reported in Siddall et al. (2005); the choice of settling velocity had negligible impact on the sediment 231Pa / 230Th activity ratios (see Fig.6 in Siddall et al. 2005). Also note that our and Siddall's model assumed that the settling velocity is the same between particles, and because of this assumption, specifying the different value depending on particle type is not possible in our model framework. Therefore, as for suggestion about one vs more classes of settling velocity, we need to repeat our previous reply "specifying the different settling speeds is unavailable in the framework of our scavenging model and require a major upgrade of its model formulation". Finally, as for the reviewer's comment about "offline" model, please note that our model is not necessarily computationally efficient in that 3D tracer calculation needs to be explicitly conduced in our model, although the tracer calculation is separately performed from the physical fields. We added the following explanation in "Materials and Methods" section of the revised manuscript.

"The "offline" means that calculation of tracer is separately performed from that of physical field; since the distributions of ^{231}Pa and ^{230}Th do not affect the physical fields at all, the results do not depend on whether the model is "offline" or "online". The offline tracer model makes it easier to perform various sensitivity experiments."

2. How nepheloid layers are simulated is not described in detail. Although line 161-165 describes a little bit, it is not explicit enough for others to follow and reproduce.

To make the methodology clearer, we explained how to introduce the bottom scavenging as follows in the revised manuscript.

"Second, we perform an experiment named BTM_EXP, in which we additionally take bottom scavenging into account. Following Rempfer et al. (2017), we simply set the deepest model grid layer as the nepheloid layer. The thickness of the nepheloid layer becomes equal to the thickness of the corresponding the deepest model grid layer which varies between 5 and 250 m depending on the depth. The intensity of the bottom scavenging depends on two parameters: the partition coefficient (K_{bottom}) and the concentration (C_{bottom}) of the bottom particles. Our treatment about C_{bottom} is the same that in Rempfer et al. (2017); we assume a globally uniform value for C_{bottom} (6.0×10^{-8} g cm^{-3}) which is within the range of 4.0×10^{-8} to 1.65×10^{-6} g cm^{-3} observed in the benthic nepheloid layers in the North Atlantic (Lam et al., 2015). As for K_{bottom}, because our formulation of the reversible scavenging is not the same as Rempfer et al. (2017), we needed to find its appropriate parameter value. For this purpose, we perform a number of simulations with different bottom scavenging intensities by changing the value of K_{bottom}."

3. Section 4.1: I appreciate the authors adding this section to have a comparison with other modelling works. However, the majority of this session is what we already know: without bottom/boundary scavenging, the model will overestimate deep dissolved Pa and Th concentrations, which is already pointed out/discussed in previous literature. This revision still lacks in-depth and quantitative comparison with other modelling works, for example, Rempfer et al., 2017.

Considering the reviewer's comment, we modified the section 4.1 to emphasize the comparison with the previous studies including Rempfer et al. (2017). The discussion about comparison with previous 231Pa/230Th modeling studies on model-data comparison, which was previously stated in the second half of the section 4.4, was moved to the section 4.1 and we tried to make quantitative comparison there in the revised manuscript.

4. Section 4.3: Three experiments (1D, 3D, CTRL) are used to decompose the processes controlling sedimentary Pa/Th. In 1D, with only reversible scavenging, the sedimentary Pa/Th is 0.093, which is common knowledge and the dissolve phase distribution in this scenario has already been discussed in previous literature (e.g., Siddall et al., 2005). Line 358-Line 367 seems to be redundant and repeats what we already know. Similar for Line371-372.

As you mentioned, these statements are not new findings. In these statements, we just intended to confirm that our result can be interpreted from common knowledge. We still think that these statements are helpful to understand/discuss the differences between our three experiments. In the revised manuscript, we added the words “although it is well known from previous studies,” in this discussion.

5. Section 4.3: The “new” finding in this section to me is how ocean transport and bottom scavenging on each Pa and Th change sedimentary Pa/Th. This part can be improved by quantitatively comparing the effect on Pa and effect on Th; and also compare with Rempfer et al., 2017 results, which has the experiment with ocean circulation (similar to 3D) and ocean circulation & bottom scavenging (similar CTRL here), so that the robustness of the current results (Figure 8e, 9) can be verified.

Thank you for this suggestion. As for your suggestion about comparing the effect on Pa and effect on Th, this was already discussed with Figure 9c (Pa bottom scavenging) and Figure 9d (Th bottom scavenging) in our manuscript. As for comparison with Rempfer et al. (2017), because they did not show the individual effect from Pa and Th, comparison of our Figures 9c and 9d with Rempfer et al. (2017) was not possible. It is also difficult for us to directly compare our Figure 8e with Rempfer et al. (2017) because differences in sedimentary Pa/Th distribution between the experiments are not explicitly shown in Rempfer et al. (2017).

6. Figures 6f, 7f, 8f are mentioned in the text, but missing in the figures.

We are sorry for the missing of the figures in our previous submission. We included these figures in the revised manuscript.

7. Line 450-452 “Our CTRL_EXP…” The authors claim that their results are more realistic than others, based on their more realistic dissolved Pa and Th. This is not convincing. If the authors can provide the quantitative comparison (different model vs same observation, RMSD and correlation as Table S1) showing better model-data agreement, then it is convincing.

This statement refers to the comparison between the work of Siddall et al. (2005) and ours. It was rewritten as follows:

“Compared with Siddall_EXP based on Siddall et al. (2005), our CTRL_EXP can realistically simulate not only oceanic distribution of ^{231}Pa and ^{230}Th but also their residence time by

introducing the bottom scavenging and the dependence of scavenging efficiency on particulate concentration."

Minor:

Line 25: There are many more references using Pa/Th sedimentary ratio on past ocean circulation, suggest adding "e.g.,"

We have revised the manuscript as you have pointed out.

Line 36-40: "For example…" and "Some modeling…" These two sentences are not logically coherent and it reads to me there is another sentence after the second sentence. I guess the authors intend to say the model suggests stronger or similar LGM AMOC, therefore implementing Pa and Th in the model is important?

Yes, to avoid the confusion, we modified the sentences as follows in the revised manuscript so that the detail description about the LGM AMOC is not explicitly mentioned.

"To use the sedimentary $^{231}Pa/^{230}Th$ ratios as a proxy for ocean circulation in a more quantitative manner, the modeling about ^{231}Pa and ^{230}Th is important."

Line 49-50: Gu and Liu, 2017; Rempfer et al., 2017; van Hulten et al., 2018; Missiaen et al., 2020a are 3D ocean models but cited as 2D models.

Thank you for pointing this out. We have cited them as studies using 3D models.

Line 243-244: Authors state that PCE_EXP matches better than KREF_EXP. From Figuren5d, the agreement above 3,500m is obviously improved in PCE_EXP, but below 3,500m, KREF_EXP seems to agree better as the observation shows maximum value ~5km (also in KREF_EXP), but in PCE_EXP, the maximum value is ~4km. The better agreement with GEOTRACES in PCE_EXP in Table S1 is probably contributed by the upper ocean. Why there is a difference between the upper ocean and the abyssal ocean. More details should be discussed.

The difference between PCE_EXP and KREF_EXP below 3.5km is explained by the change in the partition coefficients (see Figure 5c). Due to introduction of the dependence of particle concentration on scavenging efficiency, the reference partition coefficient varies with ocean region in PCE_EXP (Fig. 5c): in the deep ocean below 3,500m, it is significantly higher than 1x10^7 (=value assumed in KREF_EXP) in the low-latitude regions. This caused the lower concentration below 3.5km in PCE_EXP than KREF_EXP. As for the underestimation of Th in the abyssal ocean, we added the following statement in the section 4.5 of the revised manuscript.

"The dissolved 230Th simulated in CTRL_EXP (Fig. 5b) also tends to underestimate the observed concentration near the sea bottom. 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."

Line 371: "CTL_EXP" typo.

This is fixed in the revised manuscript. Thank you.

Response to Reviewer 2

The authors greatly appreciate the reviewer who took the time to this review. We prepared a revised manuscript by considering all comments raised by the reviewer. Below, please find our responses (black) shown after the reviewer comments (blue).

The authors nicely improved the manuscript in this revised version. They have answered most of my comments / questions / suggestions / concerns. I still have a few comments that needs to be answered and a few minor adjustments are needed before publication.

One of my concern is that the justification for choosing a comparison with the simulation(s) made by Siddall et al. (2005), i.e. rather old results, which have since been discussed and improved, does not clearly appear. The study will indeed gain from a justification of this choice that should be given at the beginning of the manuscript, i.e. end of the "Introduction" and/or "Experimental design".

We are grateful for the advice. Following the reviewer's comment, we added the following statement at the beginning of section 2.5 "Experimental design" in the revised manuscript.

"As stated in the Introduction, Siddall et al. (2005) was a pioneering 3D model for global simulation of both ^{231}Pa and ^{230}Th. This model is now a relatively old model and the reversible scavenging model introduced in this model is simpler than more recent models. However, this model appropriately reproduced the observed distribution of sedimentary $^{231}Pa/^{230}Th$ ratios as shown in their Fig. 2 which appears not necessarily inferior to that in more recent models. Therefore, in this study, we start with Siddall_EXP where this most basic reversible scavenging model of Siddall et al. (2005) is introduced."

As underlined by the other reviewer, the fact that the model is offline is not correctly emphasized. From my point of view, it is not a weakness but a strength from this model because it makes it indeed easier to manipulate.

Thank you for this comment. Following your comment, we added the following statement in "Materials and Methods" section of the revised manuscript.

"The "offline" means that calculation of tracer is separately performed from that of physical

field; since the distributions of ^{231}Pa and ^{230}Th do not affect the physical fields at all, the results do not depend on whether the model is "offline" or "online". The offline tracer model makes it easier to perform various sensitivity experiments."

Figures with "vertical profile averaged horizontally" (1, 2, 3, 4 and 5): I am not sure I understand this term correctly. An explanation must be provided. Does it mean you choose 1) to make an average of all the dissPa concentrations at a given depth along the entire transect? Or 2) to average the value along a given latitude for the entire basin for each layer?

Then what is the representativeness of this averaged value (orange points) in the Atlantic because: case 1) The north and south Atlantic have very different behaviors with 231Pa being strongly entrained in the AMOC in the north and 231Pa being strongly scavenged by opal-rich particles in the south. Case 2) the west and east basins have different behaviors for diss Pa (diss Th probably to a lesser extend).

The figure caption was modified to be "vertical profile (the latitudinal mean along 30°W in the Atlantic Ocean)" in the revised manuscript. As you pointed out, this average masks the difference between north and south parts of the Atlantic basin. But this north-south difference is explicitly shown in other figures (e.g. Fig. 1a for Pa; Fig. 2c for Th) in our manuscript.

L38-41. As you described the conclusions based on the data analyses (lines above), you should also describe the main conclusions of the 2 models you are citing in this subsection.

By responding to the other reviewer's comment, this sentence was removed and replaced by the following statement in the revised manuscript.

"To use the sedimentary $^{231}Pa/^{230}Th$ ratios as a proxy for ocean circulation in a more quantitative manner, the modeling about ^{231}Pa and ^{230}Th is important."

L45. "sinking particles scavenge 230Th more strongly ". I suggest to replace strongly by efficiently.

We have revised the manuscript as you have pointed out.

L45-47. While the work by Chase et al. is highly cited, there were much earlier studies showing the effect of opal such as Rutgers van der Loeff and Berger (Deep Sea Res. 1, 40, 1993) or Walter et al. (Earth Planet Sci Lett 149, 1997). Some of these studies are cited later in the manuscript but should also appear in the Introduction as this effect has been known for a long time.

Thank you for the references. We have cited them in revised the manuscript.

L48-51. Citations should be proofread, e.g. Rempfer et al., van Hulten et al., Missiaen et al., 2020a are not 2D ocean models: please carefully check these references and to which model they correspond.

Thank you for pointing this out. We have cited them as studies using 3D models.

L63-65. While the approach was different from that of the authors' model, the effect of efficiency of scavenging depending on particle concentration has been explored by recent models, e.g., van Hulten et al. (2018); Missiaen et al. (2020a and 2020b), even if the approach was different. It should be mentioned here.

Thank you for the suggestion. We modified the sentence in the revised manuscript by citing the references the reviewer suggested.

"... this effect has not been directly considered by recent modeling studies but some studies have evaluated the impacts of changes in particle concentration and scavenging efficiency on the distribution of ^{231}Pa *and* ^{230}Th *(van Hulten et al., 2018; Missiaen et al., 2020a and 2020b)"*

L124. I suggest that, for non-model specialists, you explain the term "convection" as you did in your answer to my initial review. This was a clear and short explanation that could make your paper more accessible to a broader audience.

Because the "convection" term is included as a part of diffusion term in our offline tracer calculation, we simplified our explanation so that the term "convection" is not explicitly described in the revised manuscript.

L165. This part needs clarification.

You write that the benthic nepheloid layer is 50 to 130m above the bottom. Do you mean thickness? i.e., bottom to bottom+50m for 50m? In addition, in your answer to reviewers, you say that the thickness of the nepheloid layer increases from 5 to 250m. There is a discrepancy with what is written in the manuscript. Please clarify this point.

The values of 50-130 mean the depth above bottom of observed maximum suspended particulate matter reported in Lam et al. (2015, DSR). We set the deepest grid cell as the nepheloid layer. Therefore, the thickness of the nepheloid layer is equal to the thickness of the corresponding deepest grid cell. The thickness of grid cell increases with depth from 5 to 250 m in our ocean model. To make this point clearer, we modified the description as follows in the revised manuscript.

"Second, we perform an experiment named BTM_EXP, in which we additionally take bottom scavenging into account. Following Rempfer et al. (2017), we simply set the deepest model grid layer as the nepheloid layer. The thickness of the nepheloid layer becomes equal to the thickness of the corresponding the deepest model grid layer which varies between 5 and 250 m depending on the depth. The intensity of the bottom scavenging depends on two parameters: the partition coefficient (K_{bottom}) and the concentration (C_{bottom}) of the bottom particles. Our treatment about C_{bottom} is the same that in Rempfer et al. (2017); we assume a globally uniform value for C_{bottom} (6.0×10^{-8} g cm^{-3}) which is within the range of 4.0×10^{-8} to 1.65×10^{-6} g cm^{-3} observed in the benthic nepheloid layers in the North Atlantic (Lam et al., 2015). As for K_{bottom}, because our formulation of the reversible scavenging is not the same as Rempfer et al. (2017), we needed to find its appropriate parameter value. For this purpose, we perform a number of simulations with different bottom scavenging intensities by changing the value of K_{bottom}."

L250-253. Since you are comparing the results of your simulations to that of other models for the Southern Ocean when discussing particulate Pa and Th (section 3.3) it would be good that have a similar comparison for the dissolved Pa and Th at the end of section 3.2. even if the other models indeed use slightly different approaches to simulate the effect of changing adsorption coefficient in the Southern Ocean (e.g. Rempfer et al., 2017 and Missiaen et al., 2020a).

Thank you for the suggestion. We added the following sentences near the end of section 3.2.

"The distributions of ^{230}Th simulated in previous modeling studies (e.g., Figs. 4 and 5 in Dutay et al., 2009; Fig. 2 in Siddall et al., 2005; Fig. 2 in Gu and Liu; Fig. 3 in Rempfer et al., 2017;

Fig. 12 in van Hulten et al., 2018; Fig. S3 in Missiaen et al., 2020a) are basically similar to our result (Fig. 6b); however, our simulation is the best at reproducing the high concentration in the Southern Ocean."

L301. typo: Missiaen (not Messiaen)

This is fixed in the revised the manuscript. Thank you.

L320. typo: GEOTRACES (not GAOTRACES)

This is fixed in the revised the manuscript. Thank you.

L334-336. "In this transect, the observational data shows a clear signal associated with hydrothermal vents": please explain the underlying mechanism, i.e. how would hydrothermal vents affect both diss. and part. Pa and Th concentrations. A reference is also needed. May be also the earlier findings of this mechanism, e.g. Shimmield and Price, Geochim Cosmochim Acta 52, 1988.

Thank you for the information. We explained the underlying mechanism related to hydrothermal activities in this transect as follows:

"It has been pointed out that trace metals from hydrothermal activities may cause additional removal of 231Pa and 230Th (Shimmield and Price 1988; Lopez et al., 2015; Rutgers van der Loeff et al., 2016; German et al., 2016). Along the GEOTRACES GP16 section, 230Th and 231Pa have been found to decrease with increasing trace metals of iron and manganese supplied from hydrothermal vents (Pavia et al., 2018)."

L337. replace "our scope" by "the scope of this study"

We have revised the manuscript by following your suggestion.

L341. "we discuss about": remove "about"

We have revised the manuscript by following your suggestion.

L401. “as a matter of course” replace by “as a matter of fact”

We have revised the manuscript by following your suggestion.

L405. “231Pa transported toward the Southern Ocean is expected to be immediately removed there due to the high opal flux”. This is an overstatement. Immediately should be replaced by “quickly” or “very quickly” or “quicker than in the open ocean”. Both data and modeling studies show that there is still some Pa exported within the Southern Ocean

Thank you for pointing out this. We have revised the manuscript by following your suggestion.

L408-410. “This result implies that scavenging of 230Th is not so efficient in the Southern Ocean as previously expected due to the dependence of scavenging efficiency on particle concentration.”. In Missiaen et al. (2020a), they simulate the effect of halving of 1) the total particle flux, 2) the POC, 3) the CaCO3 and 4) opal. They show that for Th, it results in increasing the dissolved 230Th concentration in the Southern Ocean in case 1) and further show that the main effect comes from POC and opal. How does it compare with your results on scavenging efficiency and particle concentration? Can you link both results?

Thank you for this discussion. Following your suggestion, we added the following discussion in the revised manuscript.

“Missiaen et al. (2020a) demonstrated that the dissolved ^{230}Th concentration in the Southern Ocean will increase if the effect of particle scavenging is halved and that most of this effect come from POC and opal. This implies the scavenging of ^{230}Th is controlled also by the opal in the Southern Ocean. Together with their and our results, quantification about scavenging of Th by opal in the Southern Ocean may be a key for more accurate understanding of ^{231}Pa/^{230}Th ratios in the global ocean.”

L453: also add the ref to Henderson and Anderson for residence time

We have cited the reference in revised the manuscript.

L498. “(i.e., specifying the smaller *KK*Pabottom than Figs. 2c and 2d)” do you actually mean: “(i.e., specifying smaller *KK*Pabottom than in Figs. 2c and 2d)”?

Yes. We have revised the manuscript as you have pointed out.

L819. “Mangini” not “Mangianini”

This is fixed in the revised manuscript. Thank you.