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 $^{231}\text{Pa}/^{230}\text{Th}$ 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 $^{231}\text{Pa}/^{230}\text{Th}$ ratios in terms of past rates of deep water circulation.

Model 1 relies on a net export of $^{231}\text{Pa}$ out of the Atlantic due to the residence time $^{231}\text{Pa}$ being longer than $^{230}\text{Th}$ (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 $^{231}\text{Pa}/^{230}\text{Th}$ 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 $^{231}\text{Pa}$ out of the Atlantic, but raises questions about model 2 because there is no simple relationship between $^{231}\text{Pa}/^{230}\text{Th}$ 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 GEOFVIDE 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 $\mu$Bq/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.

**Reviewer 1: R. Francois (Referee)**

This paper is an important contribution to the growing database describing the distribution of Th-230 and Pa-231 in the world ocean. I recommend publication after considering the comments and questions listed below:

Abstract; line 19-20: The reason for a weaker correlation between Pa-231 and water mass age is likely due to the much lower concentrations and rate of ingrowth of Pa compared to Th. It may be worth adding error bars to Fig. 8(a),(c),(e) to highlight this point.

Author’s response:
We agree with the reviewer that the lower concentration and slower ingrowth rate of Pa might be the reason for a weaker correlation. We have added error bars to Fig. 8(a),(c),(e), and pointed out this likely reason in the text as suggested.

P 3; line 13: why was a 236U spike added to the sample?

Author’s response:
We add the spike because we sometimes recover U during chemical separation of Th and Pa. We have made that clear in the revised manuscript.
P4: Line 20 – 23; Fig. 3: If the purpose of this figure is to show that the concentrations measured on the GEOVIDE line are much lower than measured further "downstream", I don’t think that the data from 40S are useful because they raise questions that are likely beyond the scope of this paper. The wording used in line 20 – 23 is incorrect since there is an apparent southward decrease instead of an increase between the North Atlantic and 40S. Hayes et al.'s data show a strong longitudinal gradient for both Th-230 and Pa-231. Were the averages calculated over the entire longitudinal GA03 section? Were the averages calculated for 40S comparable?

Author’s response:
We agree that 40oS data raise points outside the scope of this manuscript. We have replaced Fig 3 with a figure that compares only published data from west of the Mid-Atlantic Ridge from GA01 (i.e. this study), GA03 (Hayes) and GA02 (South Atlantic). This approach more closely follows deep-waters as they age along their flow path, and shows an increase of concentration of both nuclides.

Fig. 5: ISOW is not indicated on this figure, even though it is one of the most prominent water mass. I would suggest replacing the salinity contour lines by the contour of the main water masses reported on Fig. 4, since the purpose of this figure is to relate these water masses to the distribution of the radionuclides.

Author’s response:
We agree with the reviewer that plotting contours of the main water masses instead of salinity contours would be more useful and have modified figures accordingly.

Line 31; p5 and Fig. 6: It is not entirely clear to me how the CFC ages were determined for the GEOVIDE section. Water masses and CFC ages were determined in 2012 of the eastern side of the section. From these data, the CFC ages of the different water masses could be determined for 2012 and these ages were used to determine the CFC ages of all the samples collected in 2014 from their water mass constituents. My first problem is how you could estimate the CFC age of the water masses west of Greenland. Surely, the age of ISOW and DSOW in this section of the transect must be older than in the eastern section since these water masses are farther removed from their site of formation.

My second problem is how can you distinguish ages between 100 and 800 years with CFCs considering that CFC manufacture only started 80 years ago? Finally, and maybe more fundamentally, I question whether you can apply CFC-age to Th-230 and Pa-231. I am not an expert on this question but here are my concerns: Each (most?) of your samples consists of a mixture of different water masses. Even the red zones on Fig. 4, indicate up to 20% mixing. If you mix 2 water masses of different ages, the CFC age of the mixed sample will be biased toward the younger age because there is more CFCs in the younger end member (although CFC concentrations have quasi plateaued since 1990). On the other hand, for the same mixed sample, the Th-230 age will be biased towards the older end-member because it contains more Th-230. So, in a mixed sample, Th-230 had more time for in-growth than indicated by the CFC age. Because Pa-231 grows in slower than Th-230, this effect is less pronounced. Therefore, I believe that the ingrown Th and Pa calculated...
with CFC ages using equations 4.1 and 4.2 must be taken as minima and more so for Th than for Pa.

Author's response:
Thank you for raising the question. We have included a clear description of how CFC ages are determined, as explained as point 2 on page 1 of this Response to Reviewer (RtR). This approach allows projection of water age west of Greenland where CFC measurements were not available.

We could understand the reviewer's concern about the ISOW and DSOW ages not showing an increase in the eastern section. The approach we adopted to calculate CFC ages determines that the aging of water is due to the change in water mass composition (i.e. the mixture of water masses), rather than the increase of water spreading time.

Unlike the CFC apparent age calculated based on pCFC and the atmospheric history of CFC, the TTD approach inherently solves the mixing bias problem and is not limited by the fact that atmospheric history of CFC only started in 1930s. However, our age model does work better in the young age range between 0-80 years, and we restrict our discussion within that age range.

We agree that there exists a bias due to mixing of waters of different ages for CFC concentration age calculated based on the atmospheric history of CFC. CFC-based ages in our paper however is computed with TTD method and takes into account advection and diffusion in the mathematical framework it employs and therefore will not be biased due to mixing. As we combine TTD-derived CFC age for each SWT with water mass composition, the water mass mixing of the CFC ages is linear (unlike the CFC concentrations). This addresses the reviewer's concern that CFC ages are biased due to mixing from this perspective. There is also no bias for $^{230}$Th as the mixing is linear.

**Fig. 6: Adding water mass contour lines would be helpful**

Author's response:
As above, we have added water mass contour lines.

**Line 16; p6 “Preformed component” “..in the absence of measurement at the exact location of deep water formation: : :”.
While this statement is correct for the formation of DSOW and ISOW, I would say that station 69 is essentially at the location of LSW formation.**

Author's response:
We cannot totally agree with the reviewer that station 69 is the location of LSW formation. Pickart et al. (2003) for instance suggested that Labrador Basin is not the only Basin of LSW formation, and that Irminger Basin is a second formation site. We agree though that the wording needs some correction to make it clear that the preformed value of $^{230}$Th and $^{231}$Pa is very uncertain.
“..we set the preformed value as the average of concentrations measured in waters <100m depth for this section..”

I think the authors need to elaborate on their rationale for doing this. It is not like deepwater is formed uniquely from water sinking from a depth < 100m. Deep water convection homogenizes the water column in the Labrador and Nordic seas, which then spreads laterally at depth. Therefore, the homogenized water column at the sites of deepwater formation could be the starting point providing preformed concentrations. If so, data from station 69, as well as earlier data from Moran et al indicate that preformed Th-230 should be about 0.3 dpm/1000l both for the Labrador and Nordic seas, instead of 0.1 dpm/1000l. Such value for the Nordic Sea (based on one profile from Moran et al., 1995) is of course problematic since it is higher than measured here in the IC basin. In addition, Moran’s data from the Labrador Sea shows substantial interannual variability. I think the authors should be more nuanced in their choice of preformed values and consider how the uncertainties on this number could affect their conclusions.

Alternatively, the reasoning followed by the authors may be that cooling happens at the surface, and therefore that must be the starting point, and the higher Th concentrations found in the water below at the sites of deep water formation reflects the residence time of this surface water in the convection cell. If this is the case, that should be more explicit in the paper. I would also use the available data for surface water in the Labrador and Nordic Seas only, which are available from this study and Moran’s.

Author’s response:
1. We did not mean to give the impression that deep water is formed uniquely from water sinking from a depth <100m, nor that the values we selected for preformed values are necessarily correct. An important conclusion from our work is that the preformed values of 231Pa and 230Th make a significant and long-lasting difference to the evolution of the 231Pa/230Th ratio as waters then age. Our selection of a particular preformed values serves to illustrate this point, rather than being a statement about true preformed values. It is, of course, also possible that the various water masses have different preformed values, making the use of a single value for the whole section inappropriate if seeking a “correct” value. We have made this logic clearer during revision; that we are not implying we know the preformed values, but that whatever they are will make a substantial difference to deep-water 231Pa/230Th.

Instead, as we pointed on page 6, the preformed concentrations of dissolved 230Th and 231Pa at the time the water masses formed are unknown. We can only 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, until data are available for the Nordic Seas and the Labrador Sea during times of winter convection. The reason for the choice of < 100 m is that we consider <100 m as the surface mixed layer, where Pa and Th is well mixed.

2. This is why we discussed the effect of preformed values on the evolution of Pa and Th with water age in section 4.3. We have also added how preformed values might affect our calculation of scavenged components.
Line 30; p6: as per discussion above, calculated potential total concentration should be viewed as minima (because CFC ages underestimate the Th and Pa “ages”)

Author's response:
We hope that our opening comments above clarify that there is no age bias in our CFC ages calculated with TTD approach.

Line 4; p7: “..is the net of nuclide added from above by [add: DESORPTION FROM] settling particles: : :”

Author's response:
Changes are made as suggested.

Line 8-9; p7: Considering that the CFC ages are younger than the Th ages, potential and scavenged Th-230 must be viewed as minima. Also, the underestimation of fraction scavenged is larger for Th than Pa.

Author's response:
We hope that our opening comments above clarify that there is no age bias in our CFC ages calculated with TTD approach.

Line 14; p7: I guess the authors deduce that about 75% of the Th-230 produced is scavenged because the slope of slope of observed Th-230 vs time is 25% of the rate of production, right? If so, it would be helpful to be explicit and to report the value of the slope in Fig. 8a.

Author's response:
This is how we deduce the number. We have added the linear fit equations, giving the slope, on the figure.

Wouldn’t the intercept also provide an independent estimate of preformed? I am not sure what the intercept on scavenged Th-230 figure means.

Author's response:
We agree with the reviewer that the intercept does provide some estimate of the preformed, especially for Th with a very good linearity. However, we would expect that at t=0, there is no scavenging taking place, and the y intercept of the scavenged Th figure should be 0, while that of the observed Th figure should be preformed value. However, we have a positive y intercept in scavenged Th figure, and a negative y intercept in observed figure. We attribute this to a combination of a lack of $^{230}$Th data in younger waters and the uncertainty associated with $^{230}$Th measurement and calculation.

I would also reiterate here that 75% scavenging is a minimum, and it could be that scavenging is more underestimated in older water than younger ones, producing this apparent intercept. It is not clear to me why the same can’t be attempted for Pa, at least for the observed values.
Author's response:
As above, we do not think there is an age bias in our CFC ages based on TTD approach. We therefore do not think a change is required here.

Line 15-14; p7: “This ratio is consistent with the average 230Th for these waters, which requires that about three times more 230Th has been removed by scavenging”
Line 18; p7:” : : :the average [Pa] values indicate that about half of the 231Pa remains in the water”
I can’t follow the line of reasoning here. I think the authors need to be more explicit.

Author's response:
From Fig. 8 (a) and (c), we have:
Average of Th in these waters= 3.24 µBq/kg
and average of Pa in these waters= 1.51 µBq/kg
From Fig. 8 (b) and (d), we have:
Average of scavenged Th = 11.8
Average of scavenged Pa = 1.02

To maintain an average Th of 3.3 µBq/kg in water requires 12.1 µBq/kg to be scavenged, which is three times more Th to be removed by scavenging.
Similarly, to maintain an average of Pa 1.5 µBq/kg in water requires 1.2 µBq/kg to be scavenged. That is, half of the 231Pa remains in water and half removed by scavenging.

P2; line 23: “.. there is no simple relationship between increasing Pa/Th and age, as would be expected”
P7; Line 21; p7:”..The hypothesis that Pa/Th ratios increase as water mass ages…”

We should not expect seawater Pa/Th to simply increase with water mass age. If dissolved Th and Pa activities are initially low, Pa/Th in the water column (and underlying sediments) should initially decrease with age of water mass because Th grows in faster than Pa. It’s only once Th has regained levels closer to equilibrium with scavenging (and therefore grows more slowly) that Pa starts growing faster, resulting in increasing Pa/Th. This effect is clearly illustrated in the paper by Luo et al. (2010) (see Fig. 14 in the paper). Fig. 8e,f may show a hint of this initial decreasing trend. (Luo et al also argued “Clearly, it is impossible to constrain the history of changes in the AMOC from the evolution of 231Pa/230Th at one site, as was attempted by McManus et al.; p 395; last sentence of first paragraph).

Author's response:
We agree with the reviewer that we should not expect seawater Pa/Th to simply increase with water mass age. The model results plotted on Fig. 8e,f indicate an expectation that Pa/Th ratios can either increase or decrease in the first 10 to 15 years, depending on the preformed values. Beyond this age, Pa/Th ratios increase with age before reaching equilibrium. We have made this clearer in the manuscript accordingly.

Section 4.3: I am puzzled by Fig 9. The intercepts don’t seem to match with the
preformed values. The fit with the data is terrible. I am not sure what this section is telling us.

Author's response:
As in our opening comment 3: We thank the reviewer for pointing out this issue, which contributed to our reconsideration of the model and removal of the incorrect surface term. This change gives Y intercept matching the preformed values at t=0.

We are aware of the ‘terrible’ fit of the model with data, even in the new model with surface term removed. It however supports what we aim to convey in this section that preformed values can influence the evolution of Pa, Th and Pa/Th with water mass age. Without a better knowledge of preformed values, we cannot give a simple description of how Pa/Th evolve with water mass age, and the model cannot generate profiles that better fit observations.

Line 28; p7: wouldn’t it be better to follow density rather than depth?

Author's response:
We followed depth as the model is a function of depth, reflecting that the scavenging of $^{230}\text{Th}$ and $^{231}\text{Pa}$ takes place with particles sinking with depth instead of density. This is also the way that $^{230}\text{Th}$ and $^{231}\text{Pa}$ data have traditionally been considered.

Line 4; p8: why did you choose station 1 and 13? Is it because they are the least ventilated and therefore these parameters could be estimated from a linear fit? If this is the case, that should be indicated. However, is it reasonable to apply scavenging parameters from a margin to open ocean stations?

Author's response:
We chose station 1 and 13 as their profiles are the closest to the equilibrium profile with linear increase with water depth. We however realized, as the reviewer pointed out, that Station 1 is close to the margin, on the continental slope, and therefore used station 13 only, an open ocean station, to optimize model parameters.

Fig. 9: How can Pa/Th (zero preformed) increase with age (c) if Th (a) grows faster than Pa (b)?

Author's response:
With zero preformed Pa/Th is expected to increase from first ventilation. Despite the fact that Pa approaches an equilibrium value more slowly than Th, the Pa concentration increases more than Th concentration, i.e., more of the Th produced by decay is removed by scavenging than for Pa, at each time step. So the Pa/Th ratio increases monotonically from T=0.

Section 4.4:
This discussion should take into account that scavenged Th and Pa are underestimated and proportionally more for Th than Pa. Since potential and scavenged are underestimated by exactly the same absolute amount, calculated scavenged/potential ratios should also be taken as minima.
Author’s response:
As above, we do not think there is underestimation of scavenged Th and Pa due to bias in CFC ages and Th ages as a result of mixing.

P 9; line 20-22; I think it would be helpful for the reader to provide the details of this mass balance calculation. For instance, what volume for the North Atlantic did you use to calculate production?

Author’s response:
As we followed the Deng et al. (2014) for the calculation and used the number for the volume of the North Atlantic in that paper, which is referred to in the discussion manuscript, we did not give details here. However, we understand the reviewer’s point that it would be helpful for readers so we now repeat this information in the revised supplementary material.

It seems that the results from this mass balance corroborate that scavenged (and therefore potential) Th and Pa calculated from equation 4.3 are underestimated because of the use of CFC ages. I am a little surprised that so much Pa is scavenged in the North Atlantic. If 77% of the production is scavenged, then the average Pa/Th of N. Atlantic sediments should be ca.0.073, which is higher than measured in the central N. Atlantic basin. That means that boundary scavenging must be more significant than I thought.

Author’s response:
The 96% scavenged relative to produced for Th and 77% for Pa obtained from the mass balance calculation represents a basin-scale average in the North Atlantic. The lower scavenged to produced values from equation 4.3. represent more localized results when water masses are young, and nuclide concentrations and scavenging correspondingly low.

Roger Francois
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 input function 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 $^{233}\text{Pa}$ ($t_{1/2}=26.98$ 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 $^{232}\text{Th}/^{231}\text{Pa}$ in the Pa-samples? Was the correction for the $^{232}\text{ThH}$ interference necessary, if yes, how big was the contribution to the Pa-signal?

Author's response:
$^{232}\text{Th}/^{231}\text{Pa}$ in the Pa samples are at about 1800. $^{232}\text{ThH}$ interference contributes 0.1% of $^{233}\text{Pa}$ 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 $^{232}\text{Th}$-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 μm filters? Is the added HCl capable of leaching of the $^{232}\text{Th}$ (and $^{230}\text{Th}$ and $^{231}\text{Pa}$) from these particles?

Author’s response:
We agree with the reviewer. Henderson and Anderson (2003) suggested average of $^{238}/^{232}$ 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 $^{238}\text{U}/^{232}\text{Th}$ activity ratio to correct for the detrital contribution of $^{231}\text{Pa}$ to $^{230}\text{Th}$. 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 $^{230}\text{Th}$ 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 $^{231}\text{Pa}/^{230}\text{Th}$ 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 “$^{231}\text{Pa}/^{230}\text{Th}$ ratios increase as water mass ages forms the foundation of using $^{231}\text{Pa}/^{230}\text{Th}$ 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 $^{231}\text{Pa}/^{230}\text{Th}$ 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 $^{231}\text{Pa}/^{230}\text{Th}$ 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 $^{231}\text{Pa}$ and $^{230}\text{Th}$ rather than $^{231}\text{Pa}/^{230}\text{Th}$ 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
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:

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 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 40S 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 40S. 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.

Author’s response:
We have taken on board the reviewer’s comments here. As explained in point 1 of our opening comments above, we have now clarified that there are two conceptual models that form the foundation of the interpretation of 231Pa/230Th. This has significantly helped us to make clear the take-home messages in the conclusion section.
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 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.

Author's response:

We have corrected the missing longitude mistake.

Thank you for suggesting clarification for the calculation of CFC-based ages. We have included more detailed information describing how CFC-based ages were calculated and uncertainties associated with it, as explained as point 2 on page 1 in this RtR.

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.

Author's response:

Thank you for the suggestion. We have included the uncertainty of the CFC ages and how they affect our interpretation as supplemental information in the revised manuscript.

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.

Author's response:
Thank you for this comment. We have added the suggested sensitivity analysis in the Supplemental Information.

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?

Author's response:
Thank you for your comment. This is a good point, and we have made clear that our results are based on the assumption of the same preformed value for different water masses, and addressed the influence of this assumption based on our discussion of how preformed values affect the results.

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?

Author's response:
Thank you for pointing this out. We ran some tests with the model and realized that introducing the surface term in the model is the reason causing the 231Pa and 230Th concentration twice of the preformed values at zero age. Considering this weakness in the model, we removed the surface term in the model as explained as point 3 of our opening comments. The new model gives y intercepts at preformed values at t=0.

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.

Author's response:
We agree that it would be great to integrate all the $^{231}\text{Pa}$ and $^{230}\text{Th}$ data available from GEOTRACES in the Atlantic Ocean. We are thinking of putting together a paper with that aim. With the present manuscript paper, however, we would prefer to focus on the data from GEOVIDE and how this new addition of data can provide evidence to assess the conceptual models adopted for the interpretation of $^{231}\text{Pa}/^{230}\text{Th}$. However, we continue to include data from GA02 and GA03 to put our new GEOVIDE data in the broader context and to show horizontal gradient of $^{231}\text{Pa}$ and $^{230}\text{Th}$ in the Atlantic.

Minor comments:

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?

Author’s response:
We have reported our data in SI units adopted in the GEOTRACES data product as suggested.

page 6 line 1 “Steinfeldt” is misspelled.

Author’s response:
We have corrected the misspelling.

page 6, definition of “Ingrown component”: Are U concentrations normalized to a constant salinity? To the salinity measured for each sample? Something else?

Author’s response:
We used constant U activities for all the sample when calculating the “ingrown component”, and calculated $^{238}\text{U}$ following the equation in Owens et al., 2017, assuming a salinity of 35 permil. U-235 and U-234 activity was then calculated assuming natural abundance ratio of $^{238}\text{U}/^{235}\text{U}$=137.88 and the $^{234}\text{U}/^{238}\text{U}$ activity ratio in seawater is ~1.15.

page 7 line 16: “three times more 230Th has been removed by scavenging” than “what?” Complete the description of the comparison being made.

Author’s response:
We have added: ‘… more than it remains in the water…’

page 8 line 7: Change “run” to “ran”

Author’s response:
We have made changes as suggested.

page 8 line 27: delete the “in” prior to “from DSOW”

Author’s response:
We have made changes as suggested.

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.

Author's response:
We have included the suggested reference.

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.

Author's response:
Here, the boundary between upper and lower was defined by potential density at 32.15 kg/m$^3$ as described in the discussion manuscript on page 9 line 10. The average 230Th and 231Pa in the upper and lower limb are average concentrations of these nuclides in water with potential density < 32.15 kg/m$^3$ and > 32.15 kg/m$^3$, respectively.

page 10 line 7: Change “that” to “than” i

Author's response:
We have made changes as suggested.

Additional references (added by authors):


Evolution of $^{231}$Pa and $^{230}$Th in overflow waters of the North Atlantic

Feifei Deng1, Gideon M Henderson1, Maxi Castrillejo2,3, and Fiz F. Perez4 and Reiner Steinfeldt5

1Department of Earth Sciences, University of Oxford, South Parks Road, Oxford, OX13AN, UK.
2Laboratory of Ion Beam Physics, ETH-Zurich, Otto Stern Weg 5, Zurich, 8093, Switzerland
3Institut de Ciència i Tecnologia Ambientals & Departament de Física, Universitat Autònoma de Barcelona, Bellaterra, 08193, Spain
4Departamento de Oceanografía Instituto Investigaciones Marinas (CSIC), Eduardo Cabello 6, E36208 Vigo, Spain.
5Institut für Umweltphysik, Universität Bremen, D-28334 Bremen, Germany.

Correspondence to: Feifei Deng (feifei.deng@earth.ox.ac.uk)

Abstract. Many paleoceanographic studies have sought to use the $^{231}$Pa/$^{230}$Th ratio as a proxy for deep ocean circulation rates in the North Atlantic. As yet, however, no study has fully assessed the concentration of, or controls on, $^{230}$Th and $^{231}$Pa in waters immediately following ventilation at the start of Atlantic meridional overturning. To that end, full water-column $^{231}$Pa and $^{230}$Th concentrations were measured along the GEOVIDE section, sampling a range of young North Atlantic deep waters. $^{230}$Th and $^{231}$Pa concentrations in the water column are lower than those observed further south in the Atlantic, ranging between 0.06 and 12.01 µBq/kg, and between 0.37 and 4.80 µBq/kg, respectively. Both $^{230}$Th and $^{231}$Pa profiles generally increase with water depth from surface to deep water, followed by decrease near the seafloor, with this feature most pronounced in the Labrador Sea (LA Sea) and Irminger Sea (IR Sea). Analyzing this dataset with Extended Optimum Multi-Parameter (eOMP) Analysis and CFC-based water mass age indicates that the low values of $^{230}$Th and $^{231}$Pa in water near the seafloor of the LA Sea and IR Sea are related to the young waters present in those regions. This importance of water age is confirmed for $^{230}$Th by a strong correlation between $^{230}$Th and water mass age (though this relationship with age is less clear for $^{231}$Pa and $^{231}$Pa/$^{230}$Th ratio). Scavenged $^{231}$Pa and $^{230}$Th were estimated and compared to their potential concentrations in the water column due to ingrowth. This calculation indicates that more $^{230}$Th is scavenged (~80%) relative to $^{231}$Pa (~40%), consistent with the relatively higher particle-reactivity of $^{230}$Th. Enhanced scavenging for both nuclides is demonstrated near the seafloor in young overflow waters. Calculation of meridional transport of $^{230}$Th and $^{231}$Pa with this new GEOVIDE dataset enables a complete budget for $^{230}$Th and $^{231}$Pa for the North Atlantic. Results suggest that net transport southward of $^{230}$Th and $^{231}$Pa across GEOVIDE is smaller than transport further south in the Atlantic, and indicates that the flux to sediment in the North Atlantic is equivalent to 96% of the production of $^{230}$Th, and 74% of the production for $^{231}$Pa. This result confirms a significantly higher advective loss of $^{231}$Pa to the south relative to $^{230}$Th and supports the use of $^{231}$Pa/$^{230}$Th to assess meridional transport at a basin scale.

Key words. GEOTRACES; water-column $^{230}$Th and $^{231}$Pa; water mass ageing; scavenging; meridional transport.
1 Introduction

Several paleoceanographic proxies have been proposed that rely on the $^{231}\text{Pa}^{230}\text{Th}$ ratio in marine sediments, one of which is that $^{231}\text{Pa}^{230}\text{Th}$ may record the rate of deep-water circulation, particularly in the North Atlantic. Both $^{231}\text{Pa}$ and $^{230}\text{Th}$ are produced in seawater at a constant rate by decay of uranium, but have decay activities much lower than their parent uranium isotopes due to rapid removal by adsorption onto sinking marine particles. Both nuclides are also reversibly scavenged, leading to particularly low concentrations at the surface and increasing concentrations with depth (Nozaki et al., 1981). Advection of surface waters to depth transports water with low concentrations of $^{231}\text{Pa}$ and $^{230}\text{Th}$ into the deep ocean, where their concentrations subsequently increase towards an equilibrium value at a rate dependant on the residence time of the nuclide. The longer residence time of $^{231}\text{Pa}$ relative to $^{230}\text{Th}$ (~130 years versus ~20 years, Henderson and Anderson, 2003) means that the equilibrium concentration of $^{231}\text{Pa}$ is closer to that expected from uranium decay, and that the time taken to reach this equilibrium is longer.

This oceanic behaviour of $^{231}\text{Pa}$ and $^{230}\text{Th}$ suggests that their measurement in marine sediments may reveal information about the past environment, with one common use being as a recorder of deep-water circulation, particularly in the North Atlantic (e.g. Gherardi et al., 2005, 2009; McManus et al., 2004; Roberts et al., 2014; Yu et al., 1996). The interpretation of sedimentary $^{231}\text{Pa}^{230}\text{Th}$ ratios for such past ocean circulation is based on two end-member conceptual models:

- **Basin-scale Advection**: The longer residence time of $^{231}\text{Pa}$ than $^{230}\text{Th}$ means that deep-water contains more $^{231}\text{Pa}$ than $^{230}\text{Th}$ relative to production from decay. Advection of deep-waters out of the North Atlantic therefore removes more $^{231}\text{Pa}$ than $^{230}\text{Th}$, leaving sediments in the basin with a $^{231}\text{Pa}^{230}\text{Th}$ ratio below the production ratio. If deep-water ventilation ceased, $^{231}\text{Pa}$ removal from the North Atlantic also ceases, and sedimentary $^{231}\text{Pa}^{230}\text{Th}$ values reach their production ratio. This approach was first proposed by Yu et al. (1996) who measured $^{231}\text{Pa}^{230}\text{Th}$ in Holocene and Last Glacial Maximum (LGM) sediments from many core-top samples from the Atlantic and Southern Ocean. They found similar Holocene and LGM values at a basin scale, suggesting broadly similar overturning during the two periods. Subsequent application to sediments from Heinrich Stadial 1, initially in a single core (McManus et al., 2004) and progressively a geographical range of cores (Bradtmiller et al., 2014), revealed reduced advection of $^{231}\text{Pa}$ out of the basin at that time, suggesting decreased overturning.

- **Water-mass Evolution**: The longer residence time of $^{231}\text{Pa}$ means that, following ventilation, it takes longer for deep-water $^{231}\text{Pa}$ concentrations to reach equilibrium with respect to scavenging than is the case for $^{230}\text{Th}$. This leads to a systematic evolution of $^{231}\text{Pa}^{230}\text{Th}$ with age of the water. Sediments capture this ratio (with a fractionation due to different scavenging coefficients for the two nuclides), so capture information about the age of the water. Simple models suggest an increase of $^{231}\text{Pa}^{230}\text{Th}$ with age over about 400 years (e.g. several residence times of $^{231}\text{Pa}$). This approach to interpreting sedimentary $^{231}\text{Pa}^{230}\text{Th}$ allows for the possibility of calculating flow rates for a single water mass and from a single core rather than at a basin scale. It has been pursued by (Negre et al., 2010) to assess deep-water flow in both southerly and northerly directions by comparing sediments in the North and South Atlantic, and allowed these authors to apply a simple model (Thomas et al., 2007) to calculate flow rates.
Recent water-column measurements of $^{231}$Pa and $^{230}$Th on GEOTRACES cruises shed new light on the chemical behaviour and controls on these isotopes in seawater and provided evidence to assess the validity of the models underlying the use of sedimentary $^{213}$Pa-$^{230}$Th as a proxy for deep-water circulation. These measurements have indicated that there is considerably more net advection of $^{231}$Pa than $^{230}$Th out of the North Atlantic (Deng et al., 2014), supporting the Basin-scale Advection model for $^{231}$Pa-$^{230}$Th. But these measurements also suggest that there is no simple relationship between increasing $^{231}$Pa/$^{230}$Th and age of water, as would be expected for the Water-mass Evolution model (e.g. Deng et al., 2014). Studies using 2-D and 3-D ocean models (e.g. Marchal et al., 2010; Siddall et al., 2007) have also supported the use of sedimentary $^{231}$Pa-$^{230}$Th to constrain deep-water circulation at a basin scale, and suggested that the relationship between $^{231}$Pa/$^{230}$Th and water mass age is more complex than assumed in earlier studies (e.g. Luo et al., 2010).

Observations and model studies of $^{231}$Pa and $^{230}$Th have also suggested that other controls complicate $^{231}$Pa-$^{230}$Th as a dynamic tracer of deep-water circulation, such as the effect of boundary scavenging at seafloor and ocean margins (e.g. Anderson et al., 1994; Deng et al., 2014; Rempfer et al., 2017) and the influence of particle flux and composition (e.g. Chase et al., 2002; Hayes et al., 2014; Siddall et al., 2005).

To fully assess the behaviour of $^{231}$Pa-$^{230}$Th, and its potential as a dynamic tracer of deep-water circulation, knowledge of the concentrations and variations of these isotopes as deep waters form and enter the deep Atlantic is required. Some measurements have placed initial constraints on $^{231}$Pa and $^{230}$Th values in young North Atlantic deep waters (e.g. Moran et al., 1997, 2002; Rutgers van der Loeff and Berger, 1993), but there has not yet been a systematic study of the composition of waters in the far north Atlantic. The GEOVIDE cruise allowed waters to be collected for such a study, along a line where significant other data are available, both from that cruise and previous occupations of OVIDE. GEOVIDE provided an ideal opportunity to understand $^{231}$Pa and $^{230}$Th at the start of the ocean meridional overturning circulation, and to assess the hypotheses underlying the use of $^{231}$Pa-$^{230}$Th as a paleo-proxy for the rate of deep-water circulation.

**2 Sampling strategies and Analytical methods**

Seawater samples were collected during the GEOVIDE cruise aboard the R/V Pourquoi Pas? from 15 May to 30 June, 2014 as part of GEOTRACES Section GA01. The cruise sampled four regions in the North Atlantic between 40°-60°N: Labrador Sea (LA Sea), Irminger Sea (IR Sea), Iceland Basin (IC Basin), and Western European Basin (WE Basin) (Fig. 1).

Full-depth water-column $^{231}$Pa and $^{230}$Th for this study were collected from 11 stations (Fig. 1). Sampling followed the procedure suggested by GEOTRACES intercalibration work (Anderson et al., 2012). Briefly, seawater samples of 5 Litres were directly filtered from Niskin bottles mounted on the Stainless Steel CTD Rosette through AcroPak™ capsules with Supor® Membrane (0.45 µm pore size). Filtered seawater samples were collected into acid cleaned HDPE plastic bottles, and sealed with a screw cap and Parafilm to reduce evaporation and contamination. Samples were then double bagged for storage in boxes for transport back to the shore-based lab for analysis.
Once returned to the laboratory in Oxford, samples were weighed and then acidified with quartz distilled concentrated HCl to pH ~1.7, shaken and left for at least four days to ensure that Pa and Th was desorbed from the walls of the bottle. A mixed $^{230}$Th-$^{234}$U spike and a $^{231}$Pa spike were then added to each sample to allow measurement of Th, U (for another study), and Pa by isotope dilution MC-ICP-MS (Multi Collector-Inductively Coupled Plasma-Mass Spectrometry). The $^{233}$Pa spike was freshly made by milking from $^{235}$Np (following Regelous et al., 2004) and calibrated against a known $^{234}$U solution after complete decay of $^{231}$Pa to $^{231}$U, i.e. four to five half-lives of $^{231}$Pa ($t_{1/2}$=26.98 days, Usman and MacMahon, 2000) after spike production (Robinson et al., 2004). 50 mg of pure Fe as a chloride solution was also added to each water sample. Samples were left overnight to allow for spike equilibrium after which the pH was raised to ~8.5 using distilled NH$_4$OH to co-precipitate the actinides with insoluble Fe-oxyhydroxides. At least 48 hours were allowed for scavenging of the actinides onto Fe-oxyhydroxides. The precipitate was centrifuged and rinsed, and Th, Pa and U were separated using anion exchange chromatography following Thomas et al., 2006.

After chemical separation, Pa and Th were measured on a Nu instrument MC-ICP-MS at the University of Oxford. Mass discrimination and ion-counter gain were assessed with the measurement of a U standard, CRM-145 U, before each sample measurement. Use of a U standard for this purpose minimises memory problems that might be caused by use of a Th or Pa standard (Thomas et al., 2006). Measurements were also made 0.5 mass units either side of masses of interest to allow accurate correction for the effect of abundance sensitivity on small $^{231}$Pa and $^{230}$Th beams, and a correction for a small $^{232}$ThH interference on the $^{233}$Pa beam is made from assessment of the hydride formation rate on a $^{232}$Th standard. Concentrations of $^{231}$Pa, $^{230}$Th together with $^{232}$Th were obtained from the precise MC-ICP-MS measurement of $^{231}$Pa, $^{236}$Th/$^{232}$Th, and $^{232}$Th/$^{229}$Th ratios together with well-calibrated concentrations of $^{231}$Pa, and $^{229}$Th-$^{234}$U spikes.

Chemistry blanks were assessed by conducting the complete chemical procedure on ~100 ml of Milli-Q water with each batch of samples. Based on six blank measurements, the average blanks for dissolved $^{231}$Pa, $^{230}$Th and $^{232}$Th are 0.21±0.14 fg, 1.59±0.60 fg and 5.13±1.47 pg, respectively (uncertainties are 2 standard errors). Blank contributions account for 2-22%, 2-26%, and 0.2-16% of the dissolved $^{231}$Pa, $^{230}$Th and $^{232}$Th respectively (with the higher values being for surface samples due to their low concentrations).

### 3 Results

Measured $^{230}$Th and $^{231}$Pa concentrations were corrected for blanks, ingrowth from U in seawater since the time of sample collection, and detrital U-supported $^{230}$Th and $^{231}$Pa concentrations. Measured and corrected concentrations of $^{230}$Th, $^{231}$Pa, and $^{232}$Th, along with details of corrections, are provided in the Supplemental Information SI. Although analysis was conducted in terms of fg/kg, results are converted to the SI units adopted by GEOTRACES data product, i.e., µBq/kg for $^{235}$Th and $^{231}$Pa, and pmol/kg for $^{232}$Th. This conversion uses half-lives for $^{231}$Pa, $^{230}$Th and $^{232}$Th of 32.760 yr, 75.584 yr and 1.405×10$^{10}$ yr, respectively (Robert et al., 1969; Cheng et al., 2013; Holden, 1990). Uncertainties were propagated, including the contribution from sample weighing, spike calibration, impurities in the spikes, blank corrections, and mass spectrometric measurement.
and are reported as 2 standard errors (2 s.e.). Average total uncertainties for \(^{231}\text{Pa}, \(^{230}\text{Th}\) and \(^{232}\text{Th}\) are \(\pm0.17\ \mu\text{Bq/kg}, \pm0.17\ \mu\text{Bq/kg}, \) and \(\pm0.0032\ \text{pmol/kg},\) respectively. Vertical profiles showing the results of corrected \(^{230}\text{Th}\) and \(^{231}\text{Pa}\) concentrations in the water column are plotted by region in Fig. 2.

Th-230 concentrations in the water column range between 0.06 and 12.01 \(\mu\text{Bq/kg}\), and initially generally increase with water depth from surface to deep water. Towards the seafloor, six of the eleven stations show a prominent decrease of \(^{230}\text{Th}\), with this feature most pronounced in the LA and IR Seas.

Pa-231 concentrations in the water column range between 0.37 and 4.80 \(\mu\text{Bq/kg}\) and also increase with water depth, but less rapidly than \(^{230}\text{Th}\). \(^{231}\text{Pa}\) profiles also often exhibit a decrease near the seafloor at stations showing a \(^{230}\text{Th}\) decrease. Station 38 at the Reykjanes Ridge distinguishes itself from other \(^{231}\text{Pa}\) profiles in that an increase in \(^{231}\text{Pa}\) concentrations from low concentrations at 1000 m is observed, continuing towards the bottom.

Observed \(^{230}\text{Th}\) and \(^{231}\text{Pa}\) values at GEOVIDE are lower than those observed in inter-calibrated GEOTRACES data from further south in the Atlantic. Figure 3 compares average depth profiles for \(^{230}\text{Th}\) and \(^{231}\text{Pa}\) in the west Atlantic, covering high-latitude Northwest Atlantic (from GEOVIDE, west of the Mid-Atlantic Ridge), mid-latitude Northwest Atlantic (GEOTRACES section GA03_w, Hayes et al., 2015) and Southwest Atlantic (GEOTRACES section GA02, Deng et al., 2014).

A southward increase of both \(^{230}\text{Th}\) and \(^{231}\text{Pa}\) concentrations is observed below 1000 m.

### 4 Discussion

Early studies of water-column \(^{230}\text{Th}\) and \(^{231}\text{Pa}\) reported a linear increase of both nuclides with water depth (e.g., Anderson et al., 1983b; Nozaki et al., 1981), and introduced a reversible scavenging model with exchange of both nuclides between their dissolved and particulate phases. Later studies observed a deviation of \(^{230}\text{Th}\) and \(^{231}\text{Pa}\) profiles from this reversible scavenging model, with the expected increase with depth often inverting near the seafloor (e.g., Anderson et al., 1983a; Bacon and Anderson, 1982). This feature has been further investigated in more recent studies. Rutgers van der Loeff and Berger (1993) observed that \(^{230}\text{Th}\) concentrations decrease in the bottom water in the South Atlantic south of the Antarctic Polar Front and interpreted this as the influence of relatively young bottom water in the region. Okubo et al. (2012) also found decreasing \(^{230}\text{Th}\) values near the seafloor in the North Pacific and, in the absence of ventilation in the area, interpreted these as due to bottom scavenging. Deng et al. (2014) observed low concentrations of both \(^{230}\text{Th}\) and \(^{231}\text{Pa}\) in near-bottom water coinciding with the presence of the nepheloid layer, and interpreted the low values as a result of enhanced scavenging by resuspended particles in the nepheloid layer.

In this study, recently ventilated overflow waters are sampled at depth, particularly in the Labrador and Irminger Seas. Low values of \(^{230}\text{Th}\) and \(^{231}\text{Pa}\) near the seafloor might be expected to relate to these young waters, but the effects of scavenging must also be considered.

### 4.1 Water mass distribution and influence
The presence of multiple water masses sampled by the GEOVIDE Section allows the influence of water mass (and age) on\textsuperscript{230}Th and \textsuperscript{231}Pa to be assessed. Extended Optimum Multi-Parameter (eOMP) Analysis (García-Ibáñez et al., 2018) for the GEOVIDE section maps the presence of 10 water-mass end-members in the section (Fig. 4), including three recently ventilated waters in the GEOVIDE section.

i. Labrador Sea Water (LSW), which is formed by deep convection (Talley and McCartney, 1982), is the dominant deep water along the section, extending from 1000 to 2500 m depth in the east and from surface to 3500 m in the west of the section.

ii. Iceland–Scotland Overflow Water (ISOW), which is formed in the Norwegian Sea and subsequently entrains overlying warmer and more salty waters. This water mass initially flows along the eastern flank of the Reykjanes Ridge before spreading back northwards, after crossing the Charlie-Gibbs Fracture Zone, into the Irminger and Labrador Seas (Dickson and Brown, 1994; Saunders, 2001). A pronounced layer of this water mass is observed immediately below the LSW, and extends as deep as 4000 m west of 20°W.

iii. Denmark Strait Overflow Water (DSOW), which is formed after the Nordic Seas deep waters overflow and entrains Atlantic waters (SPMW and LSW) (Yashayaev and Dickson, 2008) with dense Greenland shelf water cascading down to the DSOW layer in the Irminger Sea (Falina et al., 2012; Olsson et al., 2005; Tanhua et al., 2005). This water occupies the deepest part of the IR and LA Seas.

In the east of the section, deep waters consist of the much older Lower North East Atlantic Deep Water (NEADW\textsubscript{1}) which is formed with a significant southern component from the Antarctic Bottom Water. A number of other water masses are also observed at shallow depths, including Mediterranean Water, and various mode waters.

Some control of water mass on \textsuperscript{230}Th and \textsuperscript{231}Pa concentration is evident in nuclide section plots (Fig. 5), particularly relatively low \textsuperscript{230}Th and \textsuperscript{231}Pa concentrations in DSOW and high values in the old NEADW. In other places, the impact of water mass is less apparent. The challenge with these nuclides is that they are not conservative tracers of water mass, but evolve significantly during transport and water aging. On the GEOVIDE Section we can analyse this evolution because the ages of the water-masses can be assessed from CFC data.

CFC measurements are not available from the GEOVIDE cruise itself. de la Paz et al. (2017), however, measured CFC concentrations along the west of the same section (covering WE Basin, IC Basin, and IR Sea) in 2012 (OVIDE/CATARINA cruise). This allowed the computing of CFC-based age with the Transit Time Distribution (TTD) method. Using the water mass distribution along GEOVIDE given by García-Ibáñez et al. (2018) and the distribution for the same water masses in 2012 (García-Ibáñez et al., 2015), we derived CFC-based ages for GEOVIDE waters (Fig. 6; further details in Supplemental Information S2). Uncertainties (1 standard error) associated with CFC-based age calculated with this approach range between 11-40%.

CFC-based water-mass ages range from \(\approx\)10 years, observed in DSOW at the bottom of the LA Sea, to \(\approx\)800 years, observed for NEADW at the bottom of the WE Basin. Because this study focuses on understanding controls on \textsuperscript{231}Pa and \textsuperscript{230}Th in recently ventilated waters, we omit detailed consideration of the upper 1km in subsequent discussion, and restrict our analysis to water sampled west of 35°W of the section where young waters (\(\approx\)50 years) dominate. A rescaled version of the CFC age...
section indicates the variation in age of ventilated waters (Fig. 6b). DSOW, occupying the deepest LA and IR Seas, is the youngest water mass in this region, with an average age of ~19 years. ISOW and LSW are slightly older, with ages ranging from 26 to 45 years and 32 to 40 years respectively.

### 4.2 Evolution of $^{230}$Th and $^{231}$Pa with water age

The presence of recently ventilated deep-waters with constrained CFC ages allows analysis of the rates at which $^{230}$Th and $^{231}$Pa concentrations increase during transport, and the rates of scavenging of these nuclides. To conduct this analysis, we define five components in the budget of $^{230}$Th and $^{231}$Pa:

- **i. Preformed component:** The $^{230}$Th or $^{231}$Pa transported from the surface into the interior. For this analysis, in the absence of measurements for the exact location of deep-water formation during winter convection, we assume the same preformed value for all water masses and set this as the average of concentrations measured in surface waters <100 m depth along GEOVIDE section. This gives preformed concentrations of $1.66 \mu$Bq/kg for $^{230}$Th and $1.31 \mu$Bq/kg for $^{231}$Pa. We recognise that true preformed values may differ from these values and between water masses, and discuss the implications of uncertainty in preformed values in the following section.

- **ii. Ingrown component:** The ingrown $^{230}$Th or $^{231}$Pa from radioactive decay of U since the water was last in contact with the surface. This component increases as the water mass ages. The concentration of this component in a water mass of age $t$ can be calculated as:

$$^{230}Th_{\text{ingrown}} = ^{230}U \times (1 - e^{-\lambda_{230}t}) \quad \text{(4.1)}$$

$$^{231}Pa_{\text{ingrown}} = ^{231}U \times (1 - e^{-\lambda_{231}t}) \quad \text{(4.2)}$$

where $^{230}Th_{\text{ingrown}}$ and $^{231}Pa_{\text{ingrown}}$ are the $^{230}$Th and $^{231}$Pa ingrown from their U parents, respectively; $^{230}U$ and $^{231}U$ are activities of $^{234}U$, and $^{234}U$ in seawater (45551.2 $\mu$Bq/kg (2801.4 dpm/1000) and 1823.8 $\mu$Bq/kg (112.2 dpm/1000), respectively, assuming a constant seawater $^{234}U$ activity of 39609.8 $\mu$Bq/kg (2436 dpm/1000) at salinity 35 psu, and seawater $^{234}U/^{238}U$ activity ratio of 1.15 and natural $^{238}/^{235}U$ abundance ratio of 137.88); $\lambda_{230}$ and $\lambda_{231}$ are decay constants of $^{230}$Th and $^{231}$Pa (9.17 x 10^{-6} yr^{-1} and 2.12 x 10^{-5} yr^{-1}, respectively).

- **iii. Potential Total component:** The $^{230}$Th and $^{231}$Pa expected in the water due to the combination of preformed and ingrown components, if there were no removal by scavenging.

- **iv. Observed component:** The $^{230}$Th and $^{231}$Pa observed in the water column, i.e. dissolved $^{230}$Th and $^{231}$Pa measured in this study (after correction for detritus and ingrowth from U since sample collection).
v. Scavenged component: The net $^{230}$Th or $^{231}$Pa removed from the water since it left the surface due to scavenging. For each depth, this component is the net of nuclide added from above by desorption from settling particles, and the removal downwards by scavenging. These components are related to each other as follows:

\[ \text{Preformed} + \text{Ingrown} = \text{Potential Total} \times \frac{^{230}\text{Th}}{\text{or} \quad ^{231}\text{Pa}} = \text{Observed} + \text{Scavenged} \quad (4.3) \]

The difference between the Potential Total and the Observed concentration of $^{230}$Th (or $^{231}$Pa), therefore provides a measure of the amount of nuclide scavenged since the water left the surface (Fig. 7).

We examine the evolution of both the observed and scavenged components of $^{230}$Th and $^{231}$Pa with water mass age (Fig. 8). Both $^{230}$Th and $^{231}$Pa show an increase in observed concentration with age of water, with the increase for $^{230}$Th much more regular than for $^{231}$Pa. This strong $^{230}$Th relationship, regardless of depth of the sample (Fig. 8a), indicates a primary control of water-mass age on the increase of $^{230}$Th in these younger waters.

For $^{230}$Th, the rate of increase with age (i.e. slope in Fig. 8a) indicates that about one quarter of the $^{230}$Th formed from U decay remains in the water, with the other three quarters being removed by scavenging. This ratio is consistent with the average $^{230}$Th for these waters, which requires that about three times more $^{230}$Th than remains in water has been removed by scavenging (Fig. 8a, 8b). The scatter between $^{231}$Pa and age (Fig. 8c) precludes the use of the scatter to assess the relative proportion of scavenged $^{231}$Pa, but the average values (Fig. 8c, 8d) indicate that about half of the $^{231}$Pa remains in the water, while half is removed by scavenging. The relative behaviour of $^{230}$Th and $^{231}$Pa is consistent with previous expectations, with a higher fraction of scavenging of $^{230}$Th than $^{231}$Pa.

The hypothesis that $^{231}$Pa/$^{230}$Th ratios increase monotonically as water mass ages forms the foundation of the Water-mass Evolution model for interpretation of sedimentary $^{231}$Pa/$^{230}$Th in terms of the rate of deep water circulation. For these young waters, however, there is no clear relationship between observed $^{231}$Pa/$^{230}$Th and age (Fig. 8e), and between the $^{231}$Pa/$^{230}$Th value scavenged to the sediment and age (Fig. 8f), calling the Water-mass Evolution model into question.

4.3 The importance of Preformed $^{230}$Th and $^{231}$Pa in young waters

To assess the controls on $^{230}$Th, $^{231}$Pa, and particularly the resulting $^{231}$Pa/$^{230}$Th ratio, we apply a simple scavenging-mixing model following Moran et al. (1997). This model was first created to assess the evolution of $^{230}$Th in a 1D water column as it ages following ventilation. Here we adopt it by modelling the nuclide evolution with age for each depth, and by also modelling $^{231}$Pa. This assumes that waters have remained at the same depth since ventilation which, though not correct in detail, still allows the model to provide insights about controls on these nuclides.

The model requires values for four parameters: particle settling speed (S), suspended particle concentration (SPM), and distribution coefficients for $^{230}$Th ($K_w^T$) and $^{231}$Pa ($K_w^P$). We select these parameters to give a good fit to the $^{230}$Th and $^{231}$Pa observations at an open ocean station, Station 13, on the east of the section (i.e. a station sampling older waters, which are
close to equilibrium) and use these values to interpret the younger waters to the west. Best fits to Station 13 suggested $S = 800$ m/yr, SPM = 25 $\mu$g/l; $K_W^{230} = 1.1 \times 10^5$ ml/g; and $K_W^{231} = 1.4 \times 10^6$ ml/g (the first three of these are close to those of Moran et al. 1997). A fuller description of the model is given in Supplemental Information S3.

We show two sets of output from the models, once with a preformed component ($C_{pre}$) equal to the nuclide concentrations observed in the upper 100 m of the GEOVIDE section (as in 4.2 above), and once with the preformed component set to zero for both nuclides. For both cases, the modelled evolution of nuclide concentrations with age between 0-50 years at 2000 m and 3500 m water depths is plotted in Fig. 9, and compared to data. As expected, modeled $^{230}$Th and $^{231}$Pa concentrations increase with age, with deeper waters having higher concentrations and $^{230}$Th increasing more rapidly initially (Fig. 9), but the preformed concentration is seen to be important in setting total nuclide concentration for several decades after ventilation.

The fit of the model to observations in young waters from GEOVIDE is improved in the model run with zero preformed nuclide, particularly for $^{230}$Th. This is surprising, given that surface-water $^{230}$Th and $^{231}$Pa values are generally non-zero, and typically close to the value observed in the GEOVIDE surface waters. For $^{230}$Th in young deep waters, even the model with zero preformed nuclide overestimates the observed value, possibly indicating additional scavenging from these waters close to the seafloor.

The most striking effect of changing the assumed preformed values in the model is on $^{231}$Pa/$^{230}$Th (Fig. 9c). When preformed values are set at zero, $^{231}$Pa/$^{230}$Th ratios always increase as water age increases, but when set at the average surface value from GEOVIDE, $^{231}$Pa/$^{230}$Th ratios initially decrease before increasing. The impact of preformed concentrations has a long-lasting impact on water-column and scavenged $^{231}$Pa/$^{230}$Th, lasting for hundreds of years following ventilation (Supplementary Information Figure S2 (c), (d)). This indicates that knowledge of the nuclide concentration at the site of deep-water formation is critical to understanding the early evolution of $^{231}$Pa/$^{230}$Th in waters and their underlying sediments.

### 4.4 Scavenging of $^{230}$Th and $^{231}$Pa

Knowledge of the CFC ages of the waters analysed on the GEOVIDE cruise allow an assessment of the scavenging rates of $^{230}$Th and $^{231}$Pa. To do so, we compare the Scavenged component to the Potential Total component (as defined in Section 4.2). An analysis of uncertainty of this assessment is given in Supplemental Information S4. The percentage of the Scavenged component relative to the Potential Total component is higher for $^{230}$Th, at an average of 80%, than for $^{231}$Pa at an average of 40% (Fig. 10), consistent with the relatively higher particle-reactivity of $^{230}$Th. For both nuclides, there is higher fraction of scavenging in samples from near the seafloor, particularly those in from DSOW in the deepest LA Sea. Bottom scavenging has been indicated in previous studies (e.g. Bacon and Anderson, 1982; Deng et al., 2014; Okubo et al., 2012), but this study indicates that this enhanced nuclide scavenging occurs even in the very young overflow waters at the start of the meridional circulation.

### 4.5 Meridional transport of $^{230}$Th and $^{231}$Pa in the North Atlantic

9
Previous calculations have indicated removal of $^{230}$Th and $^{231}$Pa from the North Atlantic by meridional transport southward. Deng et al. (2014) calculated net southward transport of 6% of the $^{230}$Th and 33% of $^{231}$Pa, relative to production of these nuclides in the water column. That calculation, however, did not provide a complete budget for $^{230}$Th and $^{231}$Pa for the North Atlantic because observations at the time did not constrain input of these nuclides from the north. Data in this study allow this calculation, and therefore a more complete budget for the modern North Atlantic.

Garcia-Ibáñez et al. (2018) calculated volume transports for the Portugal to Greenland section of the GEOVIDE section by combining the water mass fractions from eOMP analysis with the absolute geostrophic velocity field calculated using inverse model constrained by Doppler current profiler velocity measurements (Zunino et al., 2017). They separated northward flowing upper, and southward flowing lower limbs of the AMOC at isopycnal $\sigma_t$ (potential density referenced to 1000 dbar) = 32.15 kg/m$^3$, with $+18.7 \pm 2.4$ Sv and $-17.6 \pm 3.0$ Sv flow across the section above and below this value (positive value indicates northward transport). With average $^{230}$Th and $^{231}$Pa concentrations in the upper limb ($\sigma_t < 32.15$ kg/m$^3$) of 1.60 and 1.32 $\mu$Bq/kg respectively, northward transport of $^{230}$Th is $3.07 \times 10^{10}$ and of $^{231}$Pa is $2.51 \times 10^{11}$ $\mu$Bq/s. Average $^{230}$Th and $^{231}$Pa concentrations in the lower limb ($\sigma_t > 32.15$ kg/m$^3$) are 3.44 and 2.07 $\mu$Bq/kg respectively, indicating transports of $^{230}$Th and $^{231}$Pa are $-2.22 \times 10^{11}$ and $-3.74 \times 10^{10}$ $\mu$Bq/s respectively.

Net transport of $^{230}$Th and $^{231}$Pa across GEOVIDE is therefore to the south, and supplies $3.15 \times 10^{10}$ $\mu$Bq/s $^{230}$Th and $1.21 \times 10^{11}$ $\mu$Bq/s $^{231}$Pa to the North Atlantic (Fig. 11). This is a smaller net transport than further south in the Atlantic (Fig. 11), due to the lower $^{230}$Th and $^{231}$Pa concentrations in the water column close of the site of deep-water formation. The budget for these nuclides for the North Atlantic consists of: production in the water column; addition by advection from the North; loss by advection to the South and removal to the sediment. The data from this study allows this budget to be fully assessed, and indicates that the flux to the sediment is equivalent to 96% of the production of $^{230}$Th, and 74% of the production for $^{231}$Pa (Supplemental Information Table S5). For both nuclides, these fluxes are higher than in previous calculations (Deng et al. 2014) which ignored advective fluxes from the North. There is, however, still a significantly higher advective loss of $^{231}$Pa relative to $^{230}$Th. At a basin scale, therefore, $^{231}$Pa/$^{230}$Th in the sediment must be lower than the production ratio. This lower value is generated by the meridional transport of the North Atlantic, and likely to be sensitive to changes in this transport.

Use the Basin-scale Advection model to interpret sedimentary $^{231}$Pa/$^{230}$Th to assess meridional transport, as initially proposed by Yu et. al (1996), is therefore still supported by the full modern North Atlantic budget for these nuclides.

5 Conclusion

Measurement of $^{230}$Th and $^{231}$Pa in waters from GEOVIDE show some control of water mass on $^{230}$Th and $^{231}$Pa concentrations, particularly low concentrations in DSOW and high values in the old NEADW. There is, however, no close mapping of nuclide concentration to water mass.

With the availability of CFC ages on this section, the evolution of $^{230}$Th and $^{231}$Pa concentration with age is possible. A systematic increase of $^{236}$Th concentration is observed over the first 50 years following ventilation, and a similar though more

---

Deleted: defined between

Deleted: 0.098 and 0.082 dpm/1000l

Deleted: 1.83 x 10^6

Deleted: 1.53 x 10^5 dpm/s

Deleted: 0.22 and 0.15 dpm/1000l

Deleted: are -3.87 x 10^6 and -2.64 x 10^6 dpm/s

Deleted: 2.04 x 10^6 dpm/s $^{230}$Th and 2.55 x 10^6 dpm/s $^{231}$Pa, relative to production of these nuclides in the water column. That calculation, however, did not provide a complete budget for $^{230}$Th and $^{231}$Pa for the North Atlantic because observations at the time did not constrain input of these nuclides from the north. Data in this study allow this calculation, and therefore a more complete budget for the modern North Atlantic.

5 Conclusion

Measurement of $^{230}$Th and $^{231}$Pa in waters from GEOVIDE show some control of water mass on $^{230}$Th and $^{231}$Pa concentrations, particularly low concentrations in DSOW and high values in the old NEADW. There is, however, no close mapping of nuclide concentration to water mass.

With the availability of CFC ages on this section, the evolution of $^{230}$Th and $^{231}$Pa concentration with age is possible. A systematic increase of $^{236}$Th concentration is observed over the first 50 years following ventilation, and a similar though more
scattered relationship seen for 231Pa. There is no clear relationship between the 231Pa/230Th ratio and age for these young waters. The long-term evolution of 231Pa/230Th is found from a simple model to be highly dependent on the preformed concentrations for these nuclides. These results complicate the interpretation of sedimentary 231Pa/230Th as a paleo-proxy for deep water circulation based on systematic evolution of water 231Pa/230Th with age, and point to the importance of a better knowledge of preformed 230Th and 231Pa concentrations to improve interpretation. This analysis of the 230Th and 231Pa concentration relative to the age of the water not only demonstrates the influence of water mass aging on 231Pa and 230Th, but also points to the influence of scavenging. Scavenged 230Th is much more extensive that 231Pa, as expected, and enhanced removal of both nuclides is seen immediately above the seafloor, particularly for young waters.

Calculation of meridional transport of 230Th and 231Pa indicates a southward net transport of both nuclides across the GEOVIDE section. This advection is smaller than that further south in the Atlantic as a result of lower 230Th and 231Pa concentrations at GEOVIDE. Calculation of the flux across GEOVIDE allows a more complete budget for the North Atlantic to be constructed and demonstrates a significantly higher advective loss of 231Pa to the south relative to 230Th, with 2x6% of the 231Pa produced advected southward (relative to only 2x4% for 230Th). This calculation supports the interpretation of sedimentary 231Pa/230Th measurements as a proxy for overturning circulation, when based on advective loss of 231Pa at a basin scale.

Acknowledgements

Géraldine Sarthou and Pascale Lherminier are thanked for leading the GEOVIDE cruise, along with the captain, Gilles Ferrand, and crew of the R/V Pourquoi Pas?. We would like to give a special thanks to Pierre Branellec, Floriane Desprez de Gésincourt, Michel Hamon, Catherine Kermabon, Philippe Le Bot, Stéphane Leizour, Olivier Ménage, Fabien Pérault and Emmanuel de Saint Léger for their technical expertise and to Catherine Schmechtig for the GEOVIDE database management. This work was supported by the French National Research Agency (ANR-13-BS06-0014, ANR-12-PDOC-0025-01), the French National Centre for Scientific Research (CNRS-LEFE-CYBER), the LabexMER (ANR-10-LABX-19), and Ifremer. It was supported for the logistic by DT-INSU and GENAVIR. Thank you also to Yi Tang who helped with sampling during the cruise. We are grateful to Mercedes de la Paz Arandiga, and Pascale Lherminier for providing valuable input during analysis of water masses and ages, and to Maribel García-Ibáñez for early provision of the eOMP analysis presented elsewhere in this volume. We also thank Yves Plancherel for valuable insight during discussion of the results presented.

References


Figure 1: Map showing GEOVIDE cruise track (black line) and station locations (black dots). Colour bars indicate water depth. Sampling locations for water-column $^{231}$Pa and $^{230}$Th in this study are shown by coloured dots, with colours representing the ocean regions they are located in.
Figure 2: Vertical profiles of $^{230}$Th (a-d) and $^{231}$Pa (e-h) in the water column along the GEOVIDE section. Colours corresponds to the region (as in Fig. 1). LA = Labrador, IR = Irminger, IC = Iceland, WE = West European. Uncertainties represent 2 standard error (2 s.e.).
Figure 3: Vertical profiles of $^{230}$Th and $^{231}$Pa from high-latitude Northwest Atlantic (west section of GEOVIDE), mid-latitude Northwest Atlantic (GA03_w), and Southwest Atlantic (GA02). Data from all stations were sorted by water depth and averages for depth and $^{230}$Th and $^{231}$Pa concentrations were taken for surface, 25 m, 100 m, 250 m, 500 m, and every 500 m interval below. Error bars on $^{230}$Th and $^{231}$Pa concentrations averages reflect standard deviation of the mean of measurements.
Figure 4: Results of Extended Optimum MultiParameter (eOMP) analysis for the GEOVIDE section (García-Ibáñez et al., 2018). Colours reflect the fraction of water at each location assigned to the water mass shown in that panel: ENACW$_{16}$ and ENACW$_{12}$ = East North Atlantic Central Water of 16°C and 12°C; SPMW$_8$, SPMW$_7$, IrSPMW = Subpolar Mode Water of 8°C, 7°C and of the Irminger Sea; SAIW$_6$ and SAIW$_4$ = Subarctic Intermediate Water of 6°C and 4°C; MW = Mediterranean Water; PIW = Polar Intermediate Water; ISOW=Iceland–Scotland Overflow Water; LSW=Labrador Sea Water; DSOW: Denmark Strait Overflow Waters; and NEADW$_L$: Lower North East Atlantic Deep Water; ABR= Azores-Biscay Rise.

Deleted: SAIW=Subarctic Intermediate Waters;
Figure 5: Distribution of $^{230}$Th and $^{231}$Pa along the GEOVIDE section. Water masses were indicated by contours (black: DSOW; blue: ISOW; white: LSW; red: NEADW.) based on 50% level percentage composition of source water types from eOMP analysis.
Figure 6: Water mass age based on CFC data along the GEOVIDE section. (a) Full water-column data for the entire section, showing waters from 10 to 800 years in age; (b) A rescaled version of (a) omitting the upper 1000m and the older waters west of 35°W to show age variation in recently ventilated deep-waters. Water masses were indicated by contours (black: DSW; blue: ISOW; white: LSW; red: NEADW.) based on 50% level percentage composition of source water types from eOMP analysis.
Figure 7: Potential Total, Observed, and Scavenged components of $^{230}$Th and $^{231}$Pa in waters >1000 m water depth and west of 35°W.
Figure 8: Relationship between water mass age and the Observed and Scavenged components of $^{230}$Th, $^{231}$Pa and $^{231}$Pa/$^{230}$Th (colour coded by water depth). Least square fitting statistics were also given, i.e. slope and correlation coefficient $r$ of the least square line, mean value and number of the data points. Note the increase of observed concentrations for both nuclides with age. Comparison of average values indicates that about three quarters of $^{230}$Th produced by decay is scavenged, compared with about half of the $^{231}$Pa.

Deleted: Note the increase of observed concentrations for both nuclides with age. Comparison of average values indicates that about three quarters of $^{230}$Th produced by decay is scavenged, compared with about half of the $^{231}$Pa.
Figure 9: Results from a scavenging-mixing model of $^{230}$Th, $^{231}$Pa, Dissolved $^{231}$Pa/$^{230}$Th and particulate $^{231}$Pa/$^{230}$Th compared to observations. Preformed concentration ($C_{pre}$) were set at 0 (dashed line) and at the average surface concentration ($C_{surface\ average}$) from GEOVIDE section (solid line), i.e. $^{230}$Th=1.66 µBq/kg, $^{231}$Pa=1.31 µBq/kg. A version of this figure extending to older waters is available in the Supplemental Information.
Figure 10: Ratio of Scavenged component to Potential Total component for $^{230}$Th and $^{231}$Pa, providing an assessment of the relative importance of scavenging for the two nuclides, and of the location of scavenging.
Figure 11: Fluxes of $^{230}$Th (blue arrow) and $^{231}$Pa (yellow arrow) across the GEOVIDE section (blue solid line), 4.5°S (green solid line) and 45°S (red solid line). Also shown are production of $^{230}$Th (blue box) and $^{231}$Pa (yellow box) in the North Atlantic (between GEOVIDE section and 4.5°S) and in the South Atlantic (between 4.5°S and 45°S), based on calculation in Deng et al. (2014). These fluxes indicate that 4% of the $^{230}$Th produced in the North Atlantic is exported southward by ocean circulation, and 26% of the $^{231}$Pa.
Supplemental Information of

Evolution of $^{231}\text{Pa}$ and $^{230}\text{Th}$ in overflow waters of the North Atlantic

Feifei Deng et al.

Correspondence to: Feifei Deng (feifei.deng@earth.ox.ac.uk)
## S1. Data of water-column $^{231}$Pa, $^{230}$Th and $^{232}$Th concentrations, and $^{231}$Pa/$^{230}$Th ratios along GEOVIDE section

### Table S1 Water-column $^{231}$Pa, $^{230}$Th and $^{232}$Th concentrations, and $^{231}$Pa/$^{230}$Th ratios along GEOVIDE section

<table>
<thead>
<tr>
<th>Station</th>
<th>Depth m</th>
<th>$^{231}$Pa fg/kg</th>
<th>$^{231}$Pa act. fg/kg</th>
<th>$^{230}$Th fg/kg</th>
<th>$^{230}$Th act. fg/kg</th>
<th>$^{232}$Th fg/kg</th>
<th>$^{232}$Th act. fg/kg</th>
<th>$^{231}$Pa/$^{230}$Th</th>
<th>$^{232}$Th/$^{230}$Th</th>
<th>$^{231}$Pa/$^{232}$Th</th>
<th>$^{231}$Pa/$^{230}$Th corr</th>
<th>$^{232}$Th/$^{230}$Th corr</th>
<th>$^{232}$Th/$^{230}$Th 2se</th>
</tr>
</thead>
<tbody>
<tr>
<td>L</td>
<td>3445.3</td>
<td>2.279</td>
<td>3.96</td>
<td>0.17</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>40.33°N</td>
<td>2955.3</td>
<td>2.463</td>
<td>4.30</td>
<td>0.22</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>0.04°W</td>
<td>2465.8</td>
<td>1.988</td>
<td>2.46</td>
<td>3.03</td>
<td>1.77</td>
<td>1.93</td>
<td>2.10</td>
<td>3.03</td>
<td>1.77</td>
<td>1.93</td>
<td>2.10</td>
<td>3.03</td>
<td>1.77</td>
</tr>
<tr>
<td></td>
<td>495.6</td>
<td>0.861</td>
<td>1.49</td>
<td>2.65</td>
<td>0.23</td>
<td>0.35</td>
<td>0.23</td>
<td>0.35</td>
<td>0.23</td>
<td>0.35</td>
<td>0.23</td>
<td>0.35</td>
<td>0.23</td>
</tr>
<tr>
<td></td>
<td>246.9</td>
<td>0.635</td>
<td>1.09</td>
<td>2.79</td>
<td>0.20</td>
<td>0.48</td>
<td>0.20</td>
<td>0.48</td>
<td>0.20</td>
<td>0.48</td>
<td>0.20</td>
<td>0.48</td>
<td>0.20</td>
</tr>
<tr>
<td></td>
<td>49.6</td>
<td>0.641</td>
<td>0.623</td>
<td>1.09</td>
<td>0.20</td>
<td>2.84</td>
<td>0.20</td>
<td>2.84</td>
<td>0.20</td>
<td>2.84</td>
<td>0.20</td>
<td>2.84</td>
<td>0.20</td>
</tr>
<tr>
<td></td>
<td>4.7</td>
<td>0.238</td>
<td>0.210</td>
<td>0.37</td>
<td>0.08</td>
<td>3.93</td>
<td>0.37</td>
<td>0.08</td>
<td>3.93</td>
<td>0.37</td>
<td>0.08</td>
<td>3.93</td>
<td>0.37</td>
</tr>
<tr>
<td>13</td>
<td>5330.4</td>
<td>2.093</td>
<td>3.28</td>
<td>0.24</td>
<td>8.69</td>
<td>8.62</td>
<td>0.24</td>
<td>8.69</td>
<td>8.62</td>
<td>0.24</td>
<td>8.69</td>
<td>8.62</td>
<td>0.24</td>
</tr>
<tr>
<td>41.38°N</td>
<td>5263.1</td>
<td>2.421</td>
<td>2.178</td>
<td>3.80</td>
<td>0.26</td>
<td>12.66</td>
<td>3.80</td>
<td>0.26</td>
<td>12.66</td>
<td>3.80</td>
<td>0.26</td>
<td>12.66</td>
<td>3.80</td>
</tr>
<tr>
<td>53.89°W</td>
<td>5194.3</td>
<td>2.751</td>
<td>2.747</td>
<td>4.80</td>
<td>0.24</td>
<td>15.80</td>
<td>4.80</td>
<td>0.24</td>
<td>15.80</td>
<td>4.80</td>
<td>0.24</td>
<td>15.80</td>
<td>4.80</td>
</tr>
<tr>
<td></td>
<td>4903.9</td>
<td>2.013</td>
<td>2.009</td>
<td>3.51</td>
<td>0.15</td>
<td>14.79</td>
<td>3.51</td>
<td>0.15</td>
<td>14.79</td>
<td>3.51</td>
<td>0.15</td>
<td>14.79</td>
<td>3.51</td>
</tr>
<tr>
<td></td>
<td>4417.8</td>
<td>2.627</td>
<td>2.624</td>
<td>4.58</td>
<td>0.24</td>
<td>11.16</td>
<td>4.58</td>
<td>0.24</td>
<td>11.16</td>
<td>4.58</td>
<td>0.24</td>
<td>11.16</td>
<td>4.58</td>
</tr>
<tr>
<td></td>
<td>3444.3</td>
<td>2.399</td>
<td>2.396</td>
<td>4.19</td>
<td>0.14</td>
<td>8.15</td>
<td>4.19</td>
<td>0.14</td>
<td>8.15</td>
<td>4.19</td>
<td>0.14</td>
<td>8.15</td>
<td>4.19</td>
</tr>
<tr>
<td></td>
<td>2464.7</td>
<td>1.569</td>
<td>1.565</td>
<td>2.73</td>
<td>0.26</td>
<td>5.77</td>
<td>2.73</td>
<td>0.26</td>
<td>5.77</td>
<td>2.73</td>
<td>0.26</td>
<td>5.77</td>
<td>2.73</td>
</tr>
<tr>
<td></td>
<td>1187.3</td>
<td>0.869</td>
<td>0.865</td>
<td>1.51</td>
<td>0.12</td>
<td>3.24</td>
<td>1.51</td>
<td>0.12</td>
<td>3.24</td>
<td>1.51</td>
<td>0.12</td>
<td>3.24</td>
<td>1.51</td>
</tr>
<tr>
<td></td>
<td>989.1</td>
<td>1.239</td>
<td>1.234</td>
<td>2.16</td>
<td>0.15</td>
<td>4.16</td>
<td>2.16</td>
<td>0.15</td>
<td>4.16</td>
<td>2.16</td>
<td>0.15</td>
<td>4.16</td>
<td>2.16</td>
</tr>
<tr>
<td></td>
<td>248.4</td>
<td>0.602</td>
<td>0.602</td>
<td>1.05</td>
<td>0.11</td>
<td></td>
<td>1.05</td>
<td>0.11</td>
<td></td>
<td>1.05</td>
<td>0.11</td>
<td></td>
<td>1.05</td>
</tr>
<tr>
<td></td>
<td>148.5</td>
<td>0.525</td>
<td>0.425</td>
<td>0.92</td>
<td>0.13</td>
<td></td>
<td>0.92</td>
<td>0.13</td>
<td></td>
<td>0.92</td>
<td>0.13</td>
<td></td>
<td>0.92</td>
</tr>
<tr>
<td>29.7</td>
<td>0.508</td>
<td>0.507</td>
<td>0.89</td>
<td>0.11</td>
<td>0.44</td>
<td>0.43</td>
<td>0.11</td>
<td>0.44</td>
<td>0.43</td>
<td>0.11</td>
<td>0.44</td>
<td>0.43</td>
<td>0.11</td>
</tr>
<tr>
<td>4.3</td>
<td>0.330</td>
<td>0.330</td>
<td>0.58</td>
<td>0.08</td>
<td>0.28</td>
<td>0.26</td>
<td>0.08</td>
<td>0.28</td>
<td>0.26</td>
<td>0.08</td>
<td>0.28</td>
<td>0.26</td>
<td>0.08</td>
</tr>
<tr>
<td>Station</td>
<td>Depth</td>
<td>213Pa</td>
<td>231Pa</td>
<td>231Pa corr</td>
<td>2ss</td>
<td>238Th</td>
<td>232Th corr</td>
<td>2ss</td>
<td>232Th</td>
<td>2ss</td>
<td>231Pa/232Th</td>
<td></td>
<td></td>
</tr>
<tr>
<td>---------</td>
<td>-------</td>
<td>-------</td>
<td>-------</td>
<td>------------</td>
<td>-----</td>
<td>-------</td>
<td>------------</td>
<td>-----</td>
<td>-------</td>
<td>-----</td>
<td>------------</td>
<td></td>
<td></td>
</tr>
<tr>
<td>1</td>
<td>4514</td>
<td>2.605</td>
<td>2.603</td>
<td>4.55</td>
<td>0.15</td>
<td>6.09</td>
<td>6.03</td>
<td>4.58</td>
<td>0.19</td>
<td>20.99</td>
<td>0.0902</td>
<td></td>
<td></td>
</tr>
<tr>
<td>1</td>
<td>4715</td>
<td>2.404</td>
<td>2.601</td>
<td>4.20</td>
<td>0.18</td>
<td>6.21</td>
<td>6.13</td>
<td>4.66</td>
<td>0.28</td>
<td>25.11</td>
<td>0.1079</td>
<td></td>
<td></td>
</tr>
<tr>
<td>2</td>
<td>4263</td>
<td>2.413</td>
<td>2.411</td>
<td>4.21</td>
<td>0.17</td>
<td>5.76</td>
<td>5.79</td>
<td>4.33</td>
<td>0.14</td>
<td>19.57</td>
<td>0.0841</td>
<td></td>
<td></td>
</tr>
<tr>
<td>2</td>
<td>4279</td>
<td>2.497</td>
<td>2.495</td>
<td>4.36</td>
<td>0.18</td>
<td>5.91</td>
<td>5.85</td>
<td>4.45</td>
<td>0.19</td>
<td>20.48</td>
<td>0.0880</td>
<td></td>
<td></td>
</tr>
<tr>
<td>3</td>
<td>3929</td>
<td>2.081</td>
<td>2.079</td>
<td>3.63</td>
<td>0.17</td>
<td>5.47</td>
<td>5.41</td>
<td>4.11</td>
<td>0.16</td>
<td>20.11</td>
<td>0.0864</td>
<td></td>
<td></td>
</tr>
<tr>
<td>3</td>
<td>3443</td>
<td>1.930</td>
<td>1.928</td>
<td>3.37</td>
<td>0.20</td>
<td>4.63</td>
<td>4.57</td>
<td>3.47</td>
<td>0.14</td>
<td>18.16</td>
<td>0.0780</td>
<td></td>
<td></td>
</tr>
<tr>
<td>4</td>
<td>2688</td>
<td>1.114</td>
<td>1.114</td>
<td>1.95</td>
<td>0.22</td>
<td>5.16</td>
<td>5.06</td>
<td>3.85</td>
<td>0.17</td>
<td>30.49</td>
<td>0.1309</td>
<td></td>
<td></td>
</tr>
<tr>
<td>4</td>
<td>1482</td>
<td>0.995</td>
<td>0.993</td>
<td>1.73</td>
<td>0.12</td>
<td>2.48</td>
<td>2.39</td>
<td>1.82</td>
<td>0.14</td>
<td>76.83</td>
<td>0.1152</td>
<td></td>
<td></td>
</tr>
<tr>
<td>5</td>
<td>788</td>
<td>0.873</td>
<td>0.871</td>
<td>1.52</td>
<td>0.14</td>
<td>1.31</td>
<td>1.23</td>
<td>0.93</td>
<td>0.16</td>
<td>25.41</td>
<td>0.1092</td>
<td></td>
<td></td>
</tr>
<tr>
<td>5</td>
<td>445</td>
<td>0.679</td>
<td>0.676</td>
<td>1.18</td>
<td>0.14</td>
<td>2.18</td>
<td>2.05</td>
<td>1.56</td>
<td>0.14</td>
<td>39.91</td>
<td>0.1714</td>
<td></td>
<td></td>
</tr>
<tr>
<td>6</td>
<td>246</td>
<td>0.595</td>
<td>0.592</td>
<td>1.03</td>
<td>0.11</td>
<td>1.64</td>
<td>1.53</td>
<td>1.17</td>
<td>0.13</td>
<td>33.41</td>
<td>0.1435</td>
<td></td>
<td></td>
</tr>
<tr>
<td>6</td>
<td>98.1</td>
<td>0.607</td>
<td>0.603</td>
<td>1.05</td>
<td>0.14</td>
<td>1.32</td>
<td>1.23</td>
<td>0.93</td>
<td>0.12</td>
<td>28.01</td>
<td>0.1203</td>
<td></td>
<td></td>
</tr>
<tr>
<td>7</td>
<td>13.9</td>
<td>0.536</td>
<td>0.532</td>
<td>0.93</td>
<td>0.15</td>
<td>1.15</td>
<td>1.01</td>
<td>10.81</td>
<td>0.0464</td>
<td>0.0015</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>7</td>
<td>3.7</td>
<td>0.522</td>
<td>0.519</td>
<td>0.14</td>
<td>0.09</td>
<td>1.14</td>
<td>0.91</td>
<td>7.95</td>
<td>0.0342</td>
<td>0.0014</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>8</td>
<td>26</td>
<td>4116.3</td>
<td>1.162</td>
<td>1.158</td>
<td>2.02</td>
<td>3.43</td>
<td>3.29</td>
<td>2.50</td>
<td>0.17</td>
<td>42.15</td>
<td>0.1810</td>
<td></td>
<td></td>
</tr>
<tr>
<td>9</td>
<td>50.28</td>
<td>2758.5</td>
<td>0.813</td>
<td>0.807</td>
<td>1.41</td>
<td>3.04</td>
<td>2.85</td>
<td>2.17</td>
<td>0.14</td>
<td>57.40</td>
<td>0.2465</td>
<td></td>
<td></td>
</tr>
<tr>
<td>10</td>
<td>22.60</td>
<td>1973.5</td>
<td>0.745</td>
<td>0.739</td>
<td>1.29</td>
<td>3.83</td>
<td>3.65</td>
<td>2.01</td>
<td>0.16</td>
<td>56.40</td>
<td>0.2322</td>
<td></td>
<td></td>
</tr>
<tr>
<td>11</td>
<td>989</td>
<td>0.695</td>
<td>0.695</td>
<td>1.22</td>
<td>0.11</td>
<td>0.11</td>
<td>0.11</td>
<td>0.99</td>
<td>0.0017</td>
<td>0.0018</td>
<td>0.992</td>
<td></td>
<td></td>
</tr>
<tr>
<td>12</td>
<td>296.7</td>
<td>0.503</td>
<td>0.503</td>
<td>0.88</td>
<td>0.10</td>
<td>0.10</td>
<td>0.10</td>
<td>0.99</td>
<td>0.0017</td>
<td>0.0018</td>
<td>0.992</td>
<td></td>
<td></td>
</tr>
<tr>
<td>13</td>
<td>74.4</td>
<td>0.408</td>
<td>0.407</td>
<td>0.71</td>
<td>0.12</td>
<td>0.67</td>
<td>0.62</td>
<td>0.47</td>
<td>0.17</td>
<td>13.57</td>
<td>0.0583</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

**Notes:**
- Depths are in meters.
- Concentrations are in fg/kg and µBq/kg.
- Activity ratios are in pg/kg and pmol/kg.
<table>
<thead>
<tr>
<th>Station</th>
<th>Depth m</th>
<th>213Pa fg/kg</th>
<th>213Pu fg/kg</th>
<th>213Pa/239Pu</th>
<th>239Pu fg/kg</th>
<th>239Th fg/kg</th>
<th>239Th/233Th</th>
<th>233Th fg/kg</th>
<th>233Th/232Th</th>
<th>232Th fg/kg</th>
<th>232Th/231Pa</th>
<th>231Pa fg/kg</th>
</tr>
</thead>
<tbody>
<tr>
<td>S2</td>
<td>3218.5</td>
<td>0.984</td>
<td>1.22</td>
<td>0.12</td>
<td>1.68</td>
<td>1.62</td>
<td>1.24</td>
<td>0.14</td>
<td>15.25</td>
<td>0.655</td>
<td>0.0016</td>
<td>1.387</td>
</tr>
<tr>
<td>55.51°N</td>
<td>3218.5</td>
<td>0.984</td>
<td>1.22</td>
<td>0.12</td>
<td>1.68</td>
<td>1.62</td>
<td>1.24</td>
<td>0.14</td>
<td>15.25</td>
<td>0.655</td>
<td>0.0016</td>
<td>1.387</td>
</tr>
<tr>
<td>26.71°W</td>
<td>3049.9</td>
<td>1.131</td>
<td>1.97</td>
<td>0.11</td>
<td>3.27</td>
<td>3.21</td>
<td>2.44</td>
<td>0.13</td>
<td>20.44</td>
<td>0.087</td>
<td>0.0016</td>
<td>0.809</td>
</tr>
<tr>
<td>31.27°W</td>
<td>1301.6</td>
<td>1.336</td>
<td>2.33</td>
<td>0.11</td>
<td>3.34</td>
<td>3.25</td>
<td>2.47</td>
<td>0.20</td>
<td>29.18</td>
<td>0.1253</td>
<td>0.0026</td>
<td>0.943</td>
</tr>
<tr>
<td>31.27°W</td>
<td>1301.6</td>
<td>1.336</td>
<td>2.33</td>
<td>0.11</td>
<td>3.34</td>
<td>3.25</td>
<td>2.47</td>
<td>0.20</td>
<td>29.18</td>
<td>0.1253</td>
<td>0.0026</td>
<td>0.943</td>
</tr>
<tr>
<td>108.7</td>
<td>1274.4</td>
<td>1.299</td>
<td>2.22</td>
<td>0.19</td>
<td>2.78</td>
<td>2.65</td>
<td>2.01</td>
<td>0.14</td>
<td>42.89</td>
<td>0.1842</td>
<td>0.0036</td>
<td>1.102</td>
</tr>
<tr>
<td>20.3</td>
<td>0.836</td>
<td>0.835</td>
<td>1.46</td>
<td>0.13</td>
<td>0.84</td>
<td>0.80</td>
<td>0.61</td>
<td>0.13</td>
<td>11.88</td>
<td>0.0510</td>
<td>0.0016</td>
<td>2.389</td>
</tr>
<tr>
<td>5.3</td>
<td>1.230</td>
<td>1.228</td>
<td>2.15</td>
<td>0.25</td>
<td>0.80</td>
<td>0.76</td>
<td>0.58</td>
<td>0.15</td>
<td>11.59</td>
<td>0.0498</td>
<td>0.0016</td>
<td>3.719</td>
</tr>
<tr>
<td>Station</td>
<td>Depth (m)</td>
<td>210Pa</td>
<td>210Pa corr</td>
<td>228Th</td>
<td>220Th</td>
<td>228Th corr</td>
<td>220Th corr</td>
<td>228Th</td>
<td>220Th</td>
<td>228Th/220Th</td>
<td>220Th</td>
<td></td>
</tr>
<tr>
<td>---------</td>
<td>-----------</td>
<td>--------</td>
<td>------------</td>
<td>----------</td>
<td>----------</td>
<td>------------</td>
<td>------------</td>
<td>-------</td>
<td>-------</td>
<td>-------------</td>
<td>-------</td>
<td></td>
</tr>
<tr>
<td>44</td>
<td>2918.9</td>
<td>0.837</td>
<td>1.46</td>
<td>0.17</td>
<td>2.40</td>
<td>3.33</td>
<td>1.77</td>
<td>0.13</td>
<td>22.85</td>
<td>0.0981</td>
<td>0.0022</td>
<td></td>
</tr>
<tr>
<td>59.62°N</td>
<td>2878.5</td>
<td>0.873</td>
<td>1.52</td>
<td>0.15</td>
<td>2.50</td>
<td>2.43</td>
<td>1.84</td>
<td>0.14</td>
<td>24.44</td>
<td>0.1050</td>
<td>0.0023</td>
<td></td>
</tr>
<tr>
<td>58.95°W</td>
<td>2829</td>
<td>0.723</td>
<td>1.26</td>
<td>0.22</td>
<td>1.33</td>
<td>2.65</td>
<td>2.57</td>
<td>1.96</td>
<td>0.15</td>
<td>25.27</td>
<td>0.1085</td>
<td></td>
</tr>
<tr>
<td></td>
<td>2681.9</td>
<td>0.651</td>
<td>0.649</td>
<td>1.13</td>
<td>0.24</td>
<td>2.65</td>
<td>2.57</td>
<td>1.96</td>
<td>0.15</td>
<td>25.27</td>
<td>0.1085</td>
<td></td>
</tr>
<tr>
<td></td>
<td>2561</td>
<td>1.164</td>
<td>1.161</td>
<td>2.03</td>
<td>0.23</td>
<td>4.87</td>
<td>4.78</td>
<td>3.64</td>
<td>0.17</td>
<td>26.47</td>
<td>0.1137</td>
<td></td>
</tr>
<tr>
<td></td>
<td>2216.6</td>
<td>0.910</td>
<td>0.906</td>
<td>1.58</td>
<td>0.26</td>
<td>6.18</td>
<td>6.08</td>
<td>4.63</td>
<td>0.18</td>
<td>31.25</td>
<td>0.1342</td>
<td></td>
</tr>
<tr>
<td></td>
<td>1776</td>
<td>1.416</td>
<td>1.412</td>
<td>2.47</td>
<td>0.39</td>
<td>5.87</td>
<td>5.75</td>
<td>4.37</td>
<td>0.15</td>
<td>39.66</td>
<td>0.1704</td>
<td></td>
</tr>
<tr>
<td></td>
<td>1382.5</td>
<td>1.186</td>
<td>1.182</td>
<td>2.07</td>
<td>0.27</td>
<td>5.85</td>
<td>5.72</td>
<td>4.35</td>
<td>0.15</td>
<td>41.00</td>
<td>0.1761</td>
<td></td>
</tr>
<tr>
<td></td>
<td>1087.5</td>
<td>0.795</td>
<td>0.790</td>
<td>1.38</td>
<td>0.18</td>
<td>3.83</td>
<td>3.70</td>
<td>2.81</td>
<td>0.15</td>
<td>40.73</td>
<td>0.1749</td>
<td></td>
</tr>
<tr>
<td></td>
<td>593.2</td>
<td>1.116</td>
<td>1.111</td>
<td>1.94</td>
<td>0.36</td>
<td>4.28</td>
<td>4.13</td>
<td>3.14</td>
<td>0.16</td>
<td>46.81</td>
<td>0.2010</td>
<td></td>
</tr>
<tr>
<td></td>
<td>297.3</td>
<td>1.083</td>
<td>1.078</td>
<td>1.88</td>
<td>0.20</td>
<td>4.10</td>
<td>3.97</td>
<td>3.02</td>
<td>0.22</td>
<td>42.29</td>
<td>0.1816</td>
<td></td>
</tr>
<tr>
<td></td>
<td>78.8</td>
<td>0.929</td>
<td>0.925</td>
<td>1.62</td>
<td>0.19</td>
<td>3.44</td>
<td>3.31</td>
<td>2.52</td>
<td>0.14</td>
<td>39.69</td>
<td>0.1705</td>
<td></td>
</tr>
<tr>
<td></td>
<td>25.5</td>
<td>0.998</td>
<td>0.997</td>
<td>1.74</td>
<td>0.16</td>
<td>1.37</td>
<td>1.32</td>
<td>1.00</td>
<td>0.15</td>
<td>16.50</td>
<td>0.0709</td>
<td></td>
</tr>
<tr>
<td></td>
<td>5.5</td>
<td>0.722</td>
<td>0.720</td>
<td>1.26</td>
<td>0.33</td>
<td>1.31</td>
<td>1.27</td>
<td>0.96</td>
<td>0.12</td>
<td>14.99</td>
<td>0.0644</td>
<td></td>
</tr>
<tr>
<td>50</td>
<td>1710.9</td>
<td>0.844</td>
<td>0.841</td>
<td>1.47</td>
<td>0.07</td>
<td>3.77</td>
<td>3.67</td>
<td>2.79</td>
<td>0.16</td>
<td>31.42</td>
<td>0.1349</td>
<td></td>
</tr>
<tr>
<td>59.80°N</td>
<td>1652.6</td>
<td>0.881</td>
<td>0.878</td>
<td>1.53</td>
<td>0.07</td>
<td>4.03</td>
<td>3.92</td>
<td>2.98</td>
<td>0.16</td>
<td>33.28</td>
<td>0.1429</td>
<td></td>
</tr>
<tr>
<td>62.01°W</td>
<td>1603.1</td>
<td>0.845</td>
<td>0.842</td>
<td>1.47</td>
<td>0.07</td>
<td>4.10</td>
<td>3.92</td>
<td>3.04</td>
<td>0.24</td>
<td>33.74</td>
<td>0.1449</td>
<td></td>
</tr>
<tr>
<td></td>
<td>1481.2</td>
<td>0.813</td>
<td>0.809</td>
<td>1.41</td>
<td>0.07</td>
<td>4.19</td>
<td>4.08</td>
<td>3.10</td>
<td>0.17</td>
<td>33.94</td>
<td>0.1458</td>
<td></td>
</tr>
<tr>
<td></td>
<td>989.7</td>
<td>1.087</td>
<td>1.082</td>
<td>1.89</td>
<td>0.08</td>
<td>5.29</td>
<td>5.15</td>
<td>3.91</td>
<td>0.20</td>
<td>45.60</td>
<td>0.1958</td>
<td></td>
</tr>
<tr>
<td></td>
<td>495.5</td>
<td>0.911</td>
<td>0.906</td>
<td>1.58</td>
<td>0.16</td>
<td>3.69</td>
<td>3.56</td>
<td>2.71</td>
<td>0.14</td>
<td>39.63</td>
<td>0.1702</td>
<td></td>
</tr>
<tr>
<td></td>
<td>247.9</td>
<td>0.851</td>
<td>0.847</td>
<td>1.48</td>
<td>0.23</td>
<td>3.16</td>
<td>3.03</td>
<td>2.30</td>
<td>0.14</td>
<td>40.61</td>
<td>0.1744</td>
<td></td>
</tr>
<tr>
<td></td>
<td>99.1</td>
<td>0.826</td>
<td>0.822</td>
<td>1.44</td>
<td>0.20</td>
<td>3.34</td>
<td>3.23</td>
<td>1.69</td>
<td>0.13</td>
<td>34.75</td>
<td>0.1492</td>
<td></td>
</tr>
<tr>
<td></td>
<td>19.5</td>
<td>0.887</td>
<td>0.885</td>
<td>1.55</td>
<td>0.32</td>
<td>1.36</td>
<td>1.30</td>
<td>0.99</td>
<td>0.16</td>
<td>18.14</td>
<td>0.0779</td>
<td></td>
</tr>
<tr>
<td></td>
<td>3.7</td>
<td>0.829</td>
<td>0.827</td>
<td>1.44</td>
<td>0.24</td>
<td>1.53</td>
<td>1.46</td>
<td>1.11</td>
<td>0.14</td>
<td>22.32</td>
<td>0.0959</td>
<td></td>
</tr>
<tr>
<td>Station</td>
<td>Depth (m)</td>
<td>210Pa (fg/kg)</td>
<td>231Pa (fg/kg)</td>
<td>231Pacorr (fg/kg)</td>
<td>218Th (pg/kg)</td>
<td>230Th (pg/kg)</td>
<td>232Th (pg/kg)</td>
<td>234Th (pg/kg)</td>
<td>235Th (pg/kg)</td>
<td>238U (pg/kg)</td>
<td>232Th/235Th</td>
<td>230Th/232Th</td>
</tr>
<tr>
<td>---------</td>
<td>-----------</td>
<td>----------------</td>
<td>---------------</td>
<td>-------------------</td>
<td>---------------</td>
<td>--------------</td>
<td>--------------</td>
<td>---------------</td>
<td>--------------</td>
<td>--------------</td>
<td>-------------</td>
<td>-------------</td>
</tr>
<tr>
<td>64</td>
<td>2466.8</td>
<td>0.756</td>
<td>1.32</td>
<td>0.19</td>
<td>3.24</td>
<td>3.16</td>
<td>2.40</td>
<td>0.14</td>
<td>25.71</td>
<td>0.1104</td>
<td>0.0025</td>
<td>0.551</td>
</tr>
<tr>
<td>59.07°N</td>
<td>2423.6</td>
<td>0.825</td>
<td>1.44</td>
<td>0.20</td>
<td>3.71</td>
<td>3.63</td>
<td>2.76</td>
<td>0.18</td>
<td>26.12</td>
<td>0.1122</td>
<td>0.0027</td>
<td>0.523</td>
</tr>
<tr>
<td>46.08°W</td>
<td>2374.9</td>
<td>0.955</td>
<td>1.67</td>
<td>0.19</td>
<td>3.76</td>
<td>3.68</td>
<td>2.80</td>
<td>0.13</td>
<td>27.16</td>
<td>0.1166</td>
<td>0.0025</td>
<td>0.597</td>
</tr>
<tr>
<td>2226.6</td>
<td>1.051</td>
<td>1.048</td>
<td>1.83</td>
<td>0.27</td>
<td>5.33</td>
<td>5.24</td>
<td>3.99</td>
<td>0.15</td>
<td>29.28</td>
<td>0.1257</td>
<td>0.0028</td>
<td>0.460</td>
</tr>
<tr>
<td>1775.6</td>
<td>1.155</td>
<td>1.152</td>
<td>2.01</td>
<td>0.20</td>
<td>5.50</td>
<td>5.38</td>
<td>4.10</td>
<td>0.17</td>
<td>34.64</td>
<td>0.1488</td>
<td>0.0038</td>
<td>0.491</td>
</tr>
<tr>
<td>890</td>
<td>1.112</td>
<td>1.106</td>
<td>1.93</td>
<td>0.19</td>
<td>4.56</td>
<td>4.39</td>
<td>3.34</td>
<td>0.19</td>
<td>53.17</td>
<td>0.2283</td>
<td>0.0045</td>
<td>0.579</td>
</tr>
<tr>
<td>395.2</td>
<td>0.898</td>
<td>0.894</td>
<td>1.56</td>
<td>0.21</td>
<td>3.61</td>
<td>3.48</td>
<td>2.65</td>
<td>0.18</td>
<td>40.79</td>
<td>0.1752</td>
<td>0.0110</td>
<td>0.590</td>
</tr>
<tr>
<td>247.4</td>
<td>0.923</td>
<td>0.918</td>
<td>1.60</td>
<td>0.26</td>
<td>3.23</td>
<td>3.10</td>
<td>2.36</td>
<td>0.18</td>
<td>40.00</td>
<td>0.1718</td>
<td>0.0038</td>
<td>0.680</td>
</tr>
<tr>
<td>99.2</td>
<td>1.044</td>
<td>1.039</td>
<td>1.81</td>
<td>0.23</td>
<td>3.45</td>
<td>3.30</td>
<td>2.51</td>
<td>0.16</td>
<td>46.12</td>
<td>0.1981</td>
<td>0.0041</td>
<td>0.723</td>
</tr>
<tr>
<td>29.5</td>
<td>0.963</td>
<td>0.960</td>
<td>1.68</td>
<td>0.28</td>
<td>2.18</td>
<td>2.08</td>
<td>1.58</td>
<td>0.14</td>
<td>30.23</td>
<td>0.1298</td>
<td>0.0029</td>
<td>1.060</td>
</tr>
<tr>
<td>5.1</td>
<td>0.768</td>
<td>0.766</td>
<td>1.34</td>
<td>0.21</td>
<td>1.54</td>
<td>1.46</td>
<td>1.11</td>
<td>0.17</td>
<td>23.73</td>
<td>0.1019</td>
<td>0.0024</td>
<td>1.202</td>
</tr>
</tbody>
</table>

| 69      | 3676.5    | 0.404          | 0.70          | 0.10              | 1.94          | 1.88         | 1.43         | 0.21          | 19.60        | 0.0842       | 0.0025      | 0.491       |
| 55.84°N | 3637.3    | 0.336          | 0.58          | 0.12              | 1.92          | 1.86         | 1.41         | 0.16          | 19.15        | 0.0823       | 0.0023      | 0.414       |
| 48.09°W | 3589.5    | 0.438          | 0.76          | 0.11              | 2.15          | 2.08         | 1.58         | 0.19          | 21.39        | 0.0919       | 0.0024      | 0.482       |
| 3444.7  | 0.528     | 0.525          | 0.92          | 0.11              | 3.58          | 3.49         | 2.65         | 0.19          | 27.76        | 0.1192       | 0.0028      | 0.346       |
| 2951.8  | 0.581     | 0.578          | 1.01          | 0.11              | 5.75          | 5.64         | 4.29         | 0.23          | 33.80        | 0.1452       | 0.0032      | 0.235       |
| 2462.6  | 0.604     | 0.599          | 1.05          | 0.11              | 6.44          | 6.31         | 4.80         | 0.21          | 39.48        | 0.1696       | 0.0037      | 0.218       |
| 2168.1  | 0.855     | 0.850          | 1.49          | 0.12              | 6.03          | 5.90         | 4.49         | 0.22          | 40.95        | 0.1759       | 0.0039      | 0.331       |
| 1481.2  | 0.670     | 0.661          | 1.16          | 0.10              | 4.98          | 4.73         | 3.60         | 0.20          | 79.22        | 0.3402       | 0.0066      | 0.321       |
| 989     | 0.729     | 0.723          | 1.36          | 0.11              | 4.53          | 4.35         | 3.31         | 0.21          | 57.18        | 0.2456       | 0.0049      | 0.382       |
| 445.6   | 0.692     | 0.686          | 1.20          | 0.07              | 4.23          | 4.05         | 3.08         | 0.23          | 55.48        | 0.2383       | 0.0047      | 0.389       |
| 248.8   | 0.604     | 0.598          | 1.04          | 0.07              | 4.06          | 3.87         | 2.95         | 0.18          | 59.43        | 0.2552       | 0.0052      | 0.355       |
| 99.5    | 0.513     | 0.507          | 0.89          | 0.10              | 3.58          | 3.40         | 2.58         | 0.22          | 56.06        | 0.2408       | 0.0049      | 0.343       |
| 28.7    | 0.554     | 0.550          | 0.96          | 0.07              | 1.50          | 1.40         | 1.07         | 0.24          | 30.11        | 0.1293       | 0.0029      | 0.903       |
| 8.2     | 0.457     | 0.453          | 0.79          | 0.07              | 1.42          | 1.31         | 0.99         | 0.17          | 34.28        | 0.1472       | 0.0031      | 0.797       |
$^{230}$Th and $^{210}$Pa are dissolved $^{234}$Th and $^{214}$Pa activities corrected for the ingrowth from seawater $^{234}$U and $^{231}$U, respectively, since the time of collection following equations:

$$^{230}Th = ^{230}Th_m - ^{234}U \times (1 - \exp(-\lambda_{230Th} \times t))$$  \hspace{1cm} (1)

$$^{231}Pa = ^{231}Pa_m - ^{235}U \times (1 - \exp(-\lambda_{231Pa} \times t))$$  \hspace{1cm} (2)

$^{230}$Th and $^{210}$Pa are further corrected for detrital, U-supported $^{230}$Th and $^{210}$Pa concentrations as follows:

$$^{230}Th_{corr} = ^{230}Th_m - 0.6 \times ^{232}Th_m$$  \hspace{1cm} (3)

$$^{231}Pa_{corr} = ^{231}Pa_m - 0.046 \times 0.6 \times ^{232}Th_m$$  \hspace{1cm} (4)

where $^{230}Th_m$, $^{210}Pa_m$, and $^{232}Th_m$ are activities obtained from measurement; $^{235}U$ and $^{238}U$ are their average activities in seawater; $2324 \mu Bq/kg (112 dpm/1000l)$ and $45551 \mu Bq/kg (2801 dpm/1000l)$, respectively, obtained from $^{235}U$ activity of 39610 $\mu Bq/kg (2436 dpm/1000l)$ at salinity of 35 (Owens et al., 2011) and assuming natural $^{230}$U/$^{238}$U abundance ratio of 137.88 and seawater $^{234}$U/$^{238}$U activity ratio of 1.15; $\lambda_{230Th}$ and $\lambda_{231Pa}$ are decay constants of $^{230}$Th and $^{210}$Pa; $t$ is the time between sample collection and chemical separation of U from $^{210}$Pa and $^{230}$Th. 0.6 is the average $^{234}$U/$^{238}$U activity ratio in detrital material in the Atlantic (Henderson and Anderson, 2003) and 0.046 represents the $^{231}$U/$^{238}$U activity in seawater (Anderson et al., 1990). Half-lives for $^{210}$Pa, $^{230}$Th and $^{232}$Th are 32,760 yr, 75,584 yr and 1.40x10$^{10}$ yr (Robert et al., 1969; Cheng et al., 2013; Holden, 1990).
All errors are two standard errors including the contribution from sample weighing, spike calibration, $^{231}$Pa, $^{230}$Th and $^{232}$Th in the respective $^{235}$Pa and $^{230}$Th spikes, blank correction, internal precision and related corrections of mass spectrometric measurement.

S2. CFC-based Age determination

CFC measurements are not available for the GEOVIDE cruise itself. However, with the availability of CFC measurements from OVIDE section in 2012 and water mass composition estimated using extended Optimum Multi-Parameter (eOMP) analysis for both OVIDE and GEOVIDE sections, CFC-based ages can be derived for GEOVIDE section.

This allows the estimating the mean age of water masses using transient time distribution (TTD) method. A more detailed description of TTD method is given in other studies (e.g. Steinfeldt et al., 2009; Waugh et al., 2003). It is important to note that this mean age (referred to as CFC-based age hereafter and in the manuscript) is different from the age calculated based on atmospheric history of CFC (referred to as CFC apparent age hereafter), and therefore is not limited by the time span of the presence of CFC in the atmosphere and inherently age bias due to water mass mixing in CFC apparent age.

Combining CFC-based ages computed with the TTD method for each water sample with water mass composition estimated using eOMP analysis for OVIDE section in 2012 (García-Ibáñez et al., 2015), CFC-based age was calculated for each Source Water Type (SWT) defined in García-Ibáñez et al., 2015 by the equations,

$$\log[\text{CFC-based age}^j] = \sum_{i=1}^{12} \text{SWT}_i^j \times (\log[\text{CFC-based age}]_i) + \varepsilon_j$$

$$[\text{CFC - Age}^j] = \text{anti log}[\text{CFC - Age}]^j / \epsilon$$

where SWT$^j$ is the fraction of SWT $i$ to sample $j$ (obtained through the eOMP analysis); [CFC–based age]$^j$ is CFC-based age for each water sample computed with TTD method along OVIDE section 2012; and $\varepsilon_j$ is the residual, representing the portion of CFC-based age that can not be modelled by mixing of SWTs, i.e. the difference between log[CFC–based age]$^j$ and that obtained as the sum of the contributions by mixing of the individual SWT. $\sum_{i=1}^{12} \text{SWT}_i^j \times (\log[\text{CFC-based age}]_i).$

The output of log[CFC-based age], and its inversion ([CFC-age]) is given in Table S2. The squared correlation coefficient ($r^2$) and standard deviation of the residual, $\varepsilon$, are 0.94 and 0.12, respectively.

CFC-based age for GEOVIDE section was then calculated employing equation (5) with water mass composition estimated using eOMP analysis along GEOVIDE section (García-Ibáñez et al., 2018) and the output of CFC-based age for SWT (Table S2).
Table S2 Output of log(CFC-age) and the inversion [CFC-age] (i.e. CFC-based age) for source water types (SWT)

<table>
<thead>
<tr>
<th>SWT</th>
<th>log(CFC-age)</th>
<th>CFC-age</th>
</tr>
</thead>
<tbody>
<tr>
<td>ENACW16</td>
<td>1.05±0.20</td>
<td>11±5</td>
</tr>
<tr>
<td>ENACW12</td>
<td>1.11±0.03</td>
<td>13±1</td>
</tr>
<tr>
<td>SPMW8</td>
<td>1.69±0.04</td>
<td>49±5</td>
</tr>
<tr>
<td>SAIW</td>
<td>1.19±0.07</td>
<td>16±3</td>
</tr>
<tr>
<td>SPMW7</td>
<td>1.26±0.05</td>
<td>18±2</td>
</tr>
<tr>
<td>IrSPMW</td>
<td>0.98±0.03</td>
<td>10±1</td>
</tr>
<tr>
<td>LSW</td>
<td>1.54±0.02</td>
<td>35±1</td>
</tr>
<tr>
<td>MW</td>
<td>1.96±0.04</td>
<td>91±8</td>
</tr>
<tr>
<td>PIW</td>
<td>1.33±0.15</td>
<td>22±8</td>
</tr>
<tr>
<td>DSOW</td>
<td>1.22±0.07</td>
<td>17±3</td>
</tr>
<tr>
<td>ISOW</td>
<td>1.70±0.03</td>
<td>50±4</td>
</tr>
<tr>
<td>NEADWL</td>
<td>3.00±0.02</td>
<td>989±48</td>
</tr>
</tbody>
</table>

ENACW16 and ENACW12 = East North Atlantic Central Water of 16°C and 12°C; SPMW8, SPMW7, IrSPMW = Subpolar Mode Water of 8°C, 7°C and of the Irminger Sea; SAIW = Subarctic Intermediate Water; MW = Mediterranean Water; PIW = Polar Intermediate Water; ISOW = Iceland–Scotland Overflow Water; LSW = Labrador Sea Water; DSOW = Denmark Strait Overflow Waters; and NEADWL = Lower North East Atlantic Deep Water.

The output of the inversion [CFC-age] (i.e. CFC-based age) for source water types (SWT)

\[ r^2 \text{ and } \text{std(Resid)} \text{ represents the squared correlation coefficient and standard deviation of the residual, } \epsilon \text{, i.e. the difference between } \log \text{[CFC-based age]} \text{ and that obtained as the sum of the contributions by mixing of the individual SWT, } \sum_{i=1}^{12} \text{SWT}_i \times (\log \text{[CFC-based age]})_i. \]

S3. Scavenging-mixing model and parameterization

A more detailed description of the scavenging-mixing model used in this study is given in Moran et al. (1997). Briefly, the model takes into account reversible scavenging of the nuclides and water mass mixing. It describes the evolution of nuclides through time of a one-dimensional system, an ocean water column. In the Atlantic, the system is assumed to start at time \( t=0 \) in the far North Atlantic and moves southward with time. Transport of material downstream relative to the direction of water flow is permitted, to represent the effect of scavenging of radionuclides by sinking particles. Lateral exchange with water outside of the 1-D system is not permitted.

The equation to derive the dissolved concentration of each nuclide follows that of Moran et al., (1997). Dissolved concentration of the nuclide is given by:

\[ c_d = \frac{c_{\text{prev}}+P}{(K_d SPM+1)} \times \left[ 1 - \exp\left(-\frac{(K_d SPM+1)}{S K_d \tau_{\text{w}} SPM} \times \epsilon\right) \right] \]
where $C_d$ is the dissolved concentration of the nuclide; $P$ is the production rate of $^{230}$Th and $^{231}$Pa, 0.42 µBq/kg/yr ($2.57 \times 10^{-2}$ dpm/1000l/yr) and 0.039 µBq/kg/yr ($2.37 \times 10^{-3}$ dpm/1000l/yr), respectively; $K_d$ is the distribution coefficient of the nuclide; $\lambda$ is the decay constant of the nuclide; $C_{pre}$ is the preformed concentration of $^{230}$Th or $^{231}$Pa; SPM is the suspended particle concentration; and $S$ is the particle settling speed, which represents the net effect of particle sinking, disaggregation and aggregation; $T_w$ is water mass age; $z$ is the water depth.

Initial parameterization was conducted using $S=500-1000$ m/yr, $K_{dTh}=1 \times 10^7$ ml/g, $K_{dPa}=5 \times 10^5$ ml/g, SPM= 20-50 µg/l, for preformed concentrations set at 0 and surface average from GEOVIDE, i.e., $C_{preTh}=C_{surface average}=1.66$ µBq/kg, $C_{prePa}=C_{preTh}=1.31$ µBq/kg. With $T_w$ known from CFC measurements for every depth where $^{230}$Th and $^{231}$Pa was measured along GEOVIDE section, water-column profiles of both nuclides were simulated for GEOVIDE station 13 and the parameters were adjusted for the best fit between the simulated and observed profiles (Fig. S1). This gives us the optimized parameters for the analysis in discussion section 4.3, which are listed in Table S3. Our optimized parameters are consistent with values reported by other studies (also listed in Table S3).

Adopting the optimized parameters and setting preformed component ($C_{pre}$) equal to the nuclide concentrations observed in the upper 100 m of the GEOVIDE section, the modelled evolution of nuclide concentrations with age between 0-500 years at 2000 m and 3500 m water depths, together with GEOVIDE data, is plotted in Figure S2.

**Figure S1:** Modelled (dashed black lines) profiles with preformed value set at 0 and surface average concentration from GEOVIDE, and observed (solid blue lines) profiles of and 13 from GEOVIDE section. The best fit was first sought for $^{238}$Th, which gives us the optimized parameters $S$, SPM and $K_{dTh}$. These parameters were then adopted for the simulation of $^{231}$Pa profiles, adjusting only $K_{dPa}$ to obtain the best fit.
Figure S2: Results from a scavenging-mixing model of $^{230}$Th, $^{231}$Pa, Dissolved $^{231}$Pa-$^{230}$Th and Particulate $^{231}$Pa-$^{230}$Th compared to observations. Preformed concentration ($C_{\text{pre}}$) were set at 0 (dashed line) and at the average surface concentration ($C_{\text{surface average}}$) from GEOVIDE section (solid line), i.e. $^{230}$Th = 1.66 µBq/kg, $^{231}$Pa = 1.31 µBq/kg.

Deleted: Figure S1: Modelled (dashed black lines) profiles with preformed value set at 0 and surface average concentration from GEOVIDE, and observed (solid blue lines) profiles of station 1 and 13 from GEOVIDE section. The best fit was first sought for $^{230}$Th, which gives us the optimized parameters $S$, SPM and $K_{d}^T$. These parameters were then adopted for the simulation of $^{231}$Pa profiles, adjusting only $K_{d}^P$ to obtain the best fit.
Table S3 Parameterization of the scavenging-mixing model

<table>
<thead>
<tr>
<th>Parameter</th>
<th>This study</th>
<th>Literature</th>
</tr>
</thead>
<tbody>
<tr>
<td>S (m/yr)</td>
<td>800</td>
<td>500-1000 (Moran et al., 1997)</td>
</tr>
<tr>
<td>SPM (µg/l)</td>
<td>25</td>
<td>30 (Labrador Sea, Brewer et al., 1976)</td>
</tr>
<tr>
<td>$K_{h}^{S}$ (ml/g)</td>
<td>$1.1 \times 10^{7}$</td>
<td>$1.1 \times 10^{7}$ (Moran et al., 1997)</td>
</tr>
<tr>
<td>$K_{h}^{Pa}$ (ml/g)</td>
<td>$1.4 \times 10^{6}$</td>
<td>$2.2 \times 10^{6}$ (pure carbonate); $1.4 \times 10^{6}$ g/g (pure opal) (pseudo-$K_{a}$, Chase et al., 2002)</td>
</tr>
</tbody>
</table>

S4. Uncertainty analysis of the assessment of the scavenging of $^{230}$Th and $^{231}$Pa

Both the uncertainty of the CFC-based ages and the fact that the preformed values of $^{230}$Th and $^{231}$Pa are unknown contribute to the uncertainty of the scavenging of $^{230}$Th and $^{231}$Pa. Here we analyze how these two factors influence the assessment of the scavenging of $^{230}$Th and $^{231}$Pa.

1. CFC-based ages

Uncertainties (1 standard error) associated with CFC-based age range between 11-40%. This uncertainty leads to an average uncertainty of 23% and 13% in potential total $^{230}$Th and $^{231}$Pa respectively, corresponding to an average uncertainty of 30% in the scavenged component of $^{230}$Th and 40% in the scavenged component of $^{231}$Pa.

2. Preformed values

As the exact values of preformed values are unknown, we analyse the sensitivity of the scavenging component to this factor. A two-fold increase in preformed values results in an increase by a factor of 1.2 and 1.6 in the total potential component of $^{230}$Th and $^{231}$Pa, respectively, leading to an increase by a factor of 1.2 in the scavenged component of $^{230}$Th and of 2.6 in the scavenged component of $^{231}$Pa. Clearly, the scavenged component of $^{231}$Pa is more sensitive to the preformed component compared with the scavenged component of $^{230}$Th. This difference however becomes less significant when comparing the scavenging component to the potential total component, i.e. the ratio of scavenged/potential total, with an increase in the ratio of scavenged/potential total by a factor of 1.1 and 1.4 for $^{230}$Th and $^{231}$Pa, respectively. This sensitivity analysis suggests that a better knowledge of preformed values will benefit the assessment of the scavenging of both nuclides.

S5. Meridional transport of $^{230}$Th and $^{231}$Pa in the Atlantic
Table S4 Mass balance calculation of meridional transport of $^{230}$Th and $^{231}$Pa in the Atlantic

<table>
<thead>
<tr>
<th>Net meridional transport</th>
<th>Volume of seawater between two latitudes</th>
<th>Production in water column</th>
<th>Removal to sediment</th>
<th>Removal/Production</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{230}$Th $\times 10^9$ µBq/s</td>
<td>$^{231}$Pa $\times 10^7$ m$^3$</td>
<td>$^{230}$Th $\times 10^8$ µBq/s</td>
<td>$^{231}$Pa $\times 10^6$ µBq/s</td>
<td>$^{230}$Th $\times 10^8$ µBq/s</td>
</tr>
<tr>
<td>GEOVIDE-4.5°S</td>
<td>-7.76</td>
<td>-4.75</td>
<td>1.48</td>
<td>200.8</td>
</tr>
<tr>
<td>4.5°S-45°S</td>
<td>0.33</td>
<td>0.017</td>
<td>1.02</td>
<td>138.4</td>
</tr>
</tbody>
</table>

Positive value indicates northward transport; negative value indicates southward transport. Production rate of $^{230}$Th and $^{231}$Pa in water column are 0.42 and 0.039 µBq/kg/yr, respectively.

References


Deleted:

Deleted:

Deleted:


Deleted:

Deleted: