AGU PUBLICATIONS

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Supporting Information for

Radium Isotopes across the Arctic Ocean show Time Scales of Water Mass Ventilation and Increasing Shelf Inputs

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Introduction

Here we give supporting information of

1 Model equations

Differential equations describing the ^{228}Ra-^{228}Th profile model. Parameters are listed in Table 1 of the paper.

2 Analytical procedures and intercalibration.

Figure S1 gives the results of four isotopes analysed at the GEOTRACES cross over station in the Makarov Basin on board USGSC Healy (1-9-2015) and 2 weeks later on board RV Polarstern (14-9-2015). The offset in ^{226}Ra likely results from the fact that the HLY1502 analyses were performed on discrete samples whereas the PS94 analyses depended on the estimate of the cartridge collection efficiency.
Figure S2 compares total ^{228}Th data with dissolved ^{224}Ra for all in situ pump data of HLY1502 (panel a) and for the surface water data of PS94 (panel b). The reasonably good agreement supports our use of dissolved ^{224}Ra as analogue for total ^{228}Th.

3 Tracing the sources of ^{228}Ra

In **Figure S3** we use ice back-trajectories to investigate the source regions of the high ^{228}Ra activities observed in the central Arctic. In Figure S3a we present ice back-trajectories for stations sampled in 2007 and 2015. Figure S3b shows that the transit time of the ice from shelf to sampling station was shorter in 2015 than it had been in 2007.

4 Ventilation of intermediate water in the Amundsen Basin

Figure S4 presents the distribution of SF6 on a section from the Barents shelf to the Makarov Basin. High SF6 concentrations and correspondingly low age estimates are indicative of ventilation of the Amundsen Basin (Sta 81) up to about 1500m depth.
Figure S5 gives the effect of variations in the ventilation time on modeled distributions of ^{228}Ra and ^{228}Th.

5 Data

Analytical data of the expeditions in 2011 and 2015 are given as excel files:
Table S1.xlsx: PS94 (2015)- surface values
Table S2.xlsx: PS94 (2015)- depth profiles
Table S3a.xlsx: HLY1502 (2015)- Surface values
Table S3b.xlsx: HLY1502 (2015)- depth profiles
Table S3c.xlsx: HLY1502 (2015)- Quality Flags for Table S3b
Table S4.xlsx: PS78 (2011)- surface values

Model equations

The vertical distribution of ^{228}Ra and ^{228}Th is first described in a one-dimensional model with diffusion, particle settling, and radioactive decay. The equations are:

$$\frac{\partial A_R}{\partial t} = k_z \frac{\partial^2 A_R}{\partial z^2} - \lambda_R A_R \quad (1)$$

$$\frac{\partial A_{T_{diss}}}{\partial t} = k_z \frac{\partial^2 A_{T_{diss}}}{\partial z^2} + \lambda_T A_R - \lambda_T A_{T_{diss}} - k_1 A_{T_{diss}} + k_{-1} A_{T_{part}} \quad (2)$$

$$\frac{\partial A_{T_{part}}}{\partial t} = k_z \frac{\partial^2 A_{T_{part}}}{\partial z^2} - S \frac{\partial A_{T_{part}}}{\partial z} - \lambda_T A_{T_{part}} + k_1 A_{T_{diss}} - k_{-1} A_{T_{part}} \quad (3)$$

Boundary conditions are a bottom source of ^{228}Ra, and a fixed composition (^{228}Ra°, ^{228}Th°) of the surface mixed layer (thickness MLD) thought to be supplied by advection in the TPD.

Then the concept of a one-dimensional model is abandoned when we include two processes of lateral mixing:

1) lateral exchange at all depths with the margin:

$$\frac{\partial A_R}{\partial t} = (A_{R_M} - A_R)/\tau_M \quad (4)$$

and for part and diss Th: $\frac{\partial A_T}{\partial t} = (A_{T_M} - A_T)/\tau_M$ (5)

2) ventilation at 200-1500m depth with shelf water

$$\frac{\partial A_R}{\partial t} = (A_{R_K} - A_R)/\tau_K \quad (6)$$

and for part and diss Th: $\frac{\partial A_T}{\partial t} = (A_{T_K} - A_T)/\tau_K$ (7)

Figures

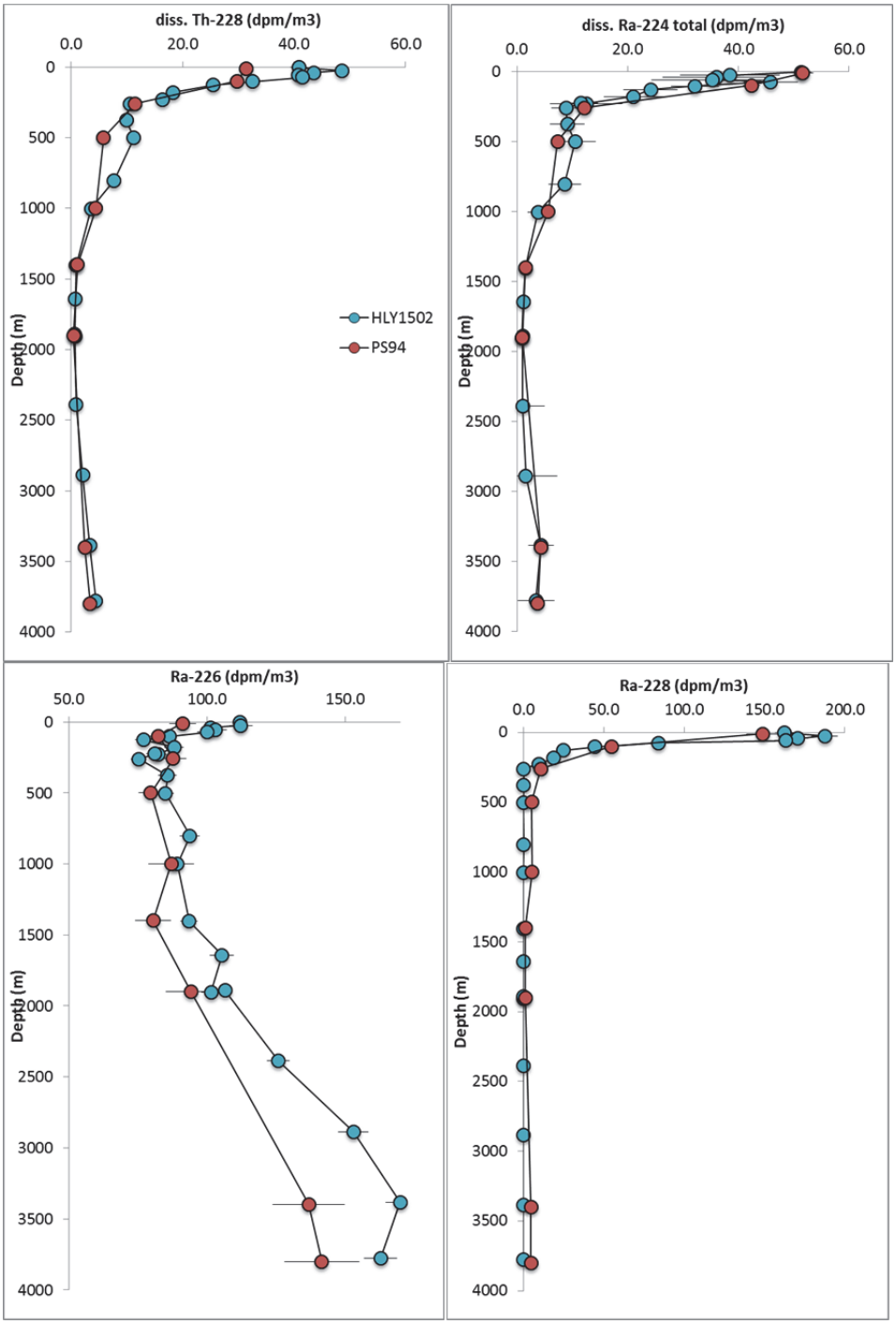

Figure S1. Intercalibration of ^{228}Th, ^{224}Ra, ^{226}Ra and ^{228}Ra for Polarstern PS94 (red) and Healy HLY1502 (blue) at the cross-over station in the Makarov Basin (87.5°N 180°E)

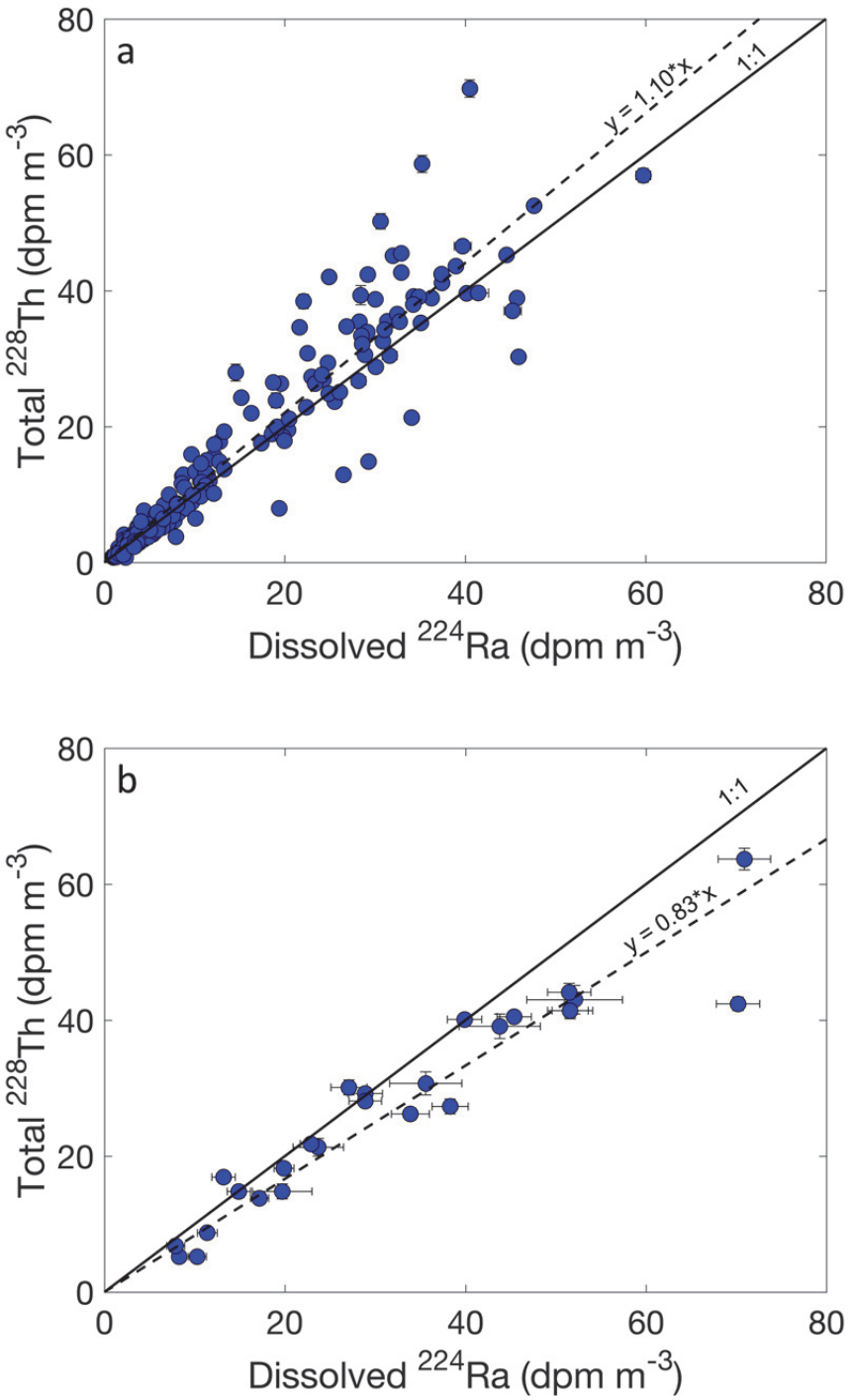

Figure S2 a: Comparison of total (dissolved + small-particulate + large-particulate) ^{228}Th and dissolved ^{224}Ra for all in situ pump data of HLY1502 and b: comparison of total (dissolved + particulate) ^{228}Th and dissolved ^{224}Ra for all surface water data of PS94 with linear regression (dashed) and identity lines (full).

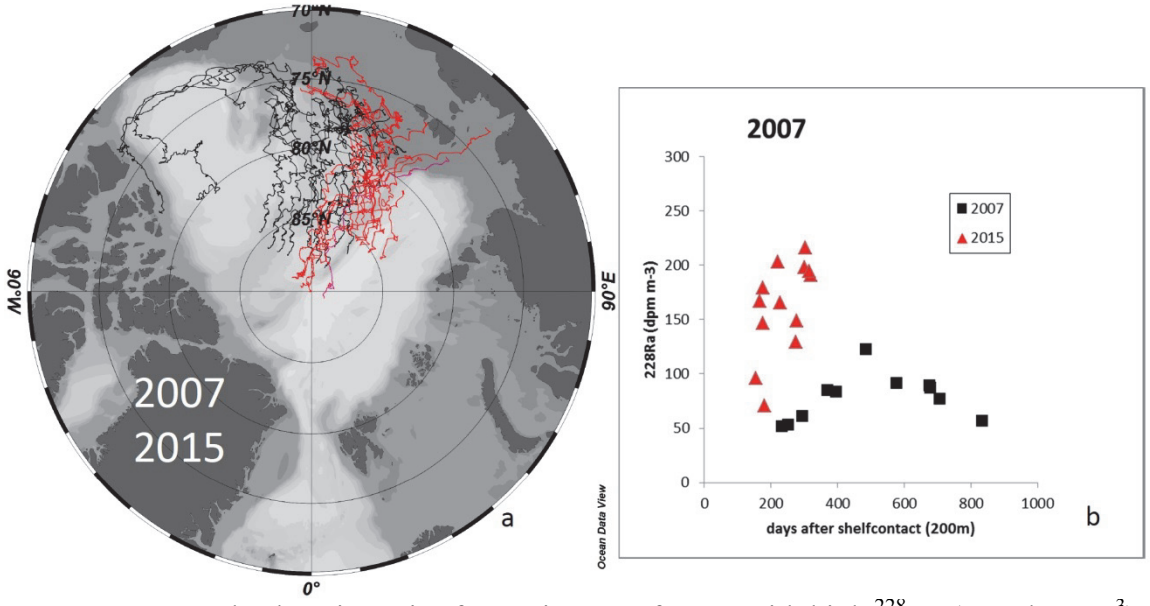

Figure S3 a: Ice back-trajectories for stations N of 84°N with high ^{228}Ra (>50 dpm m^{-3}) in surface water for PS expeditions in 2007 (black) and 2015 (red) showing no apparent difference in shelf of origin; b: ^{228}Ra activity of same stations as function of the age (days) of the ice since surface water left contact with the shelf (200m isobath).

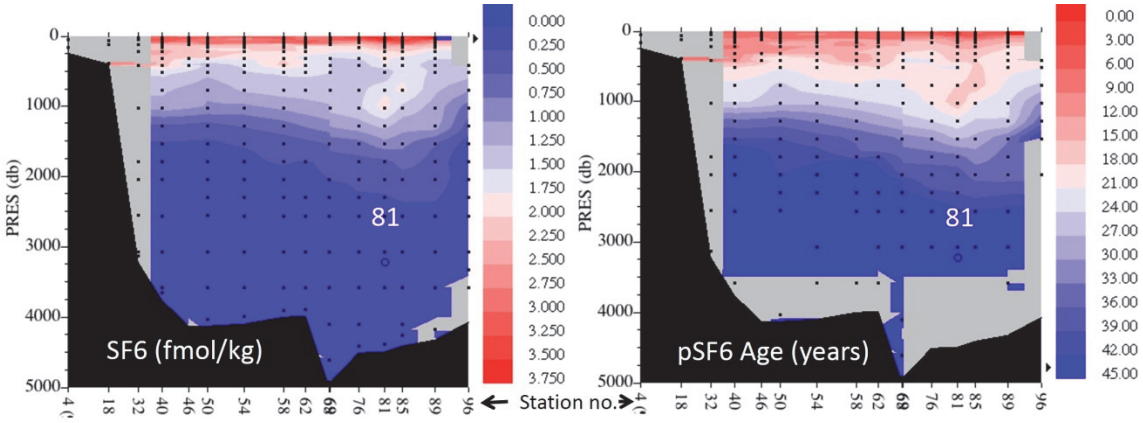

Figure S4. left: SF6 concentrations and right: pSF6 age on the section of PS94 from the Barents shelf to the Makarov Basin. The pSF6 age was calculated assuming that the surface water was in equilibrium with the atmosphere. The pSF6 age of the feature around 1000m at Sta 81 is 15-18 years.

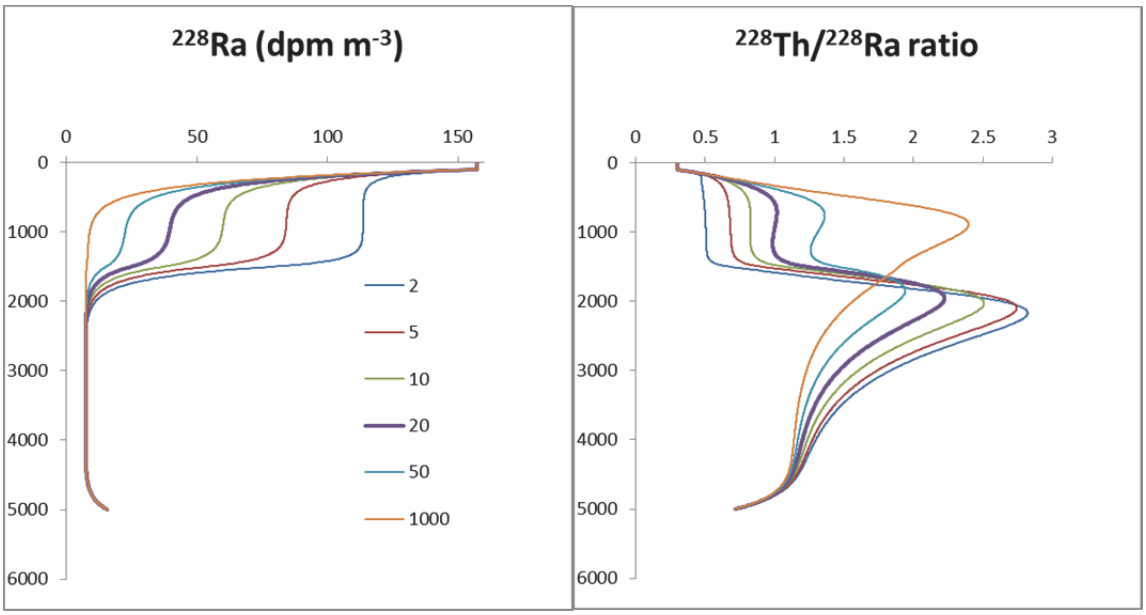

Figure S5. sensitivity of ^{228}Ra and ^{228}Th/^{228}Ra profiles on the choice of the ventilation time τ_K of the intermediate water layer

Table Captions

Table S1. Station number, sampling date, position, bottom depth, sampling depth and concentrations of dissolved ^{228}Th, excess dissolved ^{224}Ra, dissolved ^{224}Ra, ^{226}Ra, ^{228}Ra, and the $^{224}Ra_{diss}/^{228}$Ra activity ratio with 1-sigma error, for surface (10m) and water column (In Situ Pump) samples of Polarstern expedition PS94, with nearest depth of Rosette water sample with salinity, potential temperature, δ^{18}O and calculated fraction of meteoric water.

Table S2. Station number, sampling date, position, and concentrations of dissolved ^{228}Th, excess dissolved ^{224}Ra, total dissolved ^{224}Ra, particulate ^{228}Th, particulate ^{224}Ra, total ^{228}Th, ^{226}Ra, ^{228}Ra, with 1-sigma error, adsorber type, and excess total ^{224}Ra, and ocean area for surface samples of Polarstern expedition PS94.

Table S3a. Station number, position, ^{226}Ra, ^{228}Ra, ^{224}Ra, and ^{224}Ra/^{228}Ra ratio with 1-sigma errors for surface samples (2m depth) of Healy expedition HLY1502.

Table S3b. Station number, position, ^{226}Ra, ^{228}Ra, ^{224}Ra, dissolved ^{228}Th, particulate ^{228}Th, total ^{228}Th, ^{224}Ra-^{228}Ra, and ^{224}Ra/^{228}Ra ratio with 1-sigma errors and quality flags for ISP samples of Healy expedition HLY1502.

Table S3c. explanation of flags used in Table S3b

Table S4. Station number, sampling date, position, bottom depth, and concentrations of dissolved ^{224}Ra, ^{226}Ra, and ^{228}Ra, with 1-sigma error, salinity, potential temperature, δ^{18}O and calculated fraction of meteoric water for surface samples of Polarstern expedition PS78.