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Interactive comment on “Evolution of ^{231}Pa and ^{230}Th in overflow waters of the North Atlantic” *by* Feifei Deng et al.

Anonymous Referee #3

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Deng and coworkers have produced an important data set by analyzing samples collected on GEOTRACES Section GA01 (GEOVIDE) for 231Pa and 230Th. These results hold valuable implications for the use of these radionuclides as tracers of North Atlantic deep water ventilation, and its variability through time (via the analysis of 231Pa/230Th ratios, henceforth “Pa/Th”, archived in marine sediments). However, there are some major issues that should be addressed before I can recommend that the manuscript be published, as detailed in the following.

Major Comments:

1) Clarify and emphasize the principal take home message. The concluding sentence of the manuscript states “and continues to support the use of sedimentary 231Pa/230Th measurements at a basin scale to constrain overturning circulation.” This

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statement is based on the calculated southward export of dissolved Pa being substantially greater than the southward transport of Th. However, as clearly stated in the manuscript, there is no observable relationship between dissolved Pa/Th ratio and water mass age. This observation is in direct contradiction to the principles underlying the use of sedimentary Pa/Th ratios to reconstruct past variability of the ventilation of deep water in the North Atlantic Ocean, where it is assumed that dissolved Pa/Th ratios will increase monotonically with age after water mass formation due to the longer residence time of Pa compared to Th. How can the authors conclude that their results support the use of sedimentary Pa/Th ratios to constrain overturning circulation when there is no relationship between dissolved Pa/Th ratio and water mass age?

This issue becomes even more important if one considers the evolution over time of dissolved Pa/Th ratios down the length of the western Atlantic Ocean. Although the authors do not present Pa/Th ratios for the mid-latitude North Atlantic or at 40°S along with the dissolved 230Th and 213Pa data in Figure 3, eyeballing the dissolved 230Th and 231Pa profiles for these regions suggests very little change in the dissolved Pa/Th ratio from north to south, from GEOVIDE near the formation region to GA10 at 40°S. If a more rigorous analysis of the data reveals this to be true, i.e., that there is no change with water mass age in the dissolved Pa/Th ratio down the entire length of the Atlantic Ocean, then I do not see how Pa/Th ratios can be related to ventilation rate, either in the modern ocean or to reconstruct climate-related changes in ventilation rate in the past.

The manuscript would have much greater impact if this point were discussed at length, incorporating data from the entire Atlantic Ocean.

2) The development and application of CFC ages are unclear. The description in the Supplementary material (page 8) is helpful, but some of the output is not meaningful. CFC ages are not valid for time periods older than the initial introduction of CFCs into the environment in the middle of the 20th century. My colleagues who are experts in the use of CFC ages generally decline to interpret apparent ages greater than about 40

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to 50 years due to the uncertainties inherent in interpreting CFC ages in water masses last exposed to the atmosphere during the earliest days when CFCs were tagging water masses. Therefore, I do not understand how Mediterranean Water can be assigned an age of 91±8 years, or NEADW can be assigned an age of 989±48 years (Table S2). Unless there is something not explained in the paper that allows CFC ages this old to be computed, the old ages should be removed from the paper. Accordingly, Figure 6a can be removed, leaving only Figure 6b in the paper.

Related to Figure 6, it is very confusing that the two panels have different longitude scales, but the scale for Figure 6a is not shown. If there is a reason to retain Figure 6a, then include the longitude scale and note that Figure 6b incorporates only the western half (approximately) of Figure 6a.

3) The calculation and interpretation of scavenging (rates and percentages) relies strongly on the estimated CFC ages (Figures 7 through 9). Given this important sensitivity to estimated age, I recommend that the authors include a discussion of the uncertainty in the CFC ages, and how that may affect their interpretation.

4) As discussed on page 6 of the manuscript, the initial (preformed) concentrations of dissolved 230Th and 231Pa at the time the water masses formed are unknown, so the authors assume that the preformed concentrations are either equal to average concentrations in surface waters sampled along the GEOVIDE transect or that preformed concentrations are zero. Unfortunately, until data are available for the Nordic Seas and the Labrador Sea during times of winter convection, these may be the only options available for the type of analysis described here. Nevertheless, it would be helpful if the authors provided additional discussion of the sensitivity of their derived products (e.g., fraction scavenged for each isotope) to the values assumed for the preformed concentration.

In this context, it would also be helpful to discuss the possibility that each water mass has a different preformed concentration, and how this might affect the interpretation of the data presented in Figure 8. Implicit in the presentation of the data presented in Figure 8 is the assumption that all water masses have the same preformed concentrations. What if this is not the case? How would that alter the interpretation of the data?

5) There seems to be a problem with the model curves shown in Figure 9, where the solid lines depict model results for the case where average surface water concentrations determined for samples collected on the GEOVIDE cruise were used in place of the preformed concentrations. If that were the case, then why do the projected model concentrations at zero age (solid lines) intersect the Y axis at concentrations about double the values reported for average surface concentrations on page 6 (0.108 dpm/1000 L for 230Th and 0.089 dpm/1000 L for 231Pa)? If I understand the model correctly, then the concentrations at zero age should equal the assigned preformed concentrations. Is this not the case? Is the problem that the preformed concentrations are introduced twice in equation 6 (supplementary material)? Note that C(pre) and C(surface) are one and the same. Should both terms be in equation 6?

6) Summary of major comments: Given all of the uncertainties in CFC age and in initial (preformed) concentrations of Pa and Th, it seems that a stronger paper than the one under review would be produced by integrating the new data from GEOVIDE with other data from GEOTRACES cruises down the length of the Atlantic Ocean (GA02, GA03, GA10 and, perhaps, other sections with data in the GEOTRACES IDP2017, if there are any) to establish firmly whether or not the dissolved Pa/Th ratio in deep Atlantic water evolves over time as assumed in the application of sedimentary Pa/Th ratios to constrain past changes in the rate of ventilation of North Atlantic Deep Water.

Minor comments:

1) The authors report their results using historical units (dpm/1000 L) but their results will be converted to SI units when included in the next GEOTRACES data product. All of the Th and Pa data currently in the IDP2017 are presented using SI units, so

why not make this conversion before publishing the GEOVIDE data? 2) page 6 line 1 “Steinfeldt” is misspelled. 3) page 6, definition of “Ingrown component”: Are U concentrations normalized to a constant salinity? To the salinity measured for each sample? Something else? 4) page 7 line 16: “three times more 230Th has been removed by scavenging” than “what?” Complete the description of the comparison being made. 5) page 8 line 7: Change “run” to “ran” 6) page 8 line 27: delete the “in” prior to “from DSOW” 7) page 8 line 28, and elsewhere: Bottom scavenging of 230Th was first noted by Bacon and Anderson (1982) and by Anderson et al. (1983; EPSL 66(1-3), 73-90, not the paper cited by Deng et al.) in their study of the eastern tropical Pacific. These early indications of bottom scavenging should be cited. 8) page 9 line 12: What is the source of the average 230Th and 231Pa concentrations in the upper limb? The values given here are not those given on page 6 for GEOVIDE surface waters, so the source should be given. 9) page 10 line 7: Change “that” to “than”

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