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

Response to referees

We would like to thank all referees for their time reading the manuscript and giving constructive suggestions to improve the paper. We are pleased that all three referees appreciate the dataset and broadly welcome publication of this paper.

Four issues were raised by more than one reviewer. Before we respond to the points raised in individual reviews, we address these four issues.

1. Disagreement between the various referees about the key nature of findings led to a change in the introduction section, and the conclusion section accordingly.

It is interesting that referee comments varied from those that said our work confirmed the use of ^{231}Pa/^{230}Th as a paleoproxy, to those that thought we have proved the proxy does not work. It is clearly important to more clearly state how the proxy might be interpreted, and whether such interpretation is justified following our work. So we have clarified in the introduction section that there are two conceptual models that form the foundation of the interpretation of sedimentary ^{231}Pa/^{230}Th ratios in terms of past rates of deep water circulation.

Model 1 relies on a net export of ^{231}Pa out of the Atlantic due to the residence time ^{231}Pa being longer than ^{230}Th (an approach adopted by studies such as McManus et al., 2014, and Bradtmiller et al., 2014).

Model 2 is based on the systematic evolution of ^{231}Pa/^{230}Th with water mass age, which has seen its application in Negre et al. (2010).

Our study provides an opportunity to assess the validity of these models. In the conclusion section, we made clear that our result supports the Model 1 interpretation that there is a northward export of ^{231}Pa out of the Atlantic, but raises questions about model 2 because there is no simple relationship between ^{231}Pa/^{230}Th and water mass age.

2. Reviewers questioned the reliability of CFC ages, especially for older waters, and asked for more details about how these ages were calculated.

We have clarified that CFC-based ages were calculated with Transit Time Distribution (TTD) method, and were different from the CFC concentration/tracer ages based on the atmospheric history of CFC.

Briefly, we computed CFC-based ages combining CFC concentrations and water mass composition obtained from extended Optimum Multi-Parameter (eOMP) analysis. First, TTD mean ages for each source water type (SWT) were calculated from CFC concentrations and eOMP analysis from OVIDE cruise 2012. These mean ages for each SWT were then combined with water mass composition obtained from eOMP analysis for GEOVIDE 2014 to give an age for water at depths where water mass compositions are available. This approach assumes that the mixing of the ages (not the CFC concentrations) is linear, and decides that the aging of water is due to variations in water mass composition, rather than the increase of spreading time of the water. In further considering and discussing this calculation we have relied heavily on input from Reiner Steinfeldt and we have therefore added him as an author to the manuscript.

3. Reviewers questioned why ^{231}Pa and ^{230}Th concentrations given by the model did not reflect quoted preformed values at zero water age.

We have considered the modelling work carefully. On reflection, we consider our introduction of a surface term to the model to be incorrect and have consequently removed it during revision, to rely on the model exactly as originally presented in Moran et al. (1997).

4. Reviewers suggested the use of SI units adopted in GEOTRACES data product.

We have changed the units in the data table and throughout the text using μBq/kg for ^{231}Pa and ^{230}Th, and pmol/kg for ^{232}Th.

Below, we respond to the referees point by point. Reviewers' comments are in blue, and our responses are in black.

Response to Anonymous Referee #2

Here Deng et al. provide a very nice and concise piece of science. They examine crucial assumptions made for the application of 231Pa/230Th as an AMOC proxy by providing an extensive new data set of water 231Pa and 230Th concentrations. This is hard won data and the authors deserve credit for their efforts. The new data extends the former GEOTRACES transects by Hayes15 and Deng14 towards the northern North Atlantic representing a definite reality-check for the assumptions made when using 231Pa/230Th as a proxy. These assumptions have been made based on the elegant approach of measuring a kinetic tracer with a constant and well-defined inputcfunction not involved in the carbon-cycle (Yu1996). While previous studies already proofed the consistence and capability of 231Pa/230Th as an AMOC proxy the novelty of this study is the systematic examination of the behaviour of 231Pa and 230Th in the northern North Atlantic in the water masses recently influenced by NADW formation with a new set of samples and by state-of-the art analytical methods. Therefore, this manuscript certainly deserves publication in Biogeosciences. Given the published results from the 231Pa/230Th proxy of the last decades I would have wondered if this study would have come to a different conclusion. But here Deng et al. make very good cases by confirming the prerequisites for using 231Pa/230Th as AMOC proxy. The only little weakness of the manuscript is the missed opportunity of setting the new findings into the context of the attempts of using 231Pa/230Th as a large scale AMOC proxy. There are several of papers dealing with a single 231Pa/230Th profile from on location, but the results of the few which deal with comprehensive compilation approaches could be better assessed and discussed here. Besides the already mentioned Yu et al. 1996, I think in particular of Bradtmiller et al. 2014, which use the large scale 231Pa deficit for analyzing the HS1 AMOC. It would be worth of shortly recapitulate their results in the light of the new results presented here. Besides the connection to observational paleo data I also miss the comparison to theoretical or model studies. The authors should in particular present a short comparison to the predictions made by Marchal et al. 2000 and the most recent attempt by Rempfer et al. 2017. Further, most of the features reported here have been anticipated by the simple box-model approach by Luo et al. 2010. They already found a very weak correlation of 231Pa/230Th with water mass age, highlighting a vertical gradient not a horizontal in the presence of an active AMOC (see specific points below).

Thank you for suggestions of references to be included. We agree, and have included them during a rewrite of the introduction.

page 3, line 2: recurring typo of “R/V Pourquoi Pas?”

Author's response:
“R/V Pourquoi Pas?” is the correct spelling for the name of the French research vessel which undertook sampling in this study.

page 3, line 15: please specify how many several months are
Author's response:

We have specified that it is four to five half-lives of ^{233}Pa ($t_{1/2}$=26.98 days, Usman and MacMahon, 2000) after spike production.

page 3, line 19: what was the analytical yield (range)of the anion chromatography for Pa and Th?

Author's response:
Analytical yield ranges from approximately 41-91% for Th, and 30-52% for Pa.

page 3, line 25: what was the 232Th/231Pa in the Pa-samples? Was the correction for the 232ThH interference necessary, if yes, how big was the contribution to the Pa-signal?

Author's response:
232Th/231Pa in the Pa samples are at about 1800. 232ThH interference contributes 0.1% of 233Pa signal. From this perspective, it does not seem necessary to correct for ThH interference in our case. However, the Th signal in the Pa sample was not known before the Pa measurement and conducted in case Th was not very well separated from Pa.

page 5, line 14: ISOW is mentioned but not shown in Fig. 5.

Author's response:
Thank you for pointing out. We have labelled ISOW in Fig.5.

Supplement: please add a column specifying the errors to the given concentrations

Author's response:
We assume that the reviewer meant Table S1 in the supplement. In this table, it is the 232Th-corrected Pa and Th concentrations that are used for the discussion. We therefore have only given errors (2se) associated with these concentrations, and do not think it is necessary to report errors for the measured concentrations without correction.

Supplement page 7 line 7: I'm aware that this is of marginal importance given the final result, but maybe the authors could elaborate on the value 0.7 in (3) and (4). 0.7 seems a little bit high for an average, or at least unnecessarily high at the high end of the possible range according to Henderson and Anderson 2003 or Bourne et al. 2012. Further, is the detrital correction required for particles slipping through the 0.45 m filters? Is the added HCl capable of leaching of the 232Th (and 230Th and 231Pa) from these particles?

Author's response:
We agree with the reviewer. Henderson and Anderson (2003) suggested average of ^{238}U/^{232}Th activity ratio to be 0.6±0.1 in the Atlantic, and 0.7±0.1 in the Pacific Ocean. We therefore have adopted the value of 0.6 in the revised paper as ^{238}U/^{232}Th activity ratio to correct for the detrital contribution of ^{231}Pa to ^{230}Th. Equation (3) and (4) were rewritten and the data were recalculated accordingly.

Detrital correction is to correct for the contribution from the partial dissolution of lithogenic minerals to ^{230}Th pool in seawater, rather than in the sample after being collected. It is therefore necessary regardless of the filtration and the acidification.

Fig. 3,8,9: error bars are missing

Author’s response:
We have added error bars to the measured data in these figures.

page 7, line 21: It is not surprising that 231Pa/230Th does not correlate with water mass age very much. This has been already predicted by Luo et al. 2010. Much more important is the vertical decrease within one circulating mater mass (e.g. Burckel et al. 2016). Thus, the sentence that “231Pa/230 ratios increase as water mass ages forms the foundation of using 231Pa/230Th in discrete cores” is not completely accurate.

Author's response:
Thank you for pointing this out. We found this comment very helpful to improve our paper. We agree with the reviewer on the relationship between ^{231}Pa/^{230}Th and water mass age. We have clarified the two conceptual models forming the foundation of the interpretation of Pa/Th in terms of rates of deep water circulation (as explained in point 1 of our opening comments above).

page 10, line 13: typo. two times “demostrates”.

Author's response:
We have corrected the typo.

Fig. 6: I assume the x-axis has changed between (a) and (b), but they are both shown on the same longitudinal scale.

Author's response:
Thank you for pointing out this error. We have corrected the longitudinal scale for Fig.6 (a) in the revised manuscript.

Fig. 3: maybe it would be worth of showing 231Pa/230Th as well in an additional panel

Author's response:
We do not think it is not necessary as we focus on discussing the latitudinal gradient of ^{231}Pa and ^{230}Th rather than ^{231}Pa/^{230}Th at this stage.

(c). Fig. 8: please indicate water depth at the colour bar.

Author's response:
We have added to the colour bar the water depth (m).

References: Bourne, M., et al., 2012. Improved determination of marine sedimentation

rates using 230Thxs. Geochemistry Geophysics Geosystems 13.
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circulation during Heinrich Stadial 1. Nature Communications 5.
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Circulation during the last glacial (20-50 ka). Climate of the Past 12.
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Luo, Y., et al., 2010. Sediment 231Pa/230Th as a recorder of the rate of the Atlantic meridional overturning
circulation: insights from a 2-D model. Ocean Science 6.
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15.
Rempfer, J., et al., 2017. New insights into cycling of 231Pa and 230Th in the Atlantic Ocean. Earth and Planetary Science Letters 468.
Yu, E., et al., 1996. Similar rates of modern and last-glacial ocean thermohaline circulation inferred from
radiochemical data. Nature 379.

Additional references (added by authors):

Bradtmiller, L. I., McManus, J. F. and Robinson, L. F.: $^{231}Pa/^{230}Th$ evidence for a weakened but persistent Atlantic meridional overturning circulation during Heinrich Stadial 1, Nat. Commun., 5, 5817 [online] Available from: http://dx.doi.org/10.1038/ncomms6817, 2014.

Negre, C., Zahn, R., Thomas, A. L., Masqué, P., Henderson, G. M., Martínez-Méndez, G., Hall, I. R. and Mas, J. L.: Reversed flow of Atlantic deep water during the Last Glacial Maximum, Nature, 468, 84 [online] Available from: http://dx.doi.org/10.1038/nature09508, 2010.

Owens, S. A., Buesseler, K. O. and Sims, K. W. W.: Re-evaluating the ^{238}U-salinity relationship in seawater: Implications for the 238U–234Th disequilibrium method, Mar. Chem., 127(1), 31–39, doi:https://doi.org/10.1016/j.marchem.2011.07.005, 2011.

Usman, K. and MacMahon, T. D.: Determination of the half-life of ^{233}Pa, Appl. Radiat. Isot., 52(3), 585–589, doi:https://doi.org/10.1016/S0969-8043(99)00214-6, 2000.