- Review on "A global scavenging and circulation ocean
- ² model for thorium-230 and protactinium-231"

3

4 1 Scientific contribution

The authors present a model for the scavenging of ²³¹Pa and ²³⁰Th embedded within an ocean 5 biogeochemistry circulation model. Novel attributes of the model include (i) the simulation of 6 lithogenic particles within nominal "small" and "big" (which are meant to represent slow and fast 7 sinking) particle classes, (ii) the simulation of POC in small and big particles based on a the re-8 cently implemented reactivity model within PISCES, and (iii) the inclusion of tracer equations for 9 the radionuclides in the dissolved, small, and big particle phases, with the caveat that the dissolved 10 radionuclides are assumed to immediately equilibrate with small and big particulate radionuclides. 11 The authors describe an improvement in fit to the dissolved and particulate radionuclide data, com-12 pared to a previous model with similar physics but an older implementation of the biogeocehmistry 13 model within PISCES. Thus, this study presents ProThorP 0.1 as a better description of ²³¹Pa and 14 ²³⁰Th in the ocean, and one that may be contribute to our understanding of the response of these 15 radionuclides to changes in circulation and sediment resuspension in the modern ocean and the 16 ocean of the geologic past. 17

18 2 Scientific Quality

Overall, I believe the methodology and discussion of results are of good scientific quality.
 However, there are a few key points that I think need to be elaborated upon or revised:

1). For POC, GOC, biogenic silia, and $CaCO_3$ the values of K used in this study are within 21 the range found in previous laboratory experiments and field studies. While this range is large, 22 and has shown variation with both particle concentration and composition in both the field and 23 laboratory experiments, I think it is defendable to include one value of each particle type, given 24 that the goal of this study is to present a revised model for the cycling of ²³¹Pa and ²³⁰Th in the 25 ocean, and not to provide a sensitivity analysis of the radionuclide activities to variations in K (but 26 see point 3 below regarding this issue). However, I question the use of the K values for lithogenic 27 material in this study. For small particles, the values used for both isotopes are much larger than 28 \pm 1 standard deviation of the value found by *Hayes et al.* (2015), the range in *Geibert and Usbeck* 29 (2004), or the range from other previous studies (Anderson et al., 1992; Chase et al., 2002). The 30

only study I have seen a *K* value for lithogenic material greater than that reported used this study was by *Luo and Ku* (2004) ($K = 230 \times 10^6$ g/g), though this value mostly reflected the variability in *K* with the % lithogenic composition of sediment trap material from the Southern Ocean and the Equatorial Pacific. Similarly, the source for *K* for small particle POC is unclear; these values for both isotopes appear to be based on those derived by *Hayes et al.* (2015), but why was the higher end of the range used for ²³⁰Th, and the low end of the range used for ²³¹Pa? The authors should state explicitly how they arrived at the values of *K* for both POC and lithogenic material.

2). PISCES includes aggregation between small and big POC (Aumont et al., 2015), but in 38 equations 9(b,c) I do not see any terms that represent aggregation of particulate ²³¹Pa and ²³⁰Th 39 from small to big particles. The authors justify the omission of adsorption and desorption in 40 equations 9(a,b,c) by assuming an instant equilibrium between the dissolved and the two particulate 41 phases, but I do not see a similar argument for the omission of aggregation of POC given. I think 42 the authors need to clarify whether and how aggregation of the particulate radionuclide pools is or 43 is not considered. Additionally, I am curious as to how difficult it would be to include aggregation 44 of the sPOC-bound radionuclide pool in the model, since the aggregation rate can be derived from 45 eq. (39) of Aumont et al. (2015). For example, could an aggregation term be included in equations 46 9(b,c): 47

$$\frac{dA_{i,S/B}}{dt} = \dots \pm \beta A_{i,D} K_{i,POC} P^{POC} \dots,$$
(1)

where the aggregation rate represents a loss for radionuclides bound to small particles (*S*), a source for radionuclides bound to big POC (*B*), and *B* is the aggregation rate parameter (d⁻¹). Including this term in the current setup may not make sense considering the authors already assume instant equilibriation with respect to adsorption and desorption, and POC specific aggregation rates may be on the order of adsorption and desorption rates (0.1 to 1 yr⁻¹, *Murnane* (1994a)) or faster (*Burd*, 2013), but I think it is important to at least make explicit why aggregation of radionuclides in the small POC pool is or is not considered.

⁵⁵ 3). In the conclusion, along with mentioning the potential missing effects of nepheloid lay-

ers, Mn (oxyhydr)oxides, and a stronger AMOC on simulated ²³¹Pa and ²³⁰Th, I also suggest that 56 the authors mention the sensitivities of these distributions to K for the various particle phases. Al-57 though a sensitivity analysis may not be the goal of this study, I think it is at least worth speculating 58 on how changing K values from those used in this study could alter and potentially improve the 59 simulated distributions. Reported K values for any given particle type may vary by up to three 60 orders of magnitude (e.g., 10^{3.98} from *Guo et al.* (2002) vs. up to 10^{6.23} from *Geibert and Usbeck* 61 (2004) for SiO₂), may depend on whether K is derived from field observations or laboratory exper-62 iments, and may depend on differences in types of particles used in laboratory experiments (e.g., 63 inorganic vs. biogenic SiO₂ or CaCO₃). Given such large ranges, it is not unreasonable to suspect 64 that a different, yet still plausible, combination of K values could better describe the available data. 65 Minor Comments: 66

1). The authors mention that this model is an improvement of that presented in *Dutay et al.* (2009). I think this improvement should be shown in the paper. For example, a table showing the root mean square deviation and/or r for dissolved and particulate ²³⁰Th and ²³¹Pa in the dissolved and small particulate pool (there may not be enough observations to compare the fit to big particles) for both models (the one presented and the one from *Dutay et al.* (2009)) could be provided. This way, the improvement in fit obtained by the new model, which is one of the key results of this study, is made explicit.

⁷⁴ 2). The authors provide r for the fit between simulated and observed dissolved ²³⁰Th at GA02 ⁷⁵ and GA03. A similar comparison should be presented for ²³¹Pa along both sections, ²³⁰Th and ⁷⁶ ²³¹Pa in the small particle pool at GA03 (I don't think these particulate radioisotopes have been ⁷⁷ reported along GA03).

78 **3** Scientific reproducibility

The model description appears sufficient to allow the reproduction of the results, especially
 given a link to the model code has been made available in the paper.

4 Presentation quality

The overall presentation of the paper is clear, concise, and well-organized. However, I have a few specific suggestions to improve the clarity and presentation of the study. I have put suggested changes to wording in bold.

1) Page 2, lines 26-28: I suggest this sentence be rephrased: "Other factors may affect the
adsorption of radionuclides onto particles. For example, smaller particles have larger surface
area to volume ratios, which results in an increase in the number of these radionuclides
adsorbed per particle. Additionally, while the adsorption rate is expected to increase with
particle concentration, the adsorption rate may be partly limited by the coagulation of nonfilterable particles (or colloids) to filterable particles (*Honeyman et al.*, 1988; *Honeyman and Santschi*, 1989)."

2) Figure 1: I do not see DOM in the figure, but it is mentioned in the caption. I would either add this pool to the figure, or remove it from the caption. Additionally, the caption states "sPOM and "bPOM" stand for small and big Particulate Organic Matter", but the text in the figure is just written as "small" and "big," not "sPOM" and "bPOM" Again, please either amend the caption to reflect what is written in the figure, or change the text in the figure to reflect what is stated in the caption.

⁹⁸ 3) Page 5, line 7-8: *Aumont et al.* (2015) mark the boundary between small and big particles as ⁹⁹ 100 μ m. However, this boundary is nominal, so it should not matter whether whether the authors ¹⁰⁰ define the boundary as 51 μ m or 100 μ m. Still, for clarity it may be worth mentioning in this ¹⁰¹ sentence the different boundary between small and big particles defined in *Aumont et al.* (2015) ¹⁰² and this study.

4). In table 3, you should specify for each field study whether the observations are available in the dissolved pool, small particle pool, big particle pool, all pools, or some combination of the three. This can simply be done in parentheses after each expedition or each ocean basin in the "Radionuclides ²³⁰Th and ²³¹Pa" section of the table (e.g., US GT10 and GT11 (Th_{D,S,B}, Pa_{D,S,B}), to keep with the nomenclature used in the paper, would indicate that ²³⁰Th and ²³¹Pa are available in the dissolved, small, and large pools along this section). I don't think this would add too much
 more text to the table, so that it will still be easy to read.

5). Page 12, lines 8-9: I would rephrase this sentence: "Contrarily, *total* observed [CaCO₃], which is only provided along section GA03, where small particulate CaCO₃ measurements are available,..."

6). Page 22, line 7: Please change (Fig. ??) to (Fig. **S1**).

7). Page 22, line 14: Please provide a few reasons for this complexity. *Luo et al.* (2010) go into some detail on the various factor influence sedimentary ²³¹Pa/²³⁰Th, including the rate of overturning, the rate of particle scavenging, the geometry of the overturning cell, and the specific latitude of deep water formation.

8). Page 23, lines 32-33: I would rephrase the second part of this sentence: "..., particularly
when regions of high particle concentration (e.g., ocean margins and nepheloid layers) are
included."

9). Page 23, below Conclusion, line 11: delete "wanted to".

10). Page 24, line 16: I would rephrase part of this sentence: "... we improved the radionuclide
 distributions and fluxes, ..."

124 **References**

- Anderson, H. L., R. François, and S. B. Moran (1992), Experimental evidence for differential
 adsorption of Th and Pa on different solid phases in seawater, *EOS*, 73, 270.
- Aumont, O., C. Éthé, A. Tagliabue, L. Bopp, and M. Gehlen (2015), PISCES-v2: an ocean bio geochemical model for carbon and ecosystem studies, *Geoscientific Model Development*, 8(8),
 2465–2513.
- Burd, A. B. (2013), Modeling Particle Aggregation Using Size Class and Size Spectrum Approaches, *J. Geophys. Res. Oceans*, *118*(7), 3431–3443.
- ¹³² Chase, Z., R. F. Anderson, M. Q. Fleisher, and P. W. Kubik (2002), The influence of particle
 ¹³³ composition and particle flux on scavenging of Th, Pa and Be in the ocean, *Earth Planet. Sci.*¹³⁴ Lett., 204(1), 215–229.
- Dutay, J. C., F. Lacan, M. RoyBarman, and L. Bopp (2009), Influence of particle size and type
 on ²³¹Pa and ²³⁰Th simulation with a global coupled biogeochemical-ocean general circulation
 model: A first approach, *Geochemistry, Geophysics, Geosystems, 10*(1), 1–26.
- Geibert, W., and R. Usbeck (2004), Adsorption of thorium and protactinium onto different particle
 types: experimental findings, *Geochim. Cosmochim. Acta*, 68(7), 1489–1501.
- Guo, L., C. C. Hung, P. H. Santschi, and I. D. Walsh (2002), ²³⁴Th scavenging and its relationship
 to acid polysaccharide abundance in the Gulf of Mexico, *Mar. Chem.*, 78(2), 103–119.
- 142 Hayes, C. T., R. F. Anderson, M. Q. Fleisher, S. W. Vivancos, P. J. Lam, D. C. Ohnemus, K. F.
- Huang, L. F. Robinson, Y. Lu, H. Cheng, R. L. Edwards, and B. S. Moran (2015), Intensity of Th
 and Pa scavenging partitioned by particle chemistry in the North Atlantic Ocean, *Mar. Chem.*, *170*, 49–60.
- Honeyman, B. D., and P. H. Santschi (1989), A Brownian-pumping model for oceanic trace metal
 scavenging: evidence from Th isotopes, *J. Mar. Res.*, 47(4), 951–992.

- Honeyman, B. D., L. S. Balistrieri, and J. W. Murray (1988), Oceanic trace metal scavenging: the
 importance of particle concentration, *Deep Sea Res. A*, 35(2), 227–246.
- Luo, S., and T. L. Ku (2004), On the importance of opal, carbonate, and lithogenic clays in scavenging and fractionating ²³⁰Th, ²³⁰Pa and ¹⁰Be in the ocean, *Earth Planet. Sci. Lett.*, 220(1), 201–211.
- ¹⁵³ Luo, Y., R. Francois, and S. E. Allen (2010), Sediment ²³¹Pa/²³⁰Th as a recorder of the rate of the ¹⁵⁴ Atlantic meridional overturning circulation: insights from a 2-D model, *Ocean Science*, 6(1), ¹⁵⁵ 381–400.
- ¹⁵⁶ Murnane, R. J. (1994a), Determination of thorium and particulate matter cycling parameters at ¹⁵⁷ station P: A reanalysis and comparison of least squares techniques, *J. Geophys. Res. Oceans*, ¹⁵⁸ *99*(C2), 3393–3405.