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Interactive comment on "A global scavenging and circulation ocean model of thorium-230 and protactinium-231 with realistic particle dynamics (NEMO–ProThorP 0.1)" *by* Marco van Hulten et al.

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1 Response to reviewer #1

Thank you for your critical and useful comments on our manuscript. We have addressed the issues you have raised.

Concerning the term "realistic", this may be overstated; in any case, it should not have been stated as an absolute but rather a relative notion, as we do in the introduction ("more realistic"). In the title, we have replaced "realistic" for "improved"; as this is a relative term, and does not have the weight of "realism", ontologically or elsewise.

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1.1 Particle concentration effect

> While Hayes et al., 2015b (cited in the paper), found strong evidence for a particle concentration effect on K_d values in the Geotraces data sets from the North Atlantic Ocean, that is ignored in the model. The particle concentration has been documented numerous times (not just by Honeyman and Santschi, 1989, cited in Hayes et al., 2015b) for radioisotopes of Th and other elements, and is ascribed to the existence of sorbing and complexing colloidal organic matter in the filter-passing fraction. This then causes linear correlations of the log of the particle-water partition coefficient (K or K_d) vs. the log of the suspended particle concentration, as well as linear correlation of the log of the sorption rate constant vs. the log of the suspended particle concentration. It would gather that this should be pretty straightforward to incorporate that in the model. This would be important, especially if they intend to model boundary scavenging and nepheloid layer scavenging, where particle concentrations would be expected to be higher, and associated K_d values lower.

The authors are aware of the evidence for K_d values to depend on the particle concentration; we should have discussed and considered this more carefully. Indeed, especially where particle concentrations are high, as near the boundaries and in nepheloids, scavenging would be too high if they were included in our model. In the paper, we advise that future versions include those particles, and we now also note that the particle concentration effect should be considered.

The inverse correlation between $\log(K_d)$ and log(particle concentration) strongly relies on the K_d observed for very high particle concentrations. In the model presented here, particle concentrations of the different phases vary by a factor of 10 or so, so the impact on the K_d value is limited and likely within the spread of the values inferred from the data.

1.2 Choice of partition coefficients

In our model, instead of making K_d explicitly dependent on the particle concentration, there are actually different particle sizes, albeit the minimal of two. We have tested different K_d values for Th-230 and Pa-231 for a total of six particle types (small and big POC, opal, calcium carbonate and small and big lithogenic particles). Careful thinking is needed to see where more detail must be added. The particle concentration effect is certainly one to think of, but more particle types and size classes may be at least as important, or at least, in our opinion, those should be the next step in model development as the mechanisms are better understood.

We have also constrained the K_d values in terms of internal consistency. For instance, considering that smaller particles have a larger effective surface, the ratio of K_d's between small and big need to be the same for Th and Pa, at least of a given particle type. The choice for the relatively small K_d values (which are close to the range of (Geibert and Usbeck 2004)) is mostly because of the fact that we only have fast-sinking opal and calcium carbonate particles that would export too much Th and Pa if we were to set a higher value. We did not fully realise that this is a misuse of the results of (Geibert and Usbeck 2004); we are more clear about this now. We checked with W. Geibert that the authors *did* clean their samples, as is described in their methods section, giving rise to your criticism (W. Geibert, pers. comm., January 2018).

1.3 Other comments

> Also, the nomenclature and units used in this table are non-conventional. At least, it should be mentioned what the unit of K_d (Mg/g) in Table 2 is, i.e., $10^6\ gwater/gparticles.ItisnotobviousasMcanbeusedfor10^3$ and for 10^6.

“M” stands for one million (BIPM et al. 2008). We agree that it should be mentioned that it is mass of water per mass of particle, so we will amend this.

> In Table 3, they review the very limited data sets that they used for their model. By doing that, they omit data sets from many other researchers and oceans. For example, the Th and Pa nuclide data from the Gulf of Mexico (Roberts et al., 2009), or the 230Th and 234Th data sets from the North Atlantic by Guo et al. (1995), which also exhibit the common particle concentration effect, could have been used as well.

A wider range of data sources could be very useful, but we chose to only use a couple, typically more recent and larger, data sets. However, we mostly used what we came across, especially from the GEOTRACES programme, and this we deemed sufficient for a basic analysis of this first model. In the future we should see to using more data; any ready compilations of data in a convenient format would be especially handy.

> Table 4 is totally confusing and to some extent, mislabeled, and needs a better caption stating what the numbers in each column signify, and their units.

The unit is percentage for all fields with respect to all particle phases; we make this more clear now. We added a bit of linespace at the right place for an improved interpretation.

> It should therefore be more clearly stated in the manuscript what they are doing with a very limited data set from which they chose their adjustable parameters. Thus, using more realistic values of K, for example, the simulations might be different.

We have tried simulations with different values of K: they give different results indeed. Dutay et al. (2009) and others have presented several sensitivity simulations with different K's. We don't see need to repeat those. We tried to seek a compromise between "realistic simulation" and reported values. We have the impression that reported values of the K values are uncertain, and that our choice of values are still within that uncertainty. This is a somewhat subjective uncertainty in that it is bigger than reported standard deviations, because values found in the different studies often fall outside their respective internal distributions. From this we took the liberty of choosing convenient values. We have preformed one sensitivity test to investigate its impact on the Pa/Th ratio.

> Another limitation is that only towards the end of the paper is it stated that the large/small particle designation is using a 50 μm boundary. It should then also be considered that not everywhere in the ocean do the large particles sink fast, and the small ones slow. For example, Xu et al. (2011) and Hung et al. (2010, 2012) demonstrated through thorium radioisotopic evidence and microscopic analysis of sediment trap particles collected in oligotrophic waters in the Gulf of Mexico and the Pacific Ocean that the fast sinking particle assemblage is mostly composed of smaller than 50μm diatom particles. Thus, without considering this detailed data set, the modeling of van Hulten et al. is, to say the least, unrealistic.

We agree that all sinking particles are not necessarily in the > 50 micron size range. This limit is often set as an operational and somewhat arbitrary limit. However, it makes sense to keep a distinction between suspended/slowly sinking particles (defined operationally as > 0.4 micron or so) and large/rapidly sinking particles (Bacon et al. 1985). (Hung, Gong, and Santschi 2012) obtained their conclusions by comparing POC fluxes determined in sediment traps and approaches based on size-fractionated suspended particulate ^{234}Th and POC concentrations. While they question the use of 50 microns to separate small and large particles, they do not question the occurrence of two distinct behaviors. In addition, (Hung, Gong, and Santschi 2012), focused on ^{234}Th in shallow waters. ^{234}Th and ^{230}Th have similar chemical properties, they are usually studied in different environments (shallow waters versus deep waters) so that they interact with different particle types and do no correlate with the same elements/phases in trapped particles (Roy-Barman et al. 2005). Hence, it may be difficult to extend the results of Hung et al. to particles in the deep ocean.

2 Response to reviewer #2

2.1 Recent model studies

> It is remarkable that within 2017 alone three papers on implementing and applying 231Pa and 230Th isotopes into models have been presented [. . .] I just want to encourage the authors to get into contact with the modellers working on CESM1.3 and Bern3D and others and may think about starting such an inter-calibration project.

This would be an interesting idea, and such a collaboration could also result in new model improvements. Until now it was better to let each group develop its own strategy to simulate Pa and Th in order to encourage creativity, but now that several approaches have been proposed, it would be interesting to intercompare their results, for instance in the context of the GEOTRACES programme.

> General: It seems like the submission of this manuscript felt into the time range of the publication of the Guo and Liu paper. Although it was under open discussion since April, the authors may have missed it. Anyway, they

> definitely should incorporate the findings of Guo and Liu. As well they could provide a short summary (maybe as a table) on all of these recent model papers and what is different with their approaches, how particle fields are generated, how circulation, the range of adsorption/desorption coefficients etc.

We did see Gu and Liu (2017) when it came out as a discussion paper. Now that it has passed peer review, we will note their study. We believe it would be better if a future study would compare the different models in detail, possibly as part of an inter-calibration project; because that would express a more neutral point of view (and there will have been time for reflection).

2.2 Other comments

> page 1: title: I'm not sure about using the term "realistic particle dynamics" in the title. It slightly implies that previous studies have applied unrealistic particle dynamics, which might true for a few only. What about "new" or "refined" instead of realistic?

We agree that "realistic" is not the best qualification here. We will replace this word for "improved", because this is, at least for POC, spot on. Lithogenic particles are *new* in the model, but I would call the particles as a whole, including the changes in POC and calcium carbonate, *improved*.

> p2, l133: I think Siddall07 is based on Bern3D.

You are right; we will fix this.

> p3,l1: what is much too low? Please give numbers/factors.

We now do this.

> l23-26: these sentences seem redundant from p2 and 1

We will reorder and removed text.

> general: please make sure to introduce all used abbreviations. E.g. table 1: OPA?

Repaired.

> table 1: large or big ? Called “big” before.

We now call the big particles “big”, consistent with the mathematical notation.

> Fig1 and 3: I’m very sorry but my copy of the manuscript does not show the whole contents of these figures. According to the caption I miss a considerable part of the concept. I tried on a Windows and Linux system.

The “picture” environment, though part of the Copernicus LaTeX package apparently gives problems in presenting the resulting PDF on at least some systems. It is not acceptable if people cannot see the full figures, so we will compile separate PDFs from the figures and add them in the usual way in the document, now using the better suited TikZ package.

> p5, l11: Hauglustaine04 is a model approach. Why not using a satellite based particle flux?

We used a model approach for internal consistency. (Hauglustaine et al. 2004) gives a representation of dust deposition, whereas satellite based particle fluxes still need to be converted to a air–sea dust flux (through modelling). Furthermore, Hauglustaine et al. (2004) was readily available on the ORCA2 grid, and has been used for multiple studies (which would also give easy inter-comparison between different NEMO-based models using this deposition field). Given the high uncertainties in dust deposition, we decided that Hauglustaine et al. (2004) would be good enough for our purpose.

> Fig2: why are dust depositions explicitly shown, but not other fluxes? Whats the transfer function between fig 2 and fig 5d

There are no other fluxes because other particles are biogenic and generated by the biogeochemical PISCES. We run simulation imposing flux from Fig. 2 and in the first vertical level of the model, the distribution shown in Fig. 5(d) is obtained for dust when transport is applied. More concisely, running the model with the monthly flux of Fig. 2 and Eqs (1) (lines 83 and 84 of `trcsms_lith.F90`) and (2) (lines 97—100) results in Fig. 5(d).

> Table 2: I may have missed it, but why are the factors of the partition coefficients between small and big 5 for POC and 10 for lithogenics?

Since the idea is that the effective surface area is much larger for small particles compared to big, this indeed raises the question why it is 10 times much larger for Lith compared to 5 times larger for POC. Kinetics of adsorption is different for the different types of particles, so the ratio of K for big versus small particles can be different as

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well. We chose the factors 5 and 10 because the gave good results. But is it reasonable? We decided to fix, for a given particle type, a partition coefficient ratio for the different size classes that is the same for both Th and Pa, with the underlying idea that the relative amount of adsorption spots for large versus big should be the same for any metal. Between different particle types, however, we were more lenient, in that we don't assume anything about the surface structure of any particle, thus small versus big could work differently for lithogenic versus organic-carbon particles.

> p9, l14: as mentioned before it might be helpful to give the range of the reported values.

It is difficult to give a precise range of that, because different methods are involved in the different observational studies. During the early development of the model we have documented a range based on the values given by Hayes (2015), Geibert and Usbeck (2004) and several modelling studies. We will present those in a supplement to the paper. We think it is better to leave a more detailed analysis and discussion to a different study. For the present study, our effort went to the model framework, which is the embedding of the basic Th/Pa model (ProThorP) in NEMO and the passive-tracer component TOP.

> p9, l18: when it comes to particles sizes I consider Kretschmer et al. (2008) as an appropriate reference.

This is a presentation, but we found their useful paper on the same subject (doi:10.1016/j.gca.2011.09.012). We've added it in the Discussion.

> table 3: here I have a major concern. The model is compared to an arbitrary core top data set. The Oxford data has a global coverage, but the data

> is quite old and has been measured by alpha- and beta- counting which comprises large uncertanties. I know that there is little new data from the Pacific and the Southern Ocean, but when it comes to the Atlantic or the Arctic Ocean it would be recommendable to compare the model not only to the two Burckel studies. E.g.:(Bradtmiller et al., 2014; Christl et al., 2010; Hall et al., 2006; Hoffmann et al., 2013; Jonkers et al., 2015; Lippold et al., 2016; Luo et al., 2015; Negre et al., 2010; Roberts et al., 2014) (Rutgers van der Loeff et al., 2016; Voigt et al., 2017). In general it is not clear to me, how the goodness of the model versus observations is assessed (e.g. later on page 14). Wouldn't it be helpful to give some statistics?

We think that the data used is sufficient for this study: it has a good global, especially Atlantic, coverage for a first comparison. It would be useful if a new compilation, put together by "data people" for the highest quality and correct interpretation, were to come available. We are using something that is probably okay for a first comparison, and it would take a disproportionate amount of time to include such data at this time. The same is true for the statistics: we do not think it would add much to the visual sediment plots. The sediment plots are already additional to the core of this model description paper, which was to present the model. In a model evaluation paper, or in a more regular oceanographic study, a statistical analysis and more up-to-date dataset would be much more important.

> p11,fig.4: later in the text we learn that the model gives too high 231Pa/230Th values (e.g. Fig. 8d). But given that the circulation scheme of Fig. 4 is not realistic (too shallow northern overturning) this is not surprising. What parameters are responsible for creating such a shallow overturning, which reminds me rather of a glacial circulation scheme?

Low resolution models, like with the ORCA2 configuration (2°) we use, a shallow overturning is a common issue. At low resolution, models cannot realistically simulatate small-scale features like vertical mixing, which results in a too shallow overturning. We still used these circulation field because they are “the best” we have for the ORCA2 configuration, in the sense that they are used and discussed in many other studies, e.g. Gehlen et al. (2007), Arsouze et al. (2009), Dutay et al. (2009), Aumont et al. (2015). Probably, it is best if models of higher resolution are considered, but that is expensive in combination with a full biogeochemical model and we considered this from the beginning out of the scope of this study.

> p11, l 10: “. . .has a strong overturning. . .” relative to what?

Relative to some estimates. Even though it is not precisely known what the transport in the lower cell is, the model quite certainly overestimates it (citation will be added to the paper). The reasoning starts with that we are using a model with a relatively low resolution, which typically have a too shallow (upper) overturning (as the one we use does). Because of this, there is a much larger depth range in which the lower overturning cell can transport AABW northward, resulting, at least in our case, in an overestimated flux.

> Fig. 5: What are the observations shown here based on? Please give references in the caption. I’m sure there are more observations available than shown here. The color codes seem to be scaled by arbitrarily increasing numbers. However, more important, the model generates impressively realistic particle fields, but what about this high production region off Argentina?

Information on particle sources included in caption, and more information in-text. We are not aware of observations of particle concentrations that could improve our coverage. Lam, Doney and Bishop (2011) collected (consistent) data from previous studies, and should cover all data up to 2011. Since then the GEOTRACES programme has new data at the GA03 transect, but we don't know of other data. For instance, to our knowledge there are no surface concentrations of biogenic particles available in the Argentine Basin. The Argentine Basins is a specific region of high productivity with a large continental margin; of course, it would be nice if we could validate our model to concentration measurements.

> p12, l 9-11: I don't understand this sentence.

We will rephrase part of this paragraph for clarity.

> Fig. 8: I appreciate the overall good agreement with observations. But, as mentioned before, the deep could be much better with a realistic overturning scheme.

Yes.

> Fig. 10: caption: "Concentrations.." of what? Please provide units.

Units are added now.

> Table 4/p 16, l 3-4: Maybe more explanation needed here on the difference between "stock" and "particle flx". What creates the huge difference for both between bSiO2 and Litho.?

Only for POC and Litho do we have slowly sinking particles on which much of the stock is stuck, whereas the big affinity of Pa to bSiOâĆĆ mostly results in a strong export (and similarly for Th on CaCO3). We will add this to the text.

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> Fig. 11 and Fig. 13: Please use same scale for both plots (0.0 to 0.30 at max. Higher values are unrealistic). As mentioned before, please use an up-to-date data base for the Atlantic. Further it would be helpful to show the Atlantic model and observations in a 231Pa/230Th vs. depth plot like done by Gherardi et al. (2009) (their figure 5).

We have set the same colour scales with the maximum at 0.30 (and with a "Δ" for higher, unrealistic values).

In the figures presented at the end of this document, we show the dissolved Pa-231/Th-230 concentration in the seawater at the meridional GA02 (and Drake Passage), and the North Atlantic GA03 transects. Several of the patterns in the observations are recognisable in the model, but there are discrepancies as well. As of yet we don't have any interpretations of this that would give more insight in the model and ocean processes than what we got from the other figures. We could perhaps add them to an electronic supplement.

> p20: [. . .] I want to suggest that the data mentioned in the "second line 7" (hydrothermal) could be indicated by different symbols in Fig. 12. This would enable to see where the model fails and where not.

We will try to improve the scatter plots.

> chapter 6.1: the "weaker" affinity of Pa to opal is a very interesting finding. In order to strengthen this point it could be helpful to provide an overview of all the values proposed by the different studies, in particular by describing if it was a lab-experiment, in situ observation (where) or the best fit for the model.

We agree that this would be useful, but it is out of the scope of this study. The apparently weaker affinity of Pa to opal was meant as an additional illustration of the model; we do realise that it does warrant a more extensive study, and we hope that our illustration does not stop anyone from doing this in a rigorous way.

3 Response to reviewer #3

3.1 Scientific Quality

> 1. I do not believe that one of the central assumptions is justified. You assume that the absorption and desorption rates are much faster than the settling of the absorbed phases. This is marginally true for small particles sinking at 2 m/day, but is not at all true for particles sinking at 50 m/day. [. . .]
>
> The authors must either justify in detail why the assumption is valid (and thus explain why my analysis is in error) or include the absorption/desorption process.

ProThorP's aim is to scavenge and vertically transport Pa-231 and Th-230 while ocean water masses are flowing around to describe mainly (1) the amount of dissolved Pa and Th left in the water column and (2) the Pa/Th ratio accumulated in the sediments. It is clear that the equilibrium between dissolved and large particles is convenient for computing but not necessarily realistic in terms of processes at the scale of a single large particle. One idea, which is not in the equation and that might not be verified is that no "true" (chemically speaking) equilibrium between the dissolved phase and big particles exists, but that aggregation of small particles (probably through grazing) ensures the transert of Pa and Th from the solution to the large particles (Bacon and

Anderson, 1985). But this is not the real issue. The issue is to describe Pa–Th fluxes at the water column scale. Hence, when small and large particles are present (litho, POC), the K_d for the big particles is reduced compared to small particles so that large particles do not completely deplete the water column in Th and Pa. Similarly when only large particles are present (e.g., small carbonate, which is certainly not realistic either), the K_d(Th) is reduced compared to the values determined experimentally by Hayes et al. to ensure realistic fluxes.

This “fast equilibrium” hypothesis is certainly the reason for the very low dissolved ^{230}Th concentration modelled in the surface water (Fig. 9). This is equally due to small and big particles and not of great importance because we are not particularly interested in the surface water distribution or Pa and Th.

2. Why did you chose 20% small and 80% large for your lithogenic particles?

We tried 20 and 80% partitioning of dust into the lithogenic ocean particles, and this happened to work well. As explained in the manuscript, dust particles must aggregate in the upper ocean to get anywhere close to the concentration of large lithogenic particles observed in the ocean. We quickly settled on the 20/80%, because tuning would overestimate the dust source where we are missing resuspended particle sources. Further refinement should therefore be developed together with those other sources.

3. pg 16, last line : “These discrepancies ... arise from the different speeds for each type of particle” But don’t all big particles sink at the same rate? I think you could make this more clear but adding columns to Table 4: small Litho, big Litho, total Litho etc.

We think that would be too much information. We have clarified the interpretation in the text; we hope that it is more clear now.

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4. pg 22 Lines 10-20: also consider including Luo et al (2010) in this discussion as their simple model allows one to see the impact more easily

Yes, we now refer to them in this context.

3.2 Scientific Reproducibility

1. How long was the NEMO-OPA model run? Was a single year of forcing used repeatedly? If so, what year? If not, what years of forcing were used?

The simulation was produced with climatological conditions. Thus, there is no interannual variability in the forcing dataset. The model has been run for 200 years and the last year has been selected. Since the model was forced by climatological conditions, there is no interannual variability in the ocean dynamics that is simulated except for the internal variability which at 2° resolution is negligible almost everywhere except perhaps in the equatorial regions. Thus, choosing whatever year is valid.

2. Was an ice model included?

Yes, this was a simulation with NEMO–LIM.

3. Please include the fourth order equation for CaCO 3 dissolution.

Done.

4. I don't think you actually code equation (5). If I'm correct, please give the equations you actually code.

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We have not coded it exactly like this, but there will always be a translation step between the mathematical model and the implementation. The production (β) and decay ($-\lambda A$) of the radioisotopes are defined in `trcsms_protac.F90` and are straightforwardly transcribed from the equations. The sinking term ($w\mathrm{d}A/\mathrm{d}z$) is in `p4zsink.F90`, and has some subtleties, but does implement this term. Finally, the circulation including eddy-induced velocity (in the full derivative) as well as the diffusion terms ($\mathcal{A}$ for horizontal and $\mathcal{B}$ for vertical) are in OPA, and one will get the right routines through a full check-out of the NEMO repository. Since we are running PISCES and the added passive tracers off-line with respect to OPA, should we better not write out the dynamical terms. We could then just mention again that the tracers are advected by an off-line circulation field (same for eqn 2). Please, advice.

5. Under Simulations, first sentence "The model" please specify that this is the PISCES+tracer model.

Done.

6. Please include a run table, showing your physical run, your spin-up and your two analyzed runs.

We have not included a table, because there is only one sensitivity simulation, which is not central to the paper. There is one base simulation of Pa-231 and Th-230, and the dynamics is a forcing that is evaluated and used in multiple studies before.

7. Link biogenic silica and opal, in case your readers don't understand you are using them interchangeably.

Instead of linking them, we decided that we will replace the couple of occurrences of "opal" with "biogenic silica".

8. Where does one get the new scavenging model?

Near the end of the paper there is a section on code availability. There is the link to the code (https://zenodo.org/record/1009065). This archive contains updated PISCES routines, lithogenic particles and the Th-230 and Pa-231 code (including scavenging).

3.3 Presentation Quality

1. pg 1, l 19 “and the carbon”
2. Fig 1 “triple”
3. pg 5, l 7 “corresponding to”
4. pg 5, l 14 “(e.g. Van Hulten et al, 2013)”
5. pg 9, l 22 “a large number of measurements”
6. pg 9, l 23 “on this transect”
7. Table 3: over run on second last line.
8. pg 11, l 5 drop “thereof”
9. pg 11, l 11 “water sinks down”
10. pg 11, l 12 “Antarctic”
11. pg 12, l 10 “whereas we only have small particle data for the Atlantic Ocean”
12. pg 14, l 6 drop “as well”
13. pg 15, l 14 “Globally in the model,”

14. pg 16, l 12 “discrepancies”

15. pg 19, l3 or 18 depending how you count! “POC accounts for only”

Thank you for these corrections! We applied the changes.

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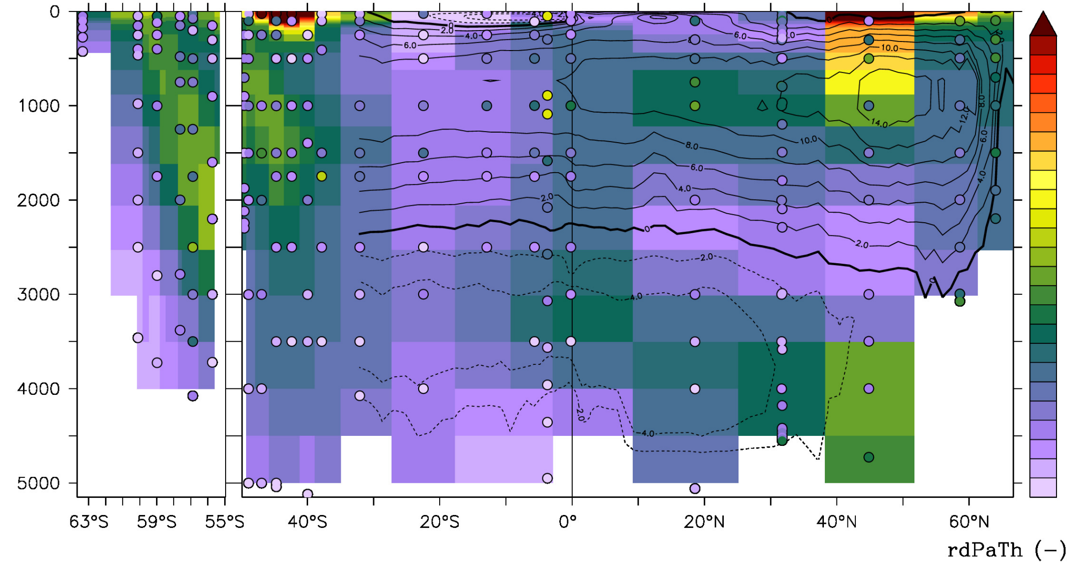

Fig. 1. West Atlantic GA02 transect, dPa/dTh

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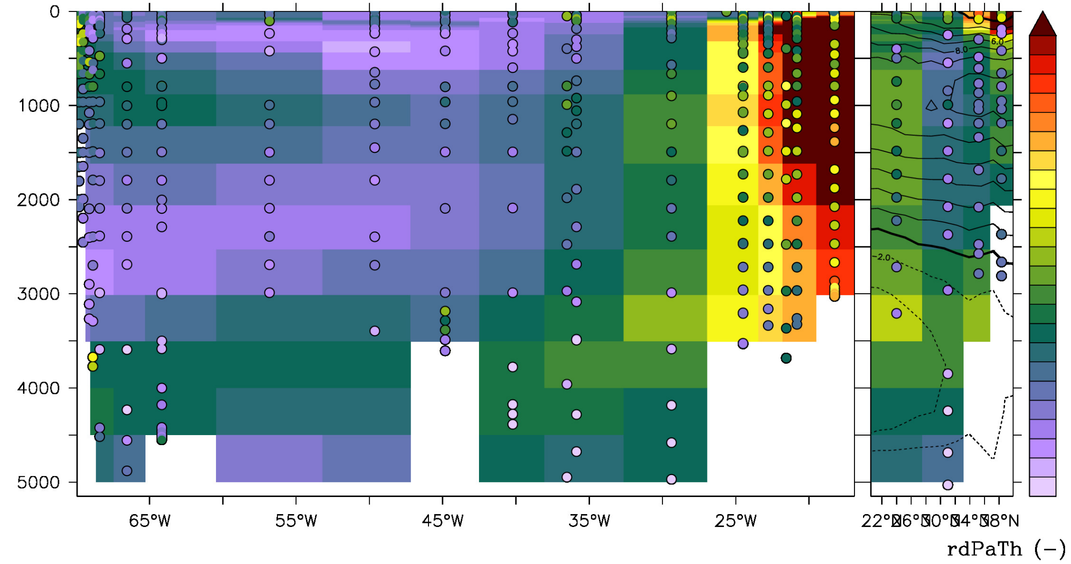

Fig. 2. North Atlantic GA03 transect, dPa/dTh

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