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## **231Pa and 230Th in the Arctic Ocean: Implications for boundary scavenging and 231Pa/230Th fractionation in the Eurasian Basin**

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### ► To cite this version:

Sandra Gdaniec, Matthieu Roy-Barman, Martin Levier, Ole Valk, Michiel Rutgers van Der Loeff, et al.. 231Pa and 230Th in the Arctic Ocean: Implications for boundary scavenging and 231Pa/230Th fractionation in the Eurasian Basin. *Chemical Geology*, 2020, 532, pp.119380. 10.1016/j.chemgeo.2019.119380 . hal-02467573

**HAL Id: hal-02467573**

**<https://hal.science/hal-02467573>**

Submitted on 21 Dec 2021

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1  **$^{231}\text{Pa}$  and  $^{230}\text{Th}$  in the Arctic Ocean: implications for Boundary**  
2 **Scavenging and  $^{231}\text{Pa}$ - $^{230}\text{Th}$  fractionation in the Eurasian Basin**

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34

35 **Abstract**

36  $^{231}\text{Pa}$ ,  $^{230}\text{Th}$  and  $^{232}\text{Th}$  were analyzed in filtered seawater ( $n = 70$ ) and suspended particles ( $n$   
37  $= 39$ ) collected along a shelf-basin transect from the Barents shelf to the Makarov Basin in the  
38 Arctic Ocean during GEOTRACES section GN04 in 2015. The distribution of dissolved  $^{231}\text{Pa}$   
39 and  $^{230}\text{Th}$  in the Arctic Ocean deviates from the linear increase expected from reversible  
40 scavenging. Higher  $^{232}\text{Th}$  concentrations were observed at the shelf, slope and in surface  
41 waters in the deep basin, pointing at lithogenic sources. Fractionation factors ( $F_{\text{Th/Pa}}$ ) observed  
42 at the Nansen margin were higher compared to  $F_{\text{Th/Pa}}$  in the central Nansen Basin, possibly  
43 due to the residual occurrence of hydrothermal particles in the deep central Nansen Basin.  
44 Application of a boundary scavenging model quantitatively accounts for the dissolved and  
45 particulate  $^{230}\text{Th}$  distributions in the Nansen Basin. Modeled dissolved  $^{231}\text{Pa}$  distributions were  
46 largely overestimated, which was attributed to the absence of incorporation of water exchange  
47 with the Atlantic Ocean in the model.  $^{231}\text{Pa}/^{230}\text{Th}$  ratios of the suspended particles of the  
48 Nansen Basin were below the  $^{231}\text{Pa}/^{230}\text{Th}$  production ratio, but top-core sediments of the  
49 Nansen margin and slope have high  $^{231}\text{Pa}/^{230}\text{Th}$ -ratios, suggesting that scavenging along the  
50 Nansen margin partly acts as a sink for the missing Arctic  $^{231}\text{Pa}$ .

51

52 **Keywords:** GEOTRACES, Protactinium, Thorium, Arctic Ocean, Boundary scavenging,

53

## 54 **1 Introduction**

55 Particle reactive isotopes in the natural uranium and thorium decay series are useful tracers  
56 of particle flux and scavenging in the oceans (e.g. Edmonds et al., 2004; Roy-Barman, 2009;  
57 Rutgers van der Loeff and Berger, 1993). In seawater,  $^{231}\text{Pa}$  and  $^{230}\text{Th}$  are produced at a  
58 constant rate by the decay of homogeneously distributed  $^{235}\text{U}$  and  $^{234}\text{U}$ , respectively. Both  
59  $^{231}\text{Pa}$  and  $^{230}\text{Th}$  are particle reactive elements, which means that they get scavenged onto  
60 settling particles and removed from the water column to the sediments. However, due to  
61 differences in their particle reactivity, the  $^{231}\text{Pa}/^{230}\text{Th}$  ratio of seawater, marine particles and  
62 sediments often differs from that of the production ratio (Rutgers van der Loeff and Berger,  
63 1993). The distribution and dispersion of  $^{231}\text{Pa}$  and  $^{230}\text{Th}$  in the water column and their ratio in  
64 sediments are of interest for processes controlling spatial and temporal variations in ocean  
65 biogeochemistry. The difference in the chemical behavior of the two tracers can result in large  
66 scale deviation of the  $^{231}\text{Pa}/^{230}\text{Th}$  ratio of marine sediments compared to the production ratio  
67 of these nuclides in the overlying water column, with higher sedimentary  $^{231}\text{Pa}/^{230}\text{Th}$  ratios at  
68 ocean margin and lower  $^{231}\text{Pa}/^{230}\text{Th}$  to the inner ocean compared to the production ratio.  
69 Differences in particle concentration, composition and flux influence scavenging rates of  $^{231}\text{Pa}$   
70 and  $^{230}\text{Th}$  in the water column (Chase et al., 2002). Environments governed by high particle  
71 flux, such as ocean margins are very effective sinks for  $^{231}\text{Pa}$  and  $^{230}\text{Th}$  (e.g. Scholten et al.,  
72 1995; Gdaniec et al., 2017). This process, usually referred to as boundary scavenging has  
73 previously been thought to be pronounced in the Arctic Ocean (Bacon et al., 1989; Cochran et  
74 al., 1995; Scholten et al., 1995). However, despite the contrasted particle fluxes over large  
75 shelf areas (receiving high river inputs) and the inner Arctic with its perennial sea ice cover,  
76 the  $^{231}\text{Pa}/^{230}\text{Th}$  ratio of arctic sediment does not vary much and is on average lower than the  
77 production ratio (e.g. Edmonds et al., 2004; Moran et al., 2005b). Not many studies have  
78 reported  $^{231}\text{Pa}/^{230}\text{Th}$  ratios slightly exceeding the production ratio (e.g. Luo and Lippold, 2015).  
79 The overall low  $^{231}\text{Pa}/^{230}\text{Th}$  ratios casted some doubts on mechanisms driving the boundary  
80 scavenging in the Arctic Ocean. While boundary scavenging does occur in the Arctic Ocean  
81 (Roy-Barman, 2009), the net export of  $^{231}\text{Pa}$  to the Atlantic Ocean through the Fram strait plays  
82 a key role in the Pa and Th budget of the Arctic Ocean (Edmonds et al., 2004; Hoffmann et al.,  
83 2013; Moran et al., 2005). However, the low vertical resolution, the relatively large analytical  
84 uncertainties and sometimes lack of particulate data limits the water column constraints on  
85 boundary scavenging in Arctic Ocean (Bacon et al., 1989; Cochran et al., 1995; Edmonds et  
86 al., 2004; Scholten et al., 1995) and more data are required for a proper modeling of boundary  
87 scavenging (Luo and Lippold, 2015).

88 In this study, dissolved and particulate  $^{231}\text{Pa}$ ,  $^{230}\text{Th}$ , and  $^{232}\text{Th}$  concentrations  
89 measured along GEOTRACES section GN04 in the Barents Sea, Nansen Basin, Amundsen



90 Basin and Makarov Basin are presented. The objective was to explore the influence of  
91 boundary scavenging and shelf-basin interactions on the observed distribution of  $^{231}\text{Pa}$  and  
92  $^{230}\text{Th}$  in the Arctic Ocean. We revisit boundary scavenging modelling using a model adapted  
93 from Roy-Barman (2009) that we compared to the current dataset and used to constrain the  
94 scavenging behavior of Pa and Th between the Arctic margin and the inner ocean.  
95

## 96 **2. Methods**

### 97 **2.1 Sampling**

98 Samples were collected on R/V Polarstern during expedition PS94 in 2015, at seven stations  
99 located in the Barents Sea, Nansen, Amundsen and Makarov Basins along the GEOTRACES  
100 GA04 section in the Arctic Ocean (Fig. 1). The samples were collected along a shelf-basin  
101 transect from the Barents shelf to the Makarov Basin to study the exchange between the  
102 margin and interior ocean. Stations 4, 161, 153 and 18 represent the shelf, station 32  
103 represents the margin, while station 40, 50, 125 (Amundsen basin) and 101 (Makarov basin)  
104 represent the interior ocean.

105 Water samples were collected in 24 L Niskin<sup>®</sup> bottles mounted on a General Oceanic<sup>®</sup>  
106 rosette equipped with a Sea-Bird Electronics CTD system (SBE911plus). The CTD-system  
107 was equipped with sensors allowing measurements of salinity, temperature and transmission  
108 (Rabe et al., 2016; van Ooijen et al., 2016). For the analysis of dissolved  $^{231}\text{Pa}$ ,  $^{230}\text{Th}$  and  $^{232}\text{Th}$ ,  
109 5 L of water were filtered directly from the Niskin bottles into sampling containers using  
110 Acropak500<sup>™</sup> cartridges (0.45  $\mu\text{m}$  pore size), which were cleaned in between stations. After  
111 filtration, water samples were acidified using concentrated ultra-pure HCl (~1 ml of acid per 1  
112 L of seawater). Samples were stored in double plastic bags until analysis.

113 Particulate samples were collected using *in-situ* pumps (McLane and Challenger  
114 Oceanic) at six stations along the GA04 section (Fig. 1). Particles (0.8  $\mu\text{m}$  pore size) were  
115 collected on Supor<sup>®</sup> polyethersulfone filters with a diameter of 142 mm.

116 At two locations (N 84° 6' 51.64", E 12° 4' 11.69" and N 84° 31.40", E 11° 6' 11.43"),  
117 "dirty ice" (ice rafted sediments incorporated into sea ice when it forms on the Arctic shelf) was  
118 collected from deck using a plastic spade and container.

119 At station 161, 32 and 101, surface sediment cores were collected using a Multi-corer  
120 (Fig. 1). The top (0-1 cm interval) of each core was analyzed for  $^{238}\text{U}$ ,  $^{234}\text{U}$ ,  $^{231}\text{Pa}$ ,  $^{232}\text{Th}$  and  
121  $^{230}\text{Th}$  at LSCE.  
122

123 2.2 Chemical preparation of Pa and Th in seawater, suspended particles, surface  
124 sediments and dirty ice

125  $^{231}\text{Pa}$ ,  $^{230}\text{Th}$  and  $^{232}\text{Th}$  were determined by isotope dilution and mass spectrometry. Seawater  
126 samples were processed as described in (Gdaniec et al., 2017). For filtered particles,  
127 approximately 1/5 (corresponding to ~25 - 200 L seawater) of the total filter material was cut  
128 onboard in a laminar flow bench and used to determine the concentrations of  $^{231}\text{Pa}$  and Th  
129 isotopes in suspended particles. The leaching of filter samples was performed at the Swedish  
130 Museum of Natural history followed by spiking and analysis at LSCE. The filters were  
131 submerged in 3N HCl and heated to  $>50^\circ\text{C}$  to wash the particles off the filters without dissolving  
132 the filters. The leachate, containing the suspended particles, was then dissolved in a mixture  
133 of concentrated  $\text{HNO}_3$  and HF following the method described in Gdaniec et al. (2017).

134 Surface sediments were dried and an aliquot (0.2 g) of crushed bulk sediment was  
135 spiked with  $^{233}\text{Pa}$ ,  $^{229}\text{Th}$  and  $^{236}\text{U}$ , followed by total dissolution in a  $\text{HNO}_3$  and HF mixture. Pa,  
136 Th and U were separated by anion exchange chromatography (Guihou et al., 2010).

137 The dirty ice samples were melted and particles were separated by centrifugation at  
138 the clean laboratory of Laboratoire des Sciences de l'Environnement Marin (LEMAR). Total  
139 dissolution of the particles (~0.2 g) was carried out at the Swedish Museum of Natural History  
140 using microwave oven digestion using HF,  $\text{HNO}_3$  and HCl followed by spiking, chemical  
141 separation and analysis of  $^{231}\text{Pa}$ ,  $^{230}\text{Th}$  and  $^{232}\text{Th}$  at Laboratoire des Sciences du Climat et de  
142 l'Environnement (LSCE).

143  $^{231}\text{Pa}$ ,  $^{230}\text{Th}$  and  $^{232}\text{Th}$  concentrations in seawater, particles and sediment samples  
144 were analyzed at LSCE by Multi Collector Inductively Coupled Plasma Mass Spectrometry  
145 (MC-ICP-MS) on a Thermo Scientific™ Neptune Plus™ instrument equipped with an Aridus  
146 II™ desolvating nebulizer and a Jet interface (Burckel et al., 2015, Gdaniec et al., 2017).

147 Uranium concentrations in seawater were estimated using the bottle salinity  
148 measured from the CTD and the U-Salinity relationship in seawater ( $U = (0.100 * S - 0.326)$ ,  
149 Owens et al., 2011). The conservative behavior of uranium in the Arctic Ocean can be  
150 questioned due to the surface and mixed layer of the Arctic contain a significant portion of a  
151 mixture of river and ice melt water. However, in the study of Not et al. (2012) the investigators  
152 apply the U vs. salinity relationship over a salinity range of ~0 – 135, and showed that the U-  
153 salinity relationship exhibits the conservative behavior over the entire range investigated.

154 Procedural blanks for seawater samples were determined by performing a complete  
155 chemical procedure on 3-11 bottles of 250 ml of Milli-Q® water with each batch of samples.  
156 Total procedural blanks for seawater samples ranged between 7.3 pg and 29.9 pg for  $^{232}\text{Th}$   
157 (average =  $15 \pm 6$  pg), 0.07 and 8.42 fg for  $^{230}\text{Th}$  (average =  $1.02 \pm 1.80$  fg) and 0.05 and 0.26

158 fg for  $^{231}\text{Pa}$  (average =  $0.17 \pm 0.18$  fg). These blanks were equivalent to 0.9 – 14 % of the  
159 measured  $^{232}\text{Th}$ , 0.2 – 21 % of the measured  $^{230}\text{Th}$  and 0.1 – 42 % of the measured  $^{231}\text{Pa}$ .

160 For particles, procedural blanks were prepared in the same way as for samples by  
161 using acid cleaned filters (0.5N  $\text{HNO}_3$ ) mounted onto the pump and deployed but not  
162 connected to the pumping system. Total procedural blanks ranged between 3.3 pg and 29.1  
163 pg for  $^{232}\text{Th}$  (average =  $4.3 \pm 1.2$  pg), 0.02 and 0.36 fg for  $^{230}\text{Th}$  (average =  $0.15 \pm 0.03$  fg) and  
164 0.004 and 0.7 fg for  $^{231}\text{Pa}$  (average =  $0.27 \pm 0.34$  fg). These blanks were equivalent to 0.1 –  
165 16 % of the measured  $^{232}\text{Th}$ , 0.1 – 19 % of the measured  $^{230}\text{Th}$  and 0.1 – 89 % of the measured  
166  $^{231}\text{Pa}$

167 All measured  $^{231}\text{Pa}$  and  $^{230}\text{Th}$  concentrations were corrected for the in-growth of  $^{231}\text{Pa}$   
168 and  $^{230}\text{Th}$  by uranium decay during the time period between sample collection and the U-Th/Pa  
169 separation. All uncertainties are expressed as 2 standard errors on the mean ( $2 \sigma_n$ ) including  
170 the propagated contribution from sample weighting, spike impurities, spike contributions, blank  
171 corrections and mass spectrometric measurements.

172 Replicates for dissolved  $^{231}\text{Pa}$ ,  $^{230}\text{Th}$  and  $^{232}\text{Th}$  concentrations were measured (Tab S1). It was  
173 initiated because the first analysis of  $^{230}\text{Th}$  at 1000 m depth for station 32 seemed obviously  
174 overestimated. An additional aliquot of the 1000 m sample was analyzed and 2 additional  
175 samples (500 m depth at St. 32 and 40) were replicated to check the method reliability. They  
176 confirmed that the first analysis of  $^{230}\text{Th}$  at 1000 m depth had suffered from contamination, but  
177 showed a good agreement (within or close to analytical uncertainties) for the other  $^{231}\text{Pa}$  and  
178  $^{230}\text{Th}$  measurements. The  $^{232}\text{Th}$  replicates can vary by as much as 30 % indicating small  $^{232}\text{Th}$   
179 contaminations, but it will have no significant impact on the discussion.

180 An inter-comparison of dissolved and particulate Pa-Th measurements between  
181 LSCE and AWI at stations 101 and 125 is given in Fig. ES1. Seawater samples at station 101  
182 (GEOTRACES crossover station of the USCGC Healy HLY1502 (GN01) and R/V Polarstern  
183 PS94 (GN04) cruises) and station 125 were duplicated and analyzed for dissolved  $^{231}\text{Pa}$ ,  $^{230}\text{Th}$   
184 and  $^{232}\text{Th}$  in order to provide an intercomparison between AWI and LSCE. These samples  
185 were obtained from the same casts, but not always the same bottle, so some differences in  
186 concentrations were expected (Fig. ES1). Concentrations of dissolved  $^{231}\text{Pa}$ ,  $^{230}\text{Th}$  and  $^{232}\text{Th}$   
187 at station 50 (Fig. 1) have been published in Valk et al., 2018 and will be used here to  
188 investigate the relationship between the Nansen margin and interior. Moreover, an  
189 intercomparison of dissolved and particulate Pa-Th measurements with AWI (Alfred Wegener  
190 Institute), UMN (Minnesota University) and LDEO (Lamont Doherty Earth Observatory) at  
191 station 101 is in progress.

192

193 **3 Results**

### 194 3.1 General circulation and Hydrography

195 The Arctic Mediterranean, like the Mediterranean Sea, transforms and exports Atlantic water  
196 of lower density entering from the adjacent ocean into high density intermediate and deep  
197 waters. The Arctic Ocean comprises two major basins, the Amerasian Basin and the Eurasian  
198 Basin, separated by the Lomonosov Ridge (Fig. 1). The Eurasian Basin is divided into the  
199 Nansen and Amundsen Basin by the Gakkel Ridge, while the Amerasian Basin is separated  
200 by the Alpha-Mendeleyev Ridge into the Makarov and the Canada Basins (Fig. 1).

201 The inflow of Atlantic Water (AW) over the Barents Sea was recognized at stations  
202 161 and 153, where high salinity ( $>35.1$ ) and high temperatures ( $\sim 2 - 7.6^\circ\text{C}$ ) were observed  
203 (Rabe et al., 2016) (Fig. ES2). Close to the Barents Sea shelf break (St. 04 and 18), the polar  
204 waters (Polar Mixed Layer and Halocline) were significantly colder ( $\leq 0.8^\circ\text{C}$ ) and fresher ( $\leq$   
205  $34.8$ ) compared to the inflow of AW (Fig. ES2). AW enters the central Arctic Ocean mainly by  
206 two branches; the Fram Strait Branch Water (FSBW) and the Barents Sea Branch Water  
207 (BSBW) (Rudels, 1994). The FSBW enters the Nansen Basin through the Fram Strait and then  
208 forms the Boundary current that flows around the Nansen Basin along the Gakkel Ridge (e.g.  
209 Rudels, 2009) (Fig. ES3). As AW enters the Arctic Basin, it encounters sea ice and the upper  
210 water masses cool ( $-1.8^\circ\text{C}$ ) and become less saline ( $\sim 34.9 - 35.0$ ) compared to underlying  
211 waters (Fig. ES2).

212 The BSBW flows over the Barents Sea shelf and enters the Nansen Basin through  
213 St. Anna Trough ( $\sim 1000$  m depth), where limited exchange with FSBW occurs (e.g. Rudels,  
214 2009) (Fig. ES3). At the eastern Siberian margin, the BSBW is divided into 3 branches: one  
215 branch flows into the Amundsen Basin while the second branch of the BSBW continues to flow  
216 towards the Fram Strait, as well as the ventilated intermediate water masses (Tanhua et al.,  
217 2009) (Fig. ES3). The third part of the BSBW prolongs into the Makarov Basin as part of the  
218 Arctic Ocean Boundary Current that travels anti-cyclonically around the Arctic Ocean (Rudels,  
219 1994; Rudels et al., 2012).

220 In the Nansen Basin (St. 32 and 40), the warm FSBW was observed between  $\sim 100$   
221 and  $\sim 1000$  m (Rabe et al., 2016) (Fig. ES2). In these waters, the Atlantic layer typically has  
222 maximum temperature of  $\sim 2.5^\circ\text{C}$  and the salinity ranges between  $34.9$  and  $35.05$  (Fig. ES2).  
223 Station 50, in the central Nansen Basin is influenced by the return flow of colder ( $-2^\circ\text{C}$ ) and  
224 less saline FSBW along the Gakkel Ridge (Rabe et al., 2016) (Fig. ES2).

225 The Eurasian Basin communicates with the Canada Basin through the boundary  
226 current that enters the Canada Basin north of Siberia and through the intra-Basin located at  
227 the central part of the Lomonosov Ridge, with a sill depth of  $\sim 1850$  m (Björk et al., 2007) (Fig.  
228 ES3). A reverse flow from the Makarov Basin to the Amundsen basin has also been identified  
229 (Björk et al., 2010, 2007)

230 Above 1700 m, stations 125 and 101 show similar water mass characteristics that fit  
 231 to the BSBW (Rabe et al., 2016) (Fig. ES2). Below this depth range, the deep waters of the  
 232 Amerasian Basin are warmer and saltier compared to the Eurasian Basin Deep Water (EBDW)  
 233 (Fig. ES2) (e.g. Aagaard et al., 1981). The deep waters from the Eurasian Basin exit the Arctic  
 234 through the Fram Strait and contribute to the deeper layers in the Nordic Seas (e.g. Rudels,  
 235 2015).

### 236 3.2 Dissolved and particulate $^{232}\text{Th}$ concentrations

237 Concentrations of dissolved  $^{232}\text{Th}$  in the Arctic Ocean ranged between 11 and 205 pg/kg and  
 238 were generally decreasing with depth (Fig. 2). Elevated concentrations of  $^{232}\text{Th}$  were observed  
 239 close to the Nansen shelf break at station 04 and 18 (72 – 205 pg/kg) and in surface waters of  
 240 station 101 (139 – 194 fg/kg). Below ~500m depth, concentrations of dissolved  $^{232}\text{Th}$  were  
 241 higher closer to the Nansen margin (24 – 80 pg/kg) compared to the Nansen interior (11 – 60  
 242 pg/kg) (Fig. 2).

243 Concentrations of particulate  $^{232}\text{Th}$  in the Barents Sea display elevated concentrations  
 244 at depth, where concentrations up to ~2500 pg/kg were observed at station 04 (Fig. 3). At the  
 245 deep stations, particulate  $^{232}\text{Th}$  ranged between 10 pg/kg and ~80 pg/kg, where lower  
 246 concentrations were observed at the ocean interior (St. 101 and 50) compared to the margin  
 247 (St. 32) (Fig. 3).

### 249 3.3 Dissolved $^{231}\text{Pa}_{xs}$ and $^{230}\text{Th}_{xs}$ concentrations

250 Measured concentrations of  $^{231}\text{Pa}$  and  $^{230}\text{Th}$  must be corrected for the presence of detrital  
 251 components due to the presence of U in lithogenic particles. The concentration of lithogenic U  
 252 is estimated from the measured concentrations of  $^{232}\text{Th}$ , which is entirely of lithogenic origin  
 253 (Brewer et al., 1980). As U is assumed to be at secular equilibrium in lithogenic phases, the  
 254 unsupported  $^{231}\text{Pa}$  and  $^{230}\text{Th}$  ( $^{231}\text{Pa}_{xs}$  and  $^{230}\text{Th}_{xs}$ ) produced solely by radioactive decay of  
 255 dissolved U-isotopes can be calculated:

$$257 \quad ^{230}\text{Th}_{xs} = ^{230}\text{Th}_m - ^{232}\text{Th}_m \times \left( \frac{^{230}\text{Th}}{^{232}\text{Th}} \right)_{Litho} \times \frac{M_{230}}{M_{232}} \quad (1)$$

$$259 \quad ^{231}\text{Pa}_{xs} = ^{231}\text{Pa}_m - ^{232}\text{Th}_m \times \left( \frac{^{230}\text{Th}}{^{232}\text{Th}} \right)_{Litho} \times \left( \frac{^{235}\text{U}}{^{238}\text{U}} \right)_{Nat} \times \frac{\lambda_{230}}{\lambda_{238}} \times \frac{\lambda_{235}}{\lambda_{231}} \times \frac{M_{231}}{M_{232}} \quad (2)$$

261 where the subscript *m* refers to measured concentrations of  $^{231}\text{Pa}$ ,  $^{230}\text{Th}$  (fg/kg) and  
 262  $^{232}\text{Th}$  (pg/kg) in seawater and particles. The  $(^{235}\text{U}/^{238}\text{U})_{Nat}$  is the natural  $^{235}\text{U}/^{238}\text{U}$  ratio of

263 1/137.88 (atom/atom) (Condon et al., 2010) and the  $(^{230}\text{Th}/^{232}\text{Th})_{\text{Litho}}$  was estimated from the  
 264 average activity ratio  $[^{238}\text{U}/^{232}\text{Th}] = 0.6 \pm 0.3$ . This range is relatively large compared to values  
 265 used previously for the Arctic Ocean ( $[^{238}\text{U}/^{232}\text{Th}] = 0.6 \pm 0.1$ , Moran et al., 2005b). It allows  
 266 covering the high value ( $[^{238}\text{U}/^{232}\text{Th}] \approx 0.9$ ) deduced from the  $^{230}\text{Th}/^{232}\text{Th}$  ( $= 4.75 \times 10^{-6}$  mol/mol)  
 267 measured in the dirty ice, as well as the low ratio ( $[^{238}\text{U}/^{232}\text{Th}] = 0.3$ ) inferred from the filtered  
 268 particles at station 04 to avoid “negative  $^{231}\text{Pa}_{\text{xs}}$ -values” in (see section 3.4). The secular  
 269 equilibrium in the lithogenic fraction of marine sediment is at has been questioned due to the  
 270 possible loss of  $^{234}\text{U}$  (through  $^{234}\text{Th}$ , its parent isotope),  $^{230}\text{Th}$  and  $^{231}\text{Pa}$  (through  $^{231}\text{Th}$ , its  
 271 parent isotope) by  $\alpha$ -recoil (Bourne et al., 2012). At present, it is unclear if this process is  
 272 sufficient to explain the discrepancy between the 2 estimates of the  $[^{238}\text{U}/^{232}\text{Th}]$  activity ratio  
 273 presented here. Therefore, we conservatively consider the range  $[^{238}\text{U}/^{232}\text{Th}] = 0.6 \pm 0.3$  as  
 274 possible and to propagate the resulting uncertainties. The same lithogenic corrections have  
 275 been made for the dissolved concentrations of  $^{231}\text{Pa}$  and  $^{230}\text{Th}$  at stations 50 and 125  
 276 measured by Valk et al., (2018).

277  $\lambda_{230}$ ,  $\lambda_{231}$ ,  $\lambda_{235}$  and  $\lambda_{238}$  are the decay constants for  $^{230}\text{Th}$  ( $\lambda_{230} = 9.16 \times 10^{-6} \text{ y}^{-1}$ ),  $^{231}\text{Pa}$   
 278 ( $\lambda_{231} = 2.11 \times 10^{-5} \text{ y}^{-1}$ ),  $^{235}\text{U}$  ( $\lambda_{235} = 9.85 \times 10^{-10} \text{ y}^{-1}$ ) and  $^{238}\text{U}$  ( $\lambda_{238} = 1.55 \times 10^{-10} \text{ y}^{-1}$ ), respectively  
 279 (Cheng et al., 1998; Condon et al., 2010).  $M_{230}$ ,  $M_{231}$  and  $M_{232}$  are the atomic masses of  
 280  $^{230}\text{Th}$  (230.033 g/mol)  $^{231}\text{Pa}$  (231.036 g/mol) and  $^{232}\text{Th}$  (232.038 g/mol). The concentrations of  
 281  $^{231}\text{Pa}$  and  $^{230}\text{Th}$  can be converted to radioactivity units ( $\mu\text{Bq/kg}$ ) by using the conversion factors  
 282 0.5724 and 1.3110, respectively.  $^{232}\text{Th}$  concentrations can be converted to pmol/kg using the  
 283 atomic mass of  $^{232}\text{Th}$  (232.038).

284 Higher lithogenic content was observed in particles and seawater collected at the  
 285 shelf, margin and in bottom waters of the deep basins. On average, the lithogenic contribution  
 286 ranged between 0.04 and 33 % for dissolved  $^{231}\text{Pa}$ , between 2.3 and 72 % for particulate  $^{231}\text{Pa}$ ,  
 287 between 0.3 and 34 % for dissolved  $^{230}\text{Th}$  and between 1.0 and 58 % for particulate  $^{230}\text{Th}$ .

288 In the deep Arctic Basin, the observed dissolved concentrations of  $^{231}\text{Pa}_{\text{xs}}$  and  $^{230}\text{Th}_{\text{xs}}$   
 289 are increasing with depth until ~2000 m, followed by decreasing or invariant concentrations  
 290 approaching the seafloor. This depletion at depth was more pronounced at the Nansen Basin  
 291 stations (32, 40, 50) compared to the Makarov Basin station (101). The depletion of  $^{230}\text{Th}_{\text{xs}}$  in  
 292 the Nansen Basin was greater compared to the decrease of dissolved  $^{231}\text{Pa}_{\text{xs}}$  in waters below  
 293 2000 m depth (Fig. 2).

294 In the Makarov Basin, dissolved  $^{231}\text{Pa}_{\text{xs}}$  ranged between 0.02 and 4.0 fg/kg and  
 295 concentrations of dissolved  $^{230}\text{Th}_{\text{xs}}$  ranged between 0.9 and 27.9 fg/kg. In contrast, lower  
 296 concentrations of dissolved  $^{231}\text{Pa}_{\text{xs}}$  (0.19 – 1.9 fg/kg) and  $^{230}\text{Th}_{\text{xs}}$  (2.1 – 8.9 fg/kg) were  
 297 observed at the deep stations of the Eurasian Basin (Fig. 2).

298 In the Barents Sea, concentrations of dissolved  $^{231}\text{Pa}_{\text{xs}}$  were lower in the polar waters  
 299 of stations 04 and 18 (0.1 – 0.2 fg/kg) compared to the inflowing Atlantic Water stations 153

300 and 161 (0.23 – 0.6 fg/kg), while the dissolved  $^{231}\text{Pa}_{\text{xs}}$  in shallow waters (<500 m) over the  
301 slope (St 32) and in the Nansen Basin (stations 40 and 50) ( $^{231}\text{Pa}_{\text{xs}} = 0.20 - 0.4 \text{ fg/kg}$ ) are  
302 intermediate between the Atlantic inflowing water (St. 153, 161) and close to Svalbard (St. 4).

303 The distribution of dissolved  $^{230}\text{Th}_{\text{xs}}$  was similar for all four shelf stations (0.5 – 1.99  
304 fg/kg) (Fig. 2) and generally lower compared to water at corresponding depth of the interior  
305 basin (2.3 – 3.8 fg/kg) (Fig. 2).

306 At station 125, located in the Amundsen Basin, the water column distribution of  
307 dissolved  $^{231}\text{Pa}$  was similar compared to the stations located in the Nansen Basin, while  
308 concentrations of dissolved  $^{230}\text{Th}$  in the Nansen Basin (St. 32, 40, 50) were similar or higher  
309 compared to the distribution of dissolved  $^{230}\text{Th}$  from 500 m to 2000 m depth at station 125 (Fig.  
310 2).

311

### 312 3.4 Particulate $^{231}\text{Pa}_{\text{xs}}$ and $^{230}\text{Th}_{\text{xs}}$ concentrations

313 The filtered particles from station 04 (Tab. ES2) and the dirty ice (Tab. ES3) were used to  
314 determine the range of [ $^{238}\text{U}/^{232}\text{Th}$ ] activity ratios of the lithogenic material. There was a high  
315 abundance of lithogenic particles at station 04 ( $^{232}\text{Th} = 46 - 2400 \text{ pg/kg}$ ) and these particles  
316 have low  $^{231}\text{Pa}/^{232}\text{Th}$  ratios that must be supported by a [ $^{238}\text{U}/^{232}\text{Th}$ ] activity ratio of  $\sim 0.3$  (to  
317 avoid negative values of  $^{231}\text{Pa}_{\text{xs}}$ ), a value that we take as the lower limit of the [ $^{238}\text{U}/^{232}\text{Th}$ ]  
318 activity ratio of the lithogenic material. Ice rafted sediments embedded in sea ice (called “dirty  
319 ice”) are an important source of lithogenic particles in the Arctic Ocean (Pfirman and Thiede,  
320 1987). As dirty ice is formed on the shelf, it may contain shelf sediments that have been  
321 incorporated into the ice as anchor ice or by sediment resuspension. This means that the dirty  
322 ice could contain some  $^{230}\text{Th}_{\text{xs}}$  that has been scavenged over the shelf. Here, the  $^{230}\text{Th}/^{232}\text{Th}$   
323 ratio ( $= 4.75 \times 10^{-6} \text{ mol/mol}$ ) of dirty ice is supported by a [ $^{238}\text{U}/^{232}\text{Th}$ ] activity ratio of  $\sim 0.9$ , a  
324 value in good agreement with the estimate of the average continental crust suggesting that  
325 dirty ice is free of  $^{230}\text{Th}_{\text{xs}}$ . Therefore, we take 0.9 as the upper limit of the [ $^{238}\text{U}/^{232}\text{Th}$ ] activity  
326 ratio of the lithogenic material.

327 Concentrations of particulate  $^{231}\text{Pa}_{\text{xs}}$  and  $^{230}\text{Th}_{\text{xs}}$  in the Arctic Ocean generally display  
328 an increasing trend with depth. Particulate concentrations of  $^{231}\text{Pa}_{\text{xs}}$  ranged between 0.0004  
329 and 0.08 fg/kg and particulate  $^{230}\text{Th}_{\text{xs}}$  concentrations ranged between 0.001 and 5.6 fg/kg.  
330 Elevated particulate  $^{231}\text{Pa}_{\text{xs}}$  and  $^{230}\text{Th}_{\text{xs}}$  concentrations were observed close to the seafloor at  
331 stations 04, 153 and 161 and in deep waters of the Nansen and Makarov Basins (Fig. 3). Due  
332 to the high abundance of particulate  $^{232}\text{Th}$  at station 04 ( $^{232}\text{Th} = 46 - 2400 \text{ pg/kg}$ ), it was not  
333 possible to estimate the unsupported particulate  $^{231}\text{Pa}_{\text{xs}}$  concentrations for this station (see  
334 above).

335 In the interior Nansen (Station 50), the distribution of particulate  $^{231}\text{Pa}_{\text{xs}}$  and  $^{230}\text{Th}_{\text{xs}}$   
336 displayed an increase with depth until ~3000 m, followed by a decrease in particulate  
337 concentrations approaching the seafloor. At station 32, concentrations of particulate  $^{230}\text{Th}_{\text{xs}}$   
338 ranged between 0.02 and 5.6 fg/kg, which was similar to concentrations observed at station  
339 50 (0.28 – 5.9 fg/kg). In contrast, particulate  $^{231}\text{Pa}_{\text{xs}}$  (>2000 m) was lower at the slope (0.05 –  
340 0.08 fg/kg) compared to the interior (0.13 – 0.18 fg/kg) (Fig. 3).

341 The distribution of particulate  $^{230}\text{Th}_{\text{xs}}$  observed at station 101 was very similar to the  
342 slope and interior of the Nansen Basin, while, below 1500 m, the particulate  $^{231}\text{Pa}_{\text{xs}}$   
343 concentrations were lower in the Makarov Basin compared to the stations located in the  
344 Nansen Basin (Fig. 3). As this manuscript was already submitted, the intercomparison work  
345 revealed that the particulate  $^{231}\text{Pa}$  concentrations at station 101 are ~ 50% lower than at the  
346 nearby station of the USCGC Healy HLY1502 (GN01) cruise in 2015 (while particulate Th  
347 isotopes are essentially similar at the 2 stations). Therefore, the interpretation of particulate Pa  
348 data requires caution.

349 Before we can compare particulate  $^{230}\text{Th}_{\text{xs}}$  and  $^{231}\text{Pa}_{\text{xs}}$  in the water column with  
350 measurements in the surface sediment, we must first evaluate if an age correction is required  
351 for the sediment samples, as the sedimentation rate in the Arctic Ocean can be very low.  
352 Sedimentation rate estimates in the Makarov Basin range from 0.4 to 4 cm/ky (e.g. Nowaczyk  
353 et al., 2001). It corresponds at most to a mean age of 1.2 ky for the 1<sup>st</sup> cm of the sediment  
354 core, so that it is not necessary to correct the  $^{231}\text{Pa}_{\text{xs}}/^{230}\text{Th}_{\text{xs}}$  ratio for radioactive decay in the  
355 sediment. The higher sedimentation rates encountered over the Barents Sea and slope make  
356 age corrections negligible (Ivanova et al., 2002). We have determined the  $^{238}\text{U}$  and  $^{234}\text{U}$  content  
357 of the 3 sediments (Tab. ES3). The  $[^{234}\text{U}/^{238}\text{U}]$  activity ratio of the 3 sediments is slightly below  
358 equilibrium suggesting the absence of seawater derived U, in agreement other arctic  
359 sediments (Hillaire-Marcel et al., 2017). Hence, we can estimate the  $[^{231}\text{Pa}_{\text{xs}}/^{230}\text{Th}_{\text{xs}}]$  activity  
360 ratio more precisely than based on the  $[^{238}\text{U}/^{232}\text{Th}]$  range determined for the suspended  
361 particles.

### 362 3.5 The Fractionation factor

363 The fractionation factor ( $F_{\text{Th/Pa}}$ ) was calculated as follows:

364  
365 
$$F_{\text{Th/Pa}} = \left( \frac{^{230}\text{Th}_{\text{xs}}/^{231}\text{Pa}_{\text{xs}}}{\text{p}} \right) / \left( \frac{^{230}\text{Th}_{\text{xs}}/^{231}\text{Pa}_{\text{xs}}}{\text{d}} \right) \quad (3)$$
  
366

367 Fractionation factors obtained in this study ranged between 2.7 and 25.4 (Fig. 4).  
368 They are comparable to values reported from other ocean basins (e.g. Hayes et al., 2015a;  
369 Moran et al., 2002, 2001; Scholten et al., 2008). The distribution of  $F_{\text{Th/Pa}}$  with depth was rather  
370 constant (Fig. 4). Below 1000 m depth, fractionation factors were consistently lower at station



371 50 in the interior of the Nansen Basin (2.7 – 13.3) compared to the Nansen margin (3.7 – 22.3)  
372 and the Makarov Basin (7.2 – 25.4) (Fig. 4).

## 373 4 Discussion

### 374 4.1 $^{230}\text{Th}_{\text{xs}}$ and $^{232}\text{Th}$ scavenging in the Arctic Ocean

375 Early studies of  $^{210}\text{Pb}_{\text{xs}}$  and  $^{230}\text{Th}_{\text{xs}}$  in Arctic sediments highlighted that their inventory in the  
376 sediments exceeded the supply from the overlying water column near the slopes and shelves  
377 and were greater at the margins compared to the interior basins (Huh et al., 1997; Smith et al.,  
378 2003). This process, usually referred to as boundary scavenging was expected to be  
379 pronounced in the Arctic Ocean due to the high proportion of shelf areas with the associated  
380 high particle flux versus the low particle flux regions in the ice-covered interior basins (Bacon  
381 et al., 1989; Cochran et al., 1995; Edmonds et al., 2004; Scholten et al., 1995). Early studies  
382 estimated the Th scavenging rates using a simple 1D production-vertical scavenging model  
383 (Bacon et al., 1989; Edmonds, 1998; Scholten et al., 1995), but this view was later challenged  
384 to include the effect of lateral transport, the effect of deep water ventilation (Scholten et al.,  
385 1995) and boundary scavenging (Roy-Barman, 2009). Box-modelling using sedimentary and  
386 dissolved  $^{230}\text{Th}_{\text{xs}}$  and  $^{231}\text{Pa}_{\text{xs}}$  data suggests that ~90 % of the *in-situ* produced  $^{230}\text{Th}_{\text{xs}}$  was  
387 removed within the Arctic Ocean by particle scavenging and that a large fraction of the  
388 scavenged  $^{230}\text{Th}_{\text{xs}}$  was removed by boundary scavenging along the Arctic margins (Moran et  
389 al., 2005, Roy-Barman, 2009). The dataset presented here will be used to revisit our  
390 understanding of the scavenging processes in the Arctic Ocean. We start the discussion with  
391  $^{230}\text{Th}$  because it is highly particle reactive (relative to Pa) and currently used to study and  
392 identify variations in scavenging intensity (e.g. Roy-Barman, 2009; Hayes et al., 2015).

393

#### 394 4.1.1. The Makarov Basin

395 When lateral transport can be neglected, the 1D reversible scavenging model predicts a linear  
396 increase of both dissolved  $^{230}\text{Th}_{\text{xs}}$  ( $^{230}\text{Th}_{\text{xs-d}}$ ) and particulate  $^{230}\text{Th}_{\text{xs}}$  ( $^{230}\text{Th}_{\text{xs-p}}$ ) concentrations  
397 with depth and consequently produces a constant  $^{230}\text{Th}_{\text{xs-p}}/^{230}\text{Th}_{\text{xs-d}}$  ratio throughout the water  
398 column (Bacon and Anderson, 1982). Among all stations reported here, these conditions seem  
399 best fulfilled at station 101 in the Makarov Basin, particularly above 2000 m depth (Fig. 2 and  
400 3). Station 101 is located in the Makarov Basin at high latitude isolated from continental  
401 margins. Therefore St. 101 can be used as a reference station to evaluate the effect of  
402 continental margins on the scavenging in the interior ocean. Neglecting horizontal transport,

403 the  $^{230}\text{Th}_{\text{xs-p}}$  concentration is a function of the settling rate ( $S$ ) of small particles and of the water  
404 depth ( $h$ ):

$$406 \quad S = \frac{P_{230\text{Th}} \times h}{^{230}\text{Th}_{\text{xs-p}}} \quad (4)$$

407  
408 where the  $P_{230\text{Th}}$  is the production rate of  $^{230}\text{Th}$  in the water column (0.56 fg/kg/y). The settling  
409 rate ( $S$ ) of particles ( $> 0.8 \mu\text{m}$ ) in the Makarov Basin ranged between 340 - 440 m/y, which is  
410 consistent with earlier estimates in the central Arctic Ocean (e.g. Edmonds et al., 2004;  
411 Scholten et al., 1995) and very close to new estimates e.g.  $S = 434 \text{ m/y}$  (Rutgers Van Der  
412 Loeff et al., 2018) However, it may represent an upper limit for the particle settling speed in the  
413 central Arctic, because this calculation neglects the possible role of boundary scavenging  
414 (Roy-Barman, 2009). The scavenging residence time of  $^{230}\text{Th}_{\text{xs}}$  ( $\sim 55 \text{ y}$ ) is much shorter than  
415 the water ventilation residence time in the Makarov Basin (e.g. Scholten et al., 1995). Since  
416 particle fluxes in the Makarov Basin are low and the water residence time is high, dissolved  
417  $^{230}\text{Th}_{\text{xs}}$  concentrations have more time to build up by the decay of uranium and stay in solution  
418 (Fig. 2). The particulate/total ratio of  $^{230}\text{Th}_{\text{xs}}$  is relatively low (6-16%) and tends to increase with  
419 depth at station 101 (Fig. 5).

420 Below 2000 m depth, dissolved  $^{230}\text{Th}_{\text{xs}}$  displayed more invariant concentrations  
421 followed by decreasing concentrations approaching the seafloor (Fig. 2). The ventilation of the  
422 deep Arctic could contribute to produce such concave profiles (Scholten et al., 1995). However,  
423 the ventilation age of the deep Makarov Basin is hundreds of years, which is much longer  
424 compared to the  $^{230}\text{Th}$  scavenging residence time (e.g. Rutgers van der Loeff et al., 2018).  
425 These depleted concentrations of dissolved  $^{230}\text{Th}_{\text{xs}}$  and  $^{232}\text{Th}$  close to the seafloor were  
426 accompanied with elevated concentrations of particulate  $^{230}\text{Th}_{\text{xs}}$  and  $^{232}\text{Th}$  (Fig. 3), suggesting  
427 enhanced removal of dissolved  $^{230}\text{Th}_{\text{xs}}$  close to the seafloor. No benthic nepheloid layer was  
428 detected by transmission (Fig. ES4). However, even a slight increase in resuspended  
429 sediments (not detectable by the transmissometer) may be sufficient to enhance scavenging  
430 and removal of dissolved  $^{230}\text{Th}$  close to the seafloor.

#### 431 4.1.2. The Eurasian Basin

432 A sharp depletion of  $^{230}\text{Th}_{\text{xs}}$  observed in deep waters ( $>2000 \text{ m}$ ) of the Nansen was recently  
433 attributed to removal of  $^{230}\text{Th}_{\text{xs}}$  related to release of dissolved iron from hydrothermal vents at  
434 the Gakkel ridge (Valk et al., 2018). Therefore, the following discussion will mainly be divided  
435 into water masses above and below 2000 m depth.

436

#### 437 4.1.2.1 Waters above 2000 m

438 On the Barents shelf, the temperature and salinity at station 153 and 161 clearly indicate the  
439 inflow of saltier and warmer AW (Fig. ES2). As a consequence, the dissolved  $^{230}\text{Th}_{\text{xs}}$   
440 concentrations (Fig. 2) compare well to surface concentrations from the northern Atlantic  
441 Ocean (Hayes et al., 2015a). Close to the seafloor at station 153 and 161 elevated particulate  
442  $^{230}\text{Th}_{\text{xs}}$  and  $^{232}\text{Th}$  concentrations were observed (Fig. ES2). The increased particulate fraction  
443 of  $^{230}\text{Th}_{\text{xs}}$  at the bottom of the water column accompanied with reduced dissolved  $^{230}\text{Th}_{\text{xs}}$   
444 indicates bottom scavenging over the shelf (Fig. 2 and 3). The low beam transmission (300 –  
445 450 m depth) reflects resuspension of bottom sediments resulting in enhanced scavenging  
446 (Fig. ES4). This probably happens due to the inflow of AW over the Barents Sea shelf which  
447 disrupts the sediments close to the seafloor (Lukashin and Shcherbinin, 2007).

448 At stations 04 and 18, the polar waters was less salty and colder compared to the  
449 Atlantic inflow, suggesting influence of ice melt and runoff close to the coast of Svalbard (Fig.  
450 3). This was also reflected by the low beam transmission in the surface and bottom waters  
451 over the Barents shelf, indicating the presence of suspended particulate material (Fig. ES4).  
452 In addition, extremely high concentrations of particulate  $^{232}\text{Th}$  (up to ~2400 pg/kg) and elevated  
453 concentrations of dissolved  $^{232}\text{Th}$  (100 – 200 pg/kg) were observed at station 04 and 18,  
454 suggesting that the particulate material is dominated by lithogenic inputs and partial dissolution  
455 of these particles (Fig. 2). Nevertheless, the dissolved  $^{230}\text{Th}_{\text{xs}}$  concentrations at stations 04 and  
456 18 compare well with those of stations 153 and 161 suggesting that the water flow rate does  
457 not leave enough time for net scavenging to occur.

458 Station 32 and 40 are located within the FSBW, where the modified Atlantic inflow  
459 (~2.5°C) can be recognized between ~100 and 1000 m depth, followed by colder and less salty  
460 deep waters (Fig. ES2). Concentrations of dissolved  $^{230}\text{Th}_{\text{xs}}$  in surface waters at station 32 and  
461 40 (2.3 – 2.7 fg/kg) were within the range but on the high side of North Atlantic ( $^{230}\text{Th}_{\text{xs}}$  = 0.76  
462 – 3.4 fg/kg, Hayes et al., 2015) and Norwegian Sea ( $^{230}\text{Th}_{\text{xs}}$  = 0.6 – 2.3 fg/kg, Moran et al.,  
463 1995) values.

464 At station 32 and 40, dissolved  $^{230}\text{Th}_{\text{xs}}$  increases linearly with depth until ~1500 m and  
465 then becomes constant (Fig. 2). This departure from the equilibrium profile suggests the  
466 possibility of enhanced scavenging at the margin. By contrast, Station 50 (Valk et al., 2018) is  
467 located within the return flow of the FSBW (Fig. ES2 and ES3). Dissolved  $^{230}\text{Th}_{\text{xs}}$   
468 concentrations increase linearly down to 2000 m and they are higher than at station 32 and 40  
469 indicating a lower scavenging rate. Surprisingly, the distribution of particulate  $^{230}\text{Th}_{\text{xs}}$  was very  
470 similar at stations 32 and 50 (and also at station 101) (Fig. 3).

471 Elevated concentrations of particulate  $^{232}\text{Th}$  at the margin (St. 32) compared to the  
472 inner ocean (St. 50) are likely due to the advection of shelf waters (St. 4) transporting  $^{232}\text{Th}$   
473 into the interior basin.

474 The dissolved  $^{230}\text{Th}_{\text{xs}}$  concentrations were considerably lower in the Amundsen basin  
475 (station 125) compared to the Makarov basin (station 101) (Fig. 2), as shown previously  
476 (Scholten et al., 1995; Valk et al., 2018). This reflects the large difference in water residence  
477 times between the Makarov and Amundsen Basins (Schlosser et al., 1997), where the longer  
478 water residence time in the Makarov Basin allows the concentrations of  $^{230}\text{Th}_{\text{xs}}$  to build up over  
479 time by the decay of U, whereas waters from the Amundsen Basin were submitted to boundary  
480 scavenging while flowing along the eastern Siberian margin.

481 Except the surface samples (<100 m), the dissolved  $^{230}\text{Th}_{\text{xs}}$  concentrations are  
482 significantly lower in the Nansen and Amundsen Basins (St. 50, 40, 32 125) compared to the  
483 Makarov Basin (St. 101). Within the Eurasian Basins (above 2000 m), a horizontal gradient of  
484 dissolved  $^{230}\text{Th}_{\text{xs}}$  content between stations 125, 32, 40 and 50 was observed (Fig. 2). At station  
485 50, dissolved  $^{230}\text{Th}_{\text{xs}}$  concentrations were highest, followed by intermediate concentrations at  
486 the Nansen margin (St. 32, 40) and the lowest dissolved  $^{230}\text{Th}_{\text{xs}}$  were observed at station 125.  
487 This suggests that  $^{230}\text{Th}_{\text{xs}}$  is scavenged along the boundaries as the dissolved  $^{230}\text{Th}_{\text{xs}}$  is  
488 decreasing towards the margin. The water at station 125 has probably experienced more  
489 scavenging on the Kara and Laptev shelf compared to the Barents shelf. Another possible  
490 reason for the low  $^{230}\text{Th}_{\text{xs}}$  concentrations observed at 125 is scavenging onto particles carried  
491 by the TPD (Trans Polar Drift) (Charette, pers. com.).

492

### 493 **Below 2000 m**

494 Below 2000 m, the  $^{230}\text{Th}_{\text{xs}}$  concentrations are relatively constant and identical within the  
495 Eurasian Basin. The sharp decrease at station 50 is clearly due to hydrothermal scavenging  
496 (Valk et al., 2018). In 2015, there was no sign of a strong hydrothermal activity in the beam  
497 transmission data suggesting that the hydrothermal event was over at the time of sampling  
498 and that the hydrothermal plume had faded away (Fig. ES4). The major part of the  
499 hydrothermal plume in the basin is expected to be transported out of the Nansen Basin along  
500 the Gakkel Ridge directly towards Fram Strait (Fig. ES3). Therefore, the distribution of  
501 dissolved  $^{230}\text{Th}_{\text{xs}}$  at stations 32, 40 and 125 are not directly downstream of station 50 and their  
502  $^{230}\text{Th}_{\text{xs}}$  do not necessarily represent hydrothermal scavenging conditions. Valk et al., (2018)  
503 identified hydrothermal plume water by their high dissolved Fe content. The dissolved Fe  
504 content of the deep waters at station 32 are higher than at station 50: it does not correspond

505 to an hydrothermal source, but to sediment resuspension at the slope as a source of dissolved  
506 Fe (Klunder et al., 2012, Rijkenberg et al., 2018).

507 An additional argument for deep enhanced scavenging comes from  $^{232}\text{Th}$ . In general,  
508 dissolved  $^{232}\text{Th}$  concentrations increases with depth in the deep ocean (e.g. Moran et al., 2002,  
509 Scholten et al., 2008, Okubo et al., 2013). In the present study,  $^{232}\text{Th}$  decrease with depth, a  
510 feature that was already observed in the Arctic (Edmonds et al., 2004), but remained  
511 unexplained. The lack of increasing dissolved  $^{232}\text{Th}$  concentrations towards the seafloor  
512 suggests enhanced bottom scavenging. The lowest dissolved  $^{232}\text{Th}$  in the deep basins occurs  
513 at station 50 suggesting the involvement of scavenging by the hydrothermal plume. However,  
514 low deep dissolved  $^{232}\text{Th}$  is also found in the Makarov Basin, where scavenging by a  
515 hydrothermal plume is not suspected. Note that the higher concentrations at station 32 do not  
516 necessarily represent a lower scavenging rate because a high scavenging rate can be  
517 balanced by a high input flux by particle resuspension/dissolution as suggested by the  
518 relatively low beam transmission and high particulate  $^{232}\text{Th}$  at this station.

519

## 520 **The particulate fraction**

521 Despite different dissolved  $^{230}\text{Th}_{\text{xs}}$  profiles at stations 32, 50 (Nansen Basin) and 101 (Makarov  
522 Basin), the particulate  $^{230}\text{Th}_{\text{xs}}$  concentrations above 2500 m depth were similar at these  
523 stations (Fig. 3). By contrast, particulate  $^{232}\text{Th}$  concentrations were higher at station 32  
524 compared to stations 50 and 101, highlighting the role of the continental margins in providing  
525 high levels of particulate matter into the low productivity interior ocean.

526 The particulate fraction of  $^{230}\text{Th}_{\text{xs}}$  increases with depth at stations 32 and 50 from 1%  
527 to 45% with a local maximum (20 %) around 200 m (Fig. 5). This subsurface maximum  
528 particulate fraction can be related to the high  $^{230}\text{Th}_{\text{xs}}$  particulate fractions associated to the  
529 bottom nepheloid layers on the shelf at station 153 and 04. Despite the high concentrations of  
530 particulate  $^{230}\text{Th}_{\text{xs}}$  in the Makarov Basin, the  $^{230}\text{Th}_{\text{xs}}$  particulate fractions are higher at stations  
531 32 and 50 because the dissolved  $^{230}\text{Th}_{\text{xs}}$  concentrations are much lower in the Nansen Basin  
532 compared to the Makarov Basin. At station 32 and 101, the increased particulate fraction of  
533  $^{230}\text{Th}_{\text{xs}}$  at the bottom of the water column accompanied with reduced dissolved  $^{230}\text{Th}_{\text{xs}}$  indicates  
534 bottom scavenging in deep waters of the open basin as it was already observed over the shelf  
535 (Fig. 2 and 3). At station 50, the increased  $^{230}\text{Th}_{\text{xs}}$  particulate fraction of the bottom waters is  
536 the relic of a hydrothermal plume induced scavenging event, possibly at a stage where the  
537 steady state situation is being achieved again (Valk et al., 2018).

538

## 539 4.2 Modelling $^{230}\text{Th}_{\text{xs}}$ scavenging in the Arctic Ocean

540 Several features listed in previous sections of this paper (increasing  $^{230}\text{Th}_{\text{xs}}$  particulate fraction  
541 with depth; decreasing dissolved  $^{230}\text{Th}_{\text{xs}}$  content in the deepest waters of the ocean margin)  
542 are not consistent with the 1D scavenging models that predict a linear increase of dissolved  
543 and particulate  $^{230}\text{Th}_{\text{xs}}$  concentrations with depth and a constant  $^{230}\text{Th}_{\text{xs}}$  particulate fraction with  
544 depth (e.g. Bacon and Anderson, 1982, Roy-Barman et al., 1996). They are not consistent with  
545 the boundary scavenging model of Roy-Barman (2009) either, because this model predicts a  
546 rather linear increase of dissolved  $^{230}\text{Th}_{\text{xs}}$  with depth at the margin and a constant  $^{230}\text{Th}_{\text{xs}}$   
547 particulate fraction with depth. This is certainly due to an oversimplification of the particle  
548 dynamics: particles were assumed to have a constant vertical flux and their transport between  
549 the margin and the ocean interior was neglected. To overcome these assumptions, we propose  
550 a boundary scavenging model where particles are introduced both at the ocean surface and  
551 also throughout the water column at the margin (hypothesis 1) and where particles are  
552 transported between the margin and the ocean interior (hypothesis 2). The input of particles at  
553 all depths of the ocean margin can be viewed as a result of nepheloid layers on the shelf or on  
554 the slope and/or to the chemical evolution of particles as they settle through the water column  
555 (precipitation of Fe-Mn oxyhydroxides at depth for example). A key point is that the particles  
556 introduced at depth are assumed to be free of  $^{230}\text{Th}_{\text{xs}}$  and  $^{231}\text{Pa}_{\text{xs}}$  when they are introduced in  
557 the water column, so that the input of particles will not be associated with an input of  $^{230}\text{Th}_{\text{xs}}$   
558 and  $^{231}\text{Pa}_{\text{xs}}$  (hypothesis 3). This means that the particles are not merely resuspended local  
559 bottom sediments, which are known to contain  $^{230}\text{Th}_{\text{xs}}$ . Instead, we hypothesize that nepheloid  
560 layers flow downward as turbidity currents along the slope with no or little mixing with  
561 surrounding waters so that they may not scavenge  $^{230}\text{Th}$  until they detach from the slope. Only  
562 then, particles spread and scavenge  $^{230}\text{Th}_{\text{xs}}$  from the deep waters. This view differs from  
563 Rutgers van der Loeff and Boudreau (1997), who assumed equilibration between seawater  
564 and particles. Hypothesis (3) is required because a dissolved  $^{230}\text{Th}_{\text{xs}}$  depletion compared to  
565 the equilibrium profile cannot be produced by resuspension of sediments that would already  
566 be “equilibrated” with overlying seawater. Hypothesis (3) is crucial for creating a water column  
567  $^{230}\text{Th}_{\text{xs}}$  profile as observed in this study. Transport of particles between the margin and the  
568 inner ocean (hypothesis 2) allows that most particles in the ice-covered central Arctic are  
569 advected from the margins.

570

### 571 4.2.1. Transport of water

572 In this box model, the Arctic Ocean is divided into 2 boxes: the ocean margin and the ocean  
573 interior (Fig. 6). The water volumes of the margin and of the ocean interior are  $V_m$  and  $V_i$  ( $\text{m}^3$ )

574 (Jakobsson, 2002). These 2 boxes exchange a total flux of water  $F$  ( $\text{m}^3/\text{s}$ ). For simplicity, we  
 575 assume that the water flows horizontally between the boxes. Vertical mixing and ventilation of  
 576 water by inputs through Fram strait and Barents Sea are neglected. Hence, at any depth in the  
 577 water column the residence time of the water with respect to horizontal exchange is  $\tau_m = V_m/F$   
 578 at the margin and  $\tau_i = V_i/F$  at the ocean interior (Tab. 1). The time constant associated to water  
 579 transport are  $k_m = 1/\tau_m$  and  $k_i = 1/\tau_i$ . The time constant associated with water exchange between  
 580 the ocean margin and the ocean interior are  $k_m = 1/\tau_m$  for the ocean margin and  $k_i = 1/\tau_i$  for the  
 581 ocean interior.

#### 582 4.2.2. Particle transport

583 In this study, the concentration of particles was not measured. The particle concentration is  
 584 embedded in the dissolved-particulate partition coefficient  $K$ , introduced in the following  
 585 section. We scale the impact of particle concentration on the partition coefficient  $K$  with a  
 586 parameter  $m$  assumed to be proportional to the particulate concentration:

587

$$588 \quad m = P(z)/P^m(0) \quad (5)$$

589 where  $P$  is the particle concentration (at the margin or in the ocean interior) and  $P^m(0)$   
 590 is the particle concentration in the surface water of the ocean margin. Hence,  $m^m = 1$  in the  
 591 surface water of the ocean margin and  $m$  is proportional to the particle concentration  
 592 elsewhere.

593 The conservation equations for  $m^m$  and  $m^i$  at the ocean margin and in the inner ocean  
 594 include particle production throughout the water column at the margin (hypothesis 1) and  
 595 particle transport between the margin and the inner ocean (hypothesis 2) and are given by:

$$596 \quad \frac{dm^m}{dt} = -S^m \frac{dm^m}{dz} + k_m(m^i - m^m) + \mu \times m^m \quad (6)$$

$$597 \quad \frac{dm^i}{dt} = -S^i \frac{dm^i}{dz} + k_i(m^m - m^i) \quad (7)$$

598 where  $S^m$  and  $S^i$  are the settling velocities of the particles at the margin and in the  
 599 inner ocean. They are both assumed to be constant with depth. We expect higher particle  
 600 settling rates at the margin compared to the interior due to increased production and the  
 601 associated particle flux (Anderson et al., 1983).  $\mu$  is an arbitrary parameter aimed to produce  
 602 an exponential increase of the particle concentration in the deep waters.  $\mu$  operates as if (1)  
 603 increasing turbulence towards the seafloor and increases the particle concentration by  
 604 formation of nepheloids and/or (2) a diffusive flux of dissolved Manganese (Mn) from the

605 sediment which allows precipitation of Mn oxides (that scavenges Pa and Th) towards the sea  
 606 floor. Assuming a steady state, we obtain:

$$607 \quad \frac{dm^m}{dz} = \frac{k_m}{S^m} (m^i - m^m) + \frac{\mu}{S^m} \times m^m \quad (8)$$

$$608 \quad \frac{dm^i}{dz} = \frac{k_i}{S^i} (m^m - m^i) \quad (9)$$

609 Bulk dissolution of particles was not considered. The observed increasing particulate  $^{230}\text{Th}$   
 610 fraction with depth points to an addition of particles with depth rather than a significant  
 611 dissolution with depth, which would induce a concave shaped  $^{230}\text{Th}$  profile (Roy-Barman et al.,  
 612 1996). Moreover, there is no direct constraint on the particle dissolution in the present study.

#### 613 4.2.3. Transport of $^{230}\text{Th}_{\text{xs}}$ and $^{231}\text{Pa}_{\text{xs}}$

614 In each box,  $^{230}\text{Th}$  and  $^{231}\text{Pa}_{\text{xs}}$  are produced by *in-situ* decay of U at a constant rate  $P$ . The  
 615 produced  $^{230}\text{Th}$  and  $^{231}\text{Pa}_{\text{xs}}$  are then transported towards the seafloor by reversible scavenging  
 616 onto sinking particles and transported horizontally by the flow of water (Fig. 6). Considering  
 617 the long half-life of  $^{230}\text{Th}$  (75 000 y) and  $^{231}\text{Pa}$  (32 700 y), the radioactive decay of the two  
 618 isotopes was neglected. Dissolved and particulate concentrations are noted as  $C_d^m$  and  $C_p^m$  for  
 619 the margin and  $C_d^i$  and  $C_p^i$  for the ocean interior. The conservation equation of total  $^{230}\text{Th}$  is  
 620 given by:

621 The ocean margin:

$$622 \quad \frac{d(C_d^m + C_p^m)}{dt} = -S^m \frac{dC_p^m}{dz} + k_m ([C_d^i + C_p^i] - [C_d^m + C_p^m]) + P_{230} \quad (10)$$

623 The ocean interior:

$$624 \quad \frac{d(C_d^i + C_p^i)}{dt} = -S^i \frac{dC_p^i}{dz} + k_i ([C_d^m + C_p^m] - [C_d^i + C_p^i]) + P_{230} \quad (11)$$

625 At the margin (equation 10), the only source term is the *in-situ* production because  
 626 the particles introduced in the water column of the margin do not contain  $^{230}\text{Th}_{\text{xs}}$  (hypothesis  
 627 3). The relationship between  $C_d$  and  $C_p$  is obtained by assuming a reversible equilibrium  
 628 between dissolved and particulate Th:

$$629 \quad C_p^i = K \times m^i \times C_d^i \quad (12)$$

$$630 \quad C_p^m = K \times m^m \times C_d^m \quad (13)$$



631 where  $K$  is the equilibrium coefficient of Th or Pa between the particulate fraction  
 632 (mass of radionuclide carried by particles per L of seawater) and the dissolved fraction (mass  
 633 of radionuclide in solution per L of seawater) ( $K = \text{concentration in particles}/\text{concentration in}$   
 634  $\text{the dissolved phase}$ ).  $K$  is assumed to be constant with depth and particle concentration.  
 635 Assuming a steady state, we obtain:

$$636 \frac{d(m^m C_d^m)}{dz} = \frac{1}{S^m} \left\{ k_m \left( (1 + K \times m^i) C_d^i - (1 + K \times m^m) C_d^m \right) + P_{230} \right\} \quad (14)$$

$$637 \frac{d(m^i C_d^i)}{dz} = \frac{1}{S^i} \left\{ k_i \left( (1 + K \times m^m) C_d^m - (1 + K \times m^i) C_d^i \right) + P_{230} \right\} \quad (15)$$

638 Equations 10, 11, 14 and 15 are solved numerically. We use circulation parameters  
 639 already obtained for modelling the boundary scavenging in the Arctic Ocean (Roy-Barman,  
 640 2009). The water residence time with respect to horizontal exchange is 10 y for the ocean  
 641 margin and 50 y for the ocean interior (Tab. 1). All the other parameter are adjusted by trial  
 642 and error in order to obtain a reasonable agreement (Fig. 7) with the dissolved and particulate  
 643 profiles of station 32 (Nansen margin) and station 50 (Nansen interior, above 2000 m to avoid  
 644 the hydrothermal scavenging that is not represented in the model). Qualitatively equivalent  
 645 results were obtained for station 32 and station 101 (not shown). The boundary conditions for  
 646 the particle concentration are  $m^m(0) = 1$  (by definition of  $m$ ) and  $m^i(0) = 1.5$  to obtain a  
 647 reasonable fit between the model and the  $^{230}\text{Th}$  profile (see discussion below). This implies  
 648 that there is an input of particles in the surface waters at the margin and in the interior ocean.  
 649 We use  $\mu = 0.5 \text{ y}^{-1}$ . As the particle settling speed at the margin is set a 600 m/y, it corresponds  
 650 to an increase of the particle concentration at the margin of a factor 2.71 every 1200 m of  
 651 depth. By default, we choose the same value for  $K (= 0.11)$ , at the margin and in the inner  
 652 ocean. If we want to reproduce the similar vertical  $^{230}\text{Th}_p$  gradients observed at the margin and  
 653 in the inner ocean (Fig. 3), the settling velocity of the particles must be higher at the margin  
 654 (600 m/y) compared to the ocean interior (340 m/y). Indeed, if  $\frac{dC_p^m}{dz} \approx \frac{dC_p^i}{dz}$ , equations 14 and 15  
 655 can be combined to yield:

$$656 \frac{S^i}{S^m} \approx \frac{P_{230} - k_i (C_{d+p}^i - C_{d+p}^m)}{P_{230} + k_m (C_{d+p}^i - C_{d+p}^m)} \quad (16)$$

657 As  $C_{d+p}^i - C_{d+p}^m > 0$ , it follows that  $S^i < S^m$

658 In other words,  $S^i < S^m$  because at the margin, an excess of  $^{230}\text{Th}$  advected from the  
 659 inner ocean must be removed by settling particles with the same vertical  $^{230}\text{Th}_p$  gradient as in  
 660 the inner ocean. Conversely, in the inner ocean, less  $^{230}\text{Th}$  must be removed by settling

661 particles with the same vertical  $^{230}\text{Th}_p$  gradient as at the margin. Note that  $S^i = S^m$ , only if  $k_i =$   
662  $k_m = 0$ . This corresponds to the 1D reversible scavenging model that would not account for the  
663 increasing  $^{230}\text{Th}$  particulate fraction with depth and for the non-linear dissolved  $^{230}\text{Th}$  profiles.

664 Given the simplicity of the model, we do not expect a full agreement with the data  
665 despite some tuning of the parameters. For example, the hydrothermal scavenging of  $^{230}\text{Th}_{xs}$   
666 below 2000 m depth, possibly occurring at station 50 is not represented in the model.  
667 Nevertheless, we believe that the model captures some effects of particle accumulation and  
668 scavenging at depth. This is in line with 3D modelling results showing that dissolved Th and  
669 Pa profiles in the Atlantic Ocean are better simulated when a parametrization of boundary and  
670 bottom scavenging is introduced (Rempfer et al., 2017).

671 As observed in the data, the modelled dissolved  $^{230}\text{Th}_{xs}$  concentration profiles  
672 increase with depth followed by a decrease in concentration approaching the seafloor (Fig. 7).  
673 The model also captures the linear increase of the particulate  $^{230}\text{Th}_{xs}$  concentrations with depth,  
674 as observed in the data (Fig. 7). Despite the reversible scavenging equilibrium hypothesis,  
675 dissolved and particulate  $^{230}\text{Th}_{xs}$  are not proportional because the mass of particles “ $m$ ”  
676 increases with depth (eq. 1). Therefore, the linear particulate  $^{230}\text{Th}_{xs}$  profiles result from the  
677 gross linear increase of the  $^{230}\text{Th}_{xs}$  *in-situ* production with depth (it is not perfectly linear due to  
678 lateral transport). Despite this linear increase of the particulate  $^{230}\text{Th}_{xs}$ , the dissolved  $^{230}\text{Th}_{xs}$   
679 concentrations can decrease with depth due to the increased particulate scavenging. As a  
680 consequence of the dissolved and particulate profile shapes, the particulate/total fraction of  
681  $^{230}\text{Th}_{xs}$  increases with depth as observed in the data (Fig. 5 and 7).

682 The measured particulate  $^{230}\text{Th}_{xs}$  concentrations are relatively similar at stations 32,  
683 50 and 101 (Fig. 3). This is surprising and probably circumstantial: the high particle mass  
684 equilibrating with a low dissolved  $^{230}\text{Th}_{xs}$  content at the margin balances the lower mass of  
685 particles equilibrating with a higher dissolved  $^{230}\text{Th}_{xs}$  concentration at the interior ocean. A  
686 direct consequence of these similar particulate  $^{230}\text{Th}_{xs}$  profiles is that the vertical flux of  $^{230}\text{Th}_{xs}$   
687 is increased at the margin where the particle settling velocity is higher compared to the ocean  
688 interior. Hence, boundary scavenging occurs.

689

#### 690 4.3 $^{231}\text{Pa}_{xs}$ profiles: scavenging versus circulation

691  $^{231}\text{Pa}_{xs}$  is less particle reactive compared to  $^{230}\text{Th}_{xs}$ , as indicated by the lower  $^{231}\text{Pa}_{xs}$  particulate  
692 fraction compared to the  $^{230}\text{Th}_{xs}$  particulate fraction (Fig. 5). As a consequence,  $^{231}\text{Pa}_{xs}$  profiles  
693 generally deviate more strongly from the linear increase expected from reversible scavenging  
694 due to a significant transport by advection (e.g. Hayes et al., 2015; Gdaniec et al., 2017). In  
695 the Arctic, Moran et al. (2005) estimated that 39 % of the  $^{231}\text{Pa}_{xs}$  produced in the Arctic Ocean  
696 is expected to be exported to the Atlantic Ocean through the Fram Strait. However, in the

697 Makarov Basin, ventilation cannot account for the non-linear  $^{231}\text{Pa}$  profile, as the residence  
698 time of the deep water is several hundreds of years (e.g. Rutgers van der Loeff et al., 2018).  
699 Therefore, this non-linear profile may be related to scavenging along the basin boundaries  
700 (Roy-Barman, 2009).

701 On the Barents Shelf (St. 153 and 161), representing the inflow of unmodified Atlantic  
702 waters, the dissolved  $^{231}\text{Pa}_{\text{xs}}$  is in the same range as the concentrations ( $^{231}\text{Pa}_{\text{xs}} = 0.33 - 0.68$   
703 fg/kg) measured in the northeastern Atlantic (Hayes et al., 2015). At stations 04 and 18, the  
704 dissolved  $^{231}\text{Pa}_{\text{xs}}$  (0.19 – 0.36 fg/kg) becomes slightly lower, possibly reflecting scavenging of  
705  $^{231}\text{Pa}_{\text{xs}}$  on the Barents Shelf (Fig. 2). Pa scavenging over the Barents Shelf is not surprising  
706 because the area is known for its diatom blooms (Wassmann et al., 1990). The dissolved  
707  $^{231}\text{Pa}_{\text{xs}}$  in shallow waters (<500 m) over the slope (St. 32) and the Nansen Basin (stations 40  
708 and 50) ( $^{231}\text{Pa}_{\text{xs}} = 0.20 - 0.4$  fg/kg) are intermediate between the Atlantic inflowing water  
709 (stations 153 and 161) and close to Svalbard (St. 4).

710 As for the  $^{230}\text{Th}_{\text{xs}}$  (and  $^{232}\text{Th}$ ) in bottom waters of station 153 and 161, depleted  
711 concentrations of dissolved  $^{231}\text{Pa}_{\text{xs}}$  were accompanied with elevated particulate  $^{231}\text{Pa}_{\text{xs}}$   
712 concentrations (Fig. 2 and 3), indicating removal of  $^{231}\text{Pa}_{\text{xs}}$  at the Barents shelf, close to the  
713 seafloor. The particulate fraction of  $^{231}\text{Pa}_{\text{xs}}$  in deep waters of the interior Nansen was very small  
714 (0.2-0.9 %) compared to  $^{230}\text{Th}_{\text{xs}}$  (25-48 %), which is expected due to the overall lower particle  
715 reactivity of  $^{231}\text{Pa}$  (compared to  $^{230}\text{Th}$ ).

716 Extremely low dissolved  $^{231}\text{Pa}_{\text{xs}}$  concentrations ( $0.02 \pm 0.005$  fg/kg) were observed in  
717 surface waters (10 – 100 m) of station 101 (Fig. 2). As the high  $^{232}\text{Th}$  content of these waters  
718 was attributed to advection by the transpolar drift (e.g. Rutgers van der Loeff et al., 2018), it is  
719 likely that these waters were completely depleted of their  $^{231}\text{Pa}$  over the shelf and that during  
720 their transit to station 101 reversible scavenging has acted to build up a linear equilibrium  
721 profile. The sea-ice cover has prevented this equilibrium profile (>100 m) from re-  
722 homogenization by wind-induced mixing.

723 In the deep basins, deep/bottom scavenging also occurs, as suggested by the  
724 elevated particulate  $^{231}\text{Pa}_{\text{xs}}$  concentrations below 2000 m depth observed at station 32 and 50  
725 that indicate removal of  $^{231}\text{Pa}_{\text{xs}}$  in deep waters of the Nansen Basin (Fig. 3). However, in  
726 contrast to  $^{230}\text{Th}_{\text{xs}}$ , higher  $^{231}\text{Pa}_{\text{xs}}$  particulate fractions were observed at station 50 relative to  
727 station 32 (Fig. 5), suggesting the presence at station 50 of hydrothermal particles which are  
728 known to have a high affinity for Pa.

729 Unlike dissolved  $^{230}\text{Th}_{\text{xs}}$  profiles, the dissolved  $^{231}\text{Pa}_{\text{xs}}$  profiles in the Nansen and  
730 Amundsen Basin have similar shapes probably due to the much longer scavenging residence  
731 time of Pa relative to Th. By contrast,  $^{231}\text{Pa}_{\text{xs}}$  concentrations are higher in the intermediate/deep  
732 Makarov Basin (St. 101) than in the Eurasian Basin due to the long term isolation of these 2  
733 basins by the Lomonosov ridge (Fig. 2). Interestingly, there are two distinct correlations

734 between dissolved silica (DSi) and dissolved  $^{231}\text{Pa}_{\text{xs}}$  in the deep waters of the Nansen and the  
735 Makarov Basins (Fig. 8). These positive correlations are likely due to the high affinity of  $^{231}\text{Pa}$   
736 for biogenic silica (BSi) (e.g. Chase et al., 2002). We propose that  $^{231}\text{Pa}$  is scavenged by BSi  
737 formed in surface waters. As these particles settle through the water column, BSi dissolves  
738 and releases  $^{231}\text{Pa}$  into solution again. The steeper slope of the dissolved  $^{231}\text{Pa}_{\text{xs}}$  versus DSi  
739 correlation in the Makarov Basin compared to the Nansen Basin is likely due to accumulation  
740 of *in-situ* produced  $^{231}\text{Pa}$  (in addition to the BSi dissolution effect) during the longer aging of  
741 the deep Makarov/Canadian waters.

742

#### 743 4.4. Applying the boundary scavenging model to Pa

744 The boundary scavenging model developed for  $^{230}\text{Th}$  is now applied to  $^{231}\text{Pa}$  with the  
745 appropriate changes (Tab. 1). The production rate is changed (0.025 fg/kg/y) and the  
746 dissolved-particulate partition coefficient is divided by 12 because we have estimated that  
747  $F_{\text{Th/Pa}} \sim 12$  (Fig. 4). Several similarities and differences arise between the model and measured  
748 data.

749 The model reproduces the non-linear shape of the dissolved  $^{231}\text{Pa}_{\text{xs}}$  profiles  
750 (particularly at St. 50) and the relatively linear particulate  $^{231}\text{Pa}_{\text{xs}}$  profiles of station 32 and 50.  
751 However, the modelled dissolved  $^{231}\text{Pa}_{\text{xs}}$  concentrations are overestimated by a factor 2 to 4  
752 (Fig. 7). Modelled particulate  $^{231}\text{Pa}_{\text{xs}}$  concentrations at the margin are also overestimated. The  
753 reason for the overestimated dissolved  $^{231}\text{Pa}_{\text{xs}}$  concentrations is probably a mix of previously  
754 discussed processes and because of the possible underestimation of the particulate Pa  
755 concentration revealed by intercomparison work at station 101 (higher particulate Pa  
756 concentration would result in a higher removal rate by scavenging on settling particles).  
757 Scavenging of  $^{231}\text{Pa}_{\text{xs}}$  by hydrothermal plumes can severely deplete the concentrations of  
758  $^{231}\text{Pa}_{\text{xs}}$  in the deep Arctic Ocean (Valk et al., 2018). Export of  $^{231}\text{Pa}$  through advection has been  
759 proposed to have a significant importance in several studies (e.g. Hoffmann et al., 2013; Moran  
760 et al., 2005). The dissolved  $^{231}\text{Pa}$  concentrations of the deep inflow through the Fram Strait  
761 Branch ( $^{231}\text{Pa} \sim 1 - 1.5$  fg/kg) agrees well with the value ( $^{231}\text{Pa} = 1.5$  fg/kg) used in Moran et al.  
762 (2005). The  $^{231}\text{Pa}$  concentrations reported here for the return in the Nansen Basin at station  
763 50 ( $^{231}\text{Pa} \sim 1 - 1.5$  fg/kg) is close to the Fram Strait Branch. By contrast, in the deep Makarov  
764 Basin, above the sill on the Lomonosov ridge is ( $^{231}\text{Pa} \sim 1.5 - 3.0$  fg/kg) consistent with Moran  
765 et al (2005) estimates. This  $^{231}\text{Pa}$  gradient between the inflow and outflow supports a net export  
766 of  $^{231}\text{Pa}$  into the Atlantic.

767 Finally, boundary scavenging along the slopes of the Eurasian Basin can reduce the  
768 dissolved  $^{231}\text{Pa}$  in the deep Arctic Ocean. Probably,  $^{231}\text{Pa}/^{230}\text{Th}$  ratios of the suspended  
769 particles of the Nansen Basin were below the  $^{231}\text{Pa}/^{230}\text{Th}$  production ratio, but top-core

770 sediments of the Nansen margin and slope have high  $^{231}\text{Pa}/^{230}\text{Th}$ -ratios, suggesting that the  
771 Nansen margin is in fact a large sink for  $^{231}\text{Pa}$  in the Arctic Ocean.

772 Roy-Barman (2009) proposed a balanced Arctic budget of  $^{231}\text{Pa}$  between the inner  
773 ocean and the margin without export of  $^{231}\text{Pa}$  through the Fram Strait. However, it was based  
774 on very low  $F_{\text{Th}/\text{Pa}}$  values ( $\sim 3$ -10) compared to the values reported in the present work.

## 775 4.4 Factors controlling Pa-Th fractionation

### 776 4.4.1 The Th-Pa fractionation factor

777  $^{231}\text{Pa}_{\text{xs}}$  is generally less particle reactive than  $^{230}\text{Th}_{\text{xs}}$ , which results in  $^{231}\text{Pa}_{\text{xs}}$  profiles that  
778 deviate more strongly from the linear increase expected from reversible scavenging due to  
779 transport by advection (e.g. Chase et al., 2002; Hayes et al., 2015; Gdaniec et al., 2017). The  
780 particulate content of  $^{231}\text{Pa}_{\text{xs}}$  relative to the total  $^{231}\text{Pa}_{\text{xs}}$  concentrations was in the order of 0.6  
781 – 1 % in the deep stations (<2000 m) and up to 15% on the shelf, while particulate/total ratios  
782 for  $^{230}\text{Th}_{\text{xs}}$  ranged between 6 % and 17 % in the deep stations and up to 80% on the shelf (Fig.  
783 5). This reflects the preferential scavenging of  $^{230}\text{Th}_{\text{xs}}$  relative to  $^{231}\text{Pa}_{\text{xs}}$ . As a consequence,  
784 the  $F_{\text{Th}/\text{Pa}}$  in the open ocean is typically around 20 (e.g. Hayes et al., 2015b), mostly when  
785 carbonate, organic and lithogenic particles dominates. Notable exceptions are environments  
786 where particulate matter is dominated by diatoms ( $F_{\text{Th}/\text{Pa}} < 5$ , e.g. Scholten et al., 2008;  
787 Venchiarutti et al., 2011a; Venchiarutti et al., 2011b) or Mn-Fe (oxy)hydroxides, such as in  
788 hydrothermal plumes ( $F_{\text{Th}/\text{Pa}} = 6 \pm 3$  for  $\text{MnO}_2$  and  $F_{\text{Th}/\text{Pa}} = 11 \pm 6$  for  $\text{Fe}(\text{OH})_3$ , Hayes et al.,  
789 2015; Pavia et al., 2018). As a consequence of these different drivers, 2 types of  $F_{\text{Th}/\text{Pa}}$  depth  
790 profiles are generally observed:

791 1) In the North Atlantic  $F_{\text{Th}/\text{Pa}}$  decreases with depth because the particulate matter  
792 composition evolves from lithogenic/carbonated in the surface water to strongly affected by  
793 Fe-Mn oxides due to hydrothermal particles above and around the Mid-Atlantic Ridge or to  
794 oxidation/precipitation of Fe-Mn released in the oxygen minimum zone off the Mauritanian  
795 coast.

796 2) In areas dominated by diatom production,  $F_{\text{Th}/\text{Pa}}$  tends to increase with depth as a  
797 consequence of the biogenic silica dissolution (Scholten et al., 2008; Venchiarutti et al., 2011),  
798 because opal tends to dissolve during the settling through the water column (Nelson et al.,  
799 1995). In the Arctic, this view is supported by the strong correlation observed between  
800 dissolved  $^{231}\text{Pa}$  and dissolved Si of the deep Makarov and Nansen Basins (Fig 8).

801 The  $F_{\text{Th}/\text{Pa}}$  profiles that we obtained on the Nansen margin (St. 32, 153 and 161) and  
802 in the Makarov Basin (St. 101) show a clear increase with depth (from  $F_{\text{Th}/\text{Pa}} \approx 2$ -5 in the shallow  
803 waters to  $F_{\text{Th}/\text{Pa}} \approx 20$  in the deep waters) suggesting surface particles dominated by diatoms  
804 and deeper particles with an increasing influence of lithogenic particles resuspended and  
805 advected from the margin. At station 50, the limited increase of  $F_{\text{Th}/\text{Pa}}$  below 2000 m ( $F_{\text{Th}/\text{Pa}} \approx$

806 10) supports that particles at this station still include some particles derived from the  
807 hydrothermal activity, even if at the time of sampling most of the plume itself had left the Gakkel  
808 ridge and cannot be clearly detected with beam transmission data (Valk et al., 2018).

809

#### 810 4.4.2 Large scale Pa-Th fractionation

811 Early studies of Pa-Th in the Arctic Ocean have emphasized that despite of the large shelf  
812 areas, the large Pa-Th fractionation observed in the Pacific Ocean sediments between the  
813 ocean margin ( $[^{231}\text{Pa}_{\text{xs}}/^{230}\text{Th}_{\text{xs}}] \approx 3 \cdot \text{production ratio}$ ) and the ocean interior ( $[^{231}\text{Pa}_{\text{xs}}/^{230}\text{Th}_{\text{xs}}] \approx$   
814  $0.3 \cdot \text{production ratio}$ ) was not present in Arctic Ocean sediments. Instead, most Arctic  
815 sediments have  $[^{231}\text{Pa}_{\text{xs}}/^{230}\text{Th}_{\text{xs}}]$  activity ratios significantly lower than the production ratio  
816 (Edmonds et al., 2004; Moran et al., 2005). Recently, a few sediment samples collected on the  
817 Siberian shelf margins were reported to have  $^{231}\text{Pa}_{\text{xs}}/^{230}\text{Th}_{\text{xs}}$  ratios that slightly exceed the  
818 production ratio (Luo and Lippold, 2015). Moreover, Hoffmann et al. (2013) observed  
819 decreasing Arctic sedimentary  $^{231}\text{Pa}/^{230}\text{Th}$  ratios with water depth, over the last 30 kyr. This  
820 general deficit in  $^{231}\text{Pa}_{\text{xs}}$  was attributed to the export of a significant fraction of the Arctic  $^{231}\text{Pa}_{\text{xs}}$   
821 into the Atlantic Ocean through the Fram Strait and to the hypothetical possibility that some of  
822 the  $^{231}\text{Pa}_{\text{xs}}$  was lost by boundary scavenging in a location not identified at that time.

823 The data obtained in this work on the suspended particles of the Nansen margin (St.  
824 32, 158 and 161) show particulate  $[^{231}\text{Pa}_{\text{xs}}/^{230}\text{Th}_{\text{xs}}]$  activity ratios below the production ratio  
825 except in the surface waters, but the calculation of the particulate  $^{231}\text{Pa}_{\text{xs}}$  is affected by large  
826 uncertainties (Tab. ES2). However, Top-core sediments from the Nansen margin (St. 161 and  
827 32) have distinctly high  $[^{231}\text{Pa}_{\text{xs}}/^{230}\text{Th}_{\text{xs}}]$  activity ratios:  $[^{231}\text{Pa}_{\text{xs}}/^{230}\text{Th}_{\text{xs}}] = 0.40$  at station 161 and  
828  $0.18$  at station 32 (Tab S3). Hence, the  $[^{231}\text{Pa}_{\text{xs}}/^{230}\text{Th}_{\text{xs}}]$  activity ratios of these two surface  
829 sediment samples are much higher than the corresponding ratio in the suspended particles.  
830 These ratios at station 161 and 32 are well above the values reported by Moran et al. (2005).  
831 So boundary scavenging along the Nansen margin could contribute to the  $^{231}\text{Pa}$  depletion in  
832 the Arctic Ocean. Scavenging by the hydrothermal plume turns out to be an alternative sink.

833 Moreover, the discrepancies between the sediment and particulate  $^{231}\text{Pa}/^{230}\text{Th}$  ratios  
834 might be related to the different timescales of these data records. Still, particulate samples in  
835 the deep part of the station 50 have  $^{231}\text{Pa}_{\text{xs}}/^{230}\text{Th}_{\text{xs}}$  ratios below the production ratio despite the  
836 relatively low  $F_{\text{Th}/\text{Pa}}$  (Tab. ES2). Whether these particles are still dominated by hydrothermal  
837 particles or not remains ambiguous (Valk et al., 2018).

## 838 **5 Conclusions**

839 The distribution of  $^{231}\text{Pa}_{\text{xs}}$  and  $^{230}\text{Th}_{\text{xs}}$  in the Arctic Ocean deviates from the linear increase  
840 expected from reversible scavenging in absence of lateral advection. While the role of  
841 hydrothermal scavenging of  $^{230}\text{Th}$  recently has been highlighted in the Nansen Basin (Valk et  
842 al., 2018), we show here that boundary scavenging also removes significant amounts of  $^{230}\text{Th}$   
843 at the Nansen margin.

844 Scavenging on particles derived from hydrothermal activity is associated with a  
845 relatively low  $F_{\text{Th/Pa}}$  fractionation factor ( $\approx 10$ ), while higher  $F_{\text{Th/Pa}}$  ( $\approx 20$ ) were observed for deep  
846 and bottom waters both of the Eurasian and Makarov Basin.

847 The modified boundary scavenging model developed here for the Nansen Basin,  
848 successfully modeled the increasing  $^{230}\text{Th}_{\text{xs}}$  particulate fraction with depth and the decrease of  
849 dissolved Pa-Th contents approaching the seafloor observed on various margins. Modeled  
850 dissolved  $^{231}\text{Pa}$  distributions were largely overestimated. These results suggest that advection  
851 of dissolved  $^{231}\text{Pa}$  out the Atlantic is an important sink for the Arctic  $^{231}\text{Pa}$  budget. However,  
852 the high sedimentary  $^{231}\text{Pa}_{\text{xs}}/^{230}\text{Th}_{\text{xs}}$  ratios observed at the Barents Sea shelf and Nansen  
853 margin indicate that the Arctic margins could indeed act as a major sink for the missing Arctic  
854  $^{231}\text{Pa}$ . More data focused on shelves and slopes of the Arctic Ocean are required to better  
855 constrain this effect and the chemical nature of the particles and their physical mode of  
856 resuspension have to be addressed more precisely.

## 857 **6 Acknowledgements**

858 This work was conducted in the framework of the GEOTRACES program and was supported  
859 by the Swedish Research Council (VR 349-202-6287). The authors are pleased to thank the  
860 captains and the crew of the research vessel R/V Polarstern. Ronja Paffrath is thanked for the  
861 sampling of seawater. Karin Wallner is acknowledged for their critical roles in the laboratory  
862 work. Aridane G. González is acknowledged for melting ice rafted sediments samples. Lise  
863 Missiaen was supported by an ERC funding to the ACCLIMATE project. The constructive  
864 comments of 2 anonymous reviewers greatly improved the present manuscript.

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### 1033 **Figures and tables**

1034

1035 **Figure 1:** Samples for the analysis of dissolved and particulate  $^{231}\text{Pa}$ ,  $^{230}\text{Th}$  and  $^{232}\text{Th}$  were  
1036 collected at 9 stations along the GEOTRACES GN04 section in the Arctic Ocean. Crossed  
1037 symbols denote sampling for particulate and dissolved samples and non-crossed points are  
1038 stations which were sampled for the analysis of dissolved concentrations.

1039

1040 **Figure 2:** Dissolved concentrations of  $^{231}\text{Pa}_{\text{xs}}$ ,  $^{230}\text{Th}_{\text{xs}}$  and  $^{232}\text{Th}$  for shelf stations (upper  
1041 panel) and deep stations (lower panel). Diamonds: Nansen Basin, squares: Amundsen  
1042 basin, triangles: Makarov Basin and circles: shelf stations.

1043

1044 **Figure 3:** Particulate concentrations of  $^{231}\text{Pa}_{\text{xs}}$ ,  $^{230}\text{Th}_{\text{xs}}$  and  $^{232}\text{Th}$  for shelf stations (upper  
1045 panel) and deep stations (lower panel). Diamonds: Nansen Basin, triangles: Makarov Basin  
1046 and circles: shelf stations

1047

1048 **Figure 4:** Pa-Th Fractionation factors. Diamonds: Nansen Basin, triangles: Makarov Basin  
1049 and circles: shelf stations.

1050 **Figure 5:** Depth profiles of (a) the particulate/total ratios of  $^{231}\text{Pa}_{\text{xs}}$  and (b) particulate/total  
1051 ratios for  $^{230}\text{Th}_{\text{xs}}$ . Diamonds: Nansen Basin, triangles: Makarov Basin and circles: shelf  
1052 stations.

1053 **Figure 6:** Schematic representation of the boundary scavenging profile model: the margin and  
1054 open ocean boxes exchange a total flux of water (F). Vertical mixing is neglected. Particles are  
1055 introduced in the surface waters of the ocean margin and interior and at all depths in the margin  
1056 box (bent arrows). Particles are then transported by currents between the margin and ocean  
1057 interior.

1058

1059 **Figure 7:** Boundary scavenging model outputs. Modelled profiles of (a and d): particle  
1060 abundance and the  $^{230}\text{Th}$  particulate fraction, (b and e): dissolved and particulate  $^{230}\text{Th}_{\text{xs}}$  and  
1061 c and f): dissolved and particulate  $^{231}\text{Pa}_{\text{xs}}$  in comparison with measured data obtained at  
1062 station 32 (margin) and 50 (interior). Pink open diamonds (st 32) and orange diamonds (st  
1063 50) represent measured data while the pink lines represent the margin model and orange  
1064 lines represent the interior ocean model.

1065

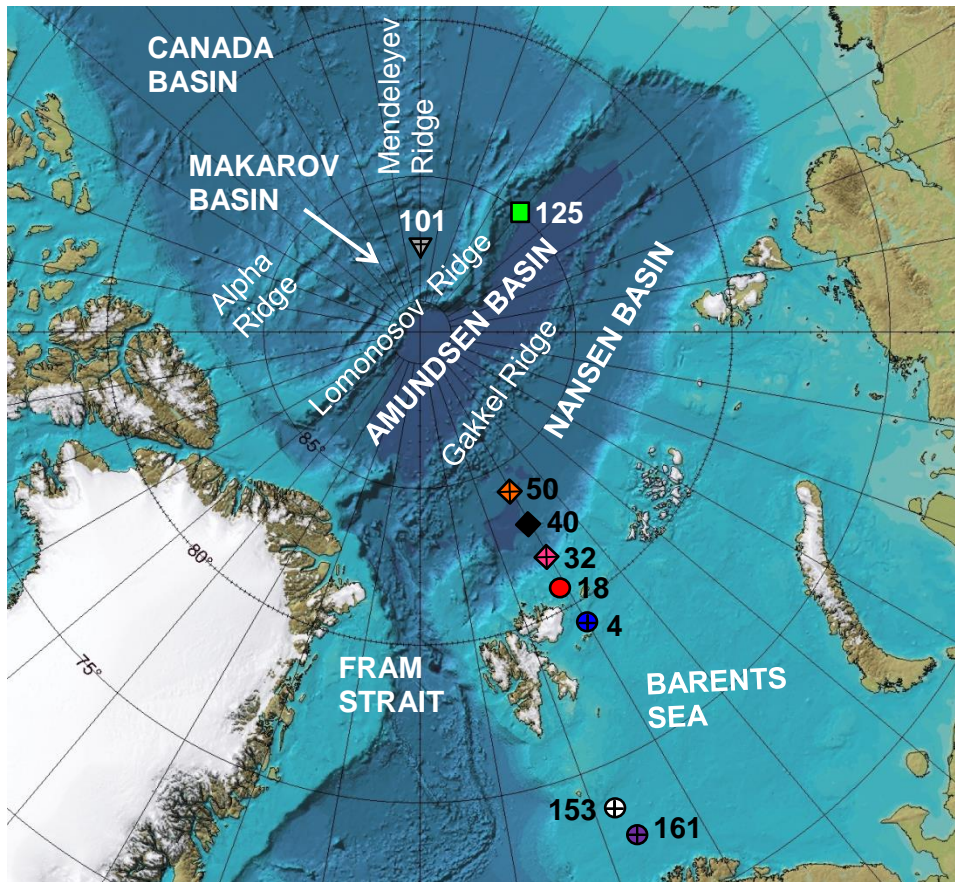
1066 **Figure 8:** Dissolved  $^{231}\text{Pa}_{\text{xs}}$  versus DSi measured during PS94 (Van Ooijen et al., 2016).  
1067 Diamonds: Nansen Basin, squares: Amundsen basin, triangles: Makarov Basin and circles:  
1068 shelf stations.

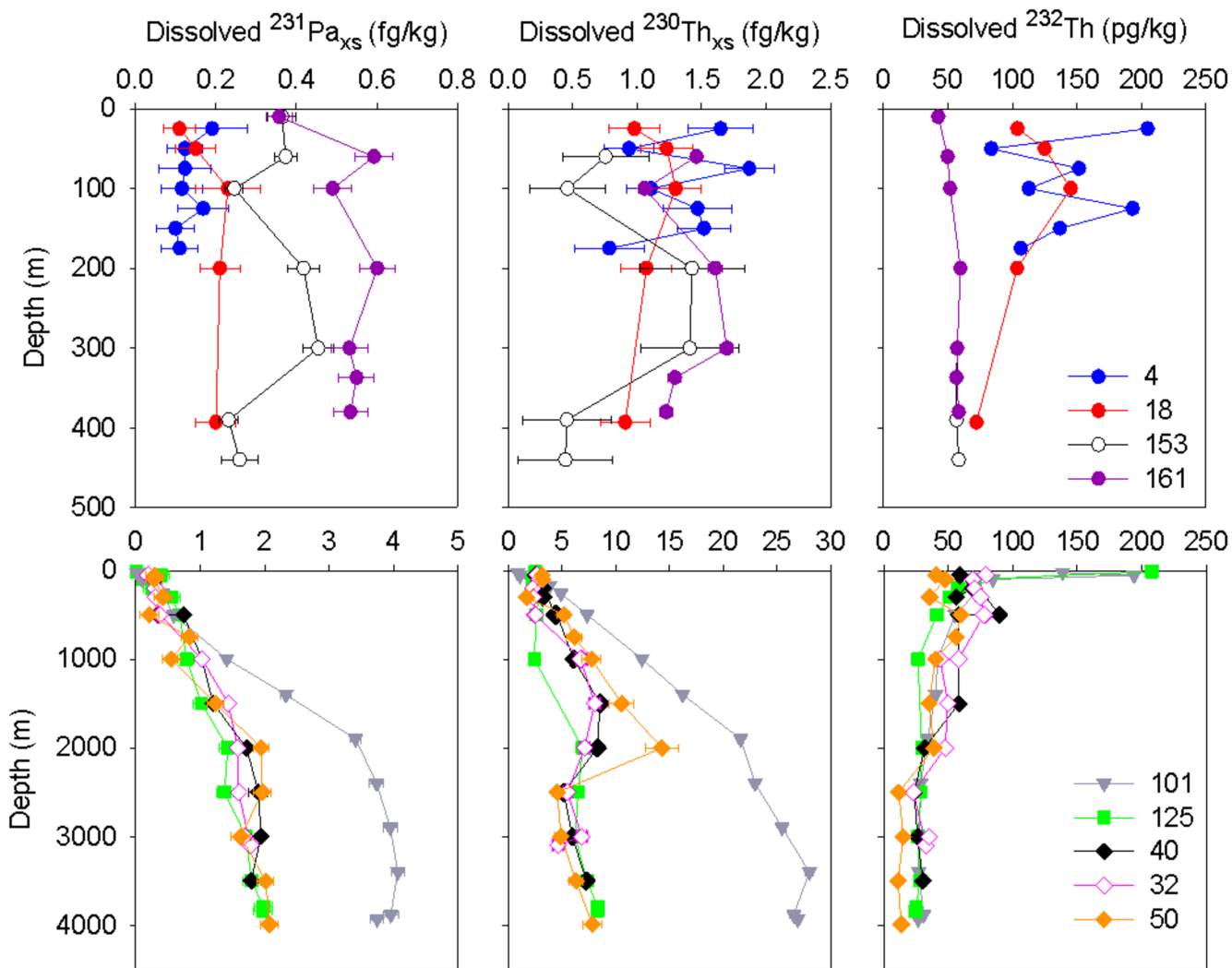
1069

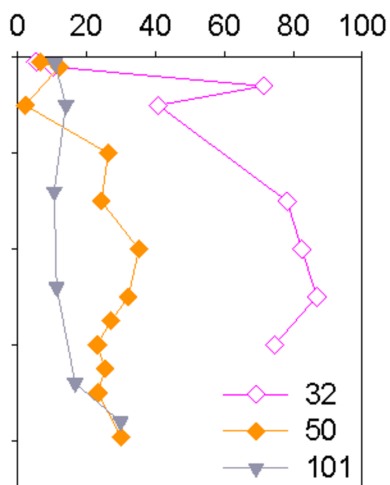
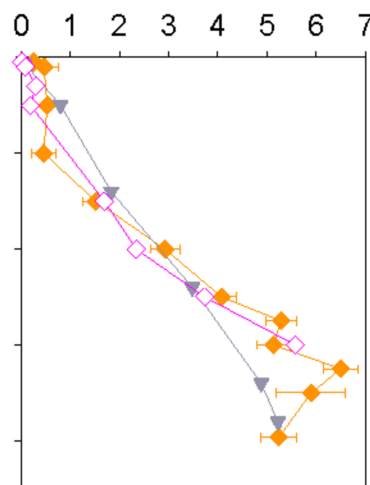
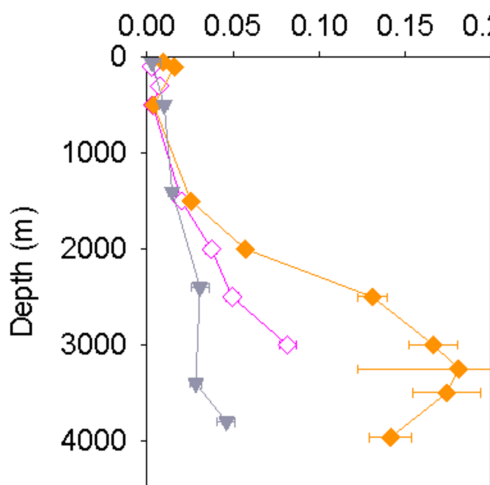
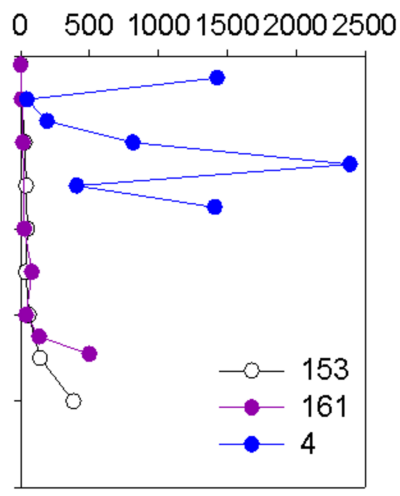
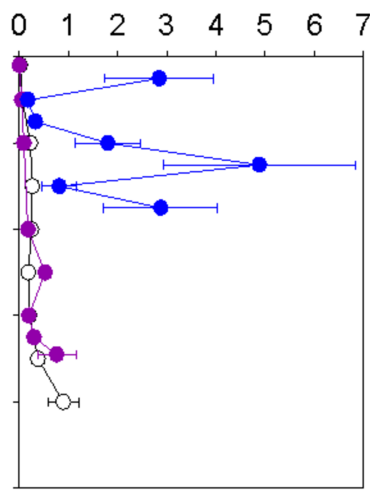
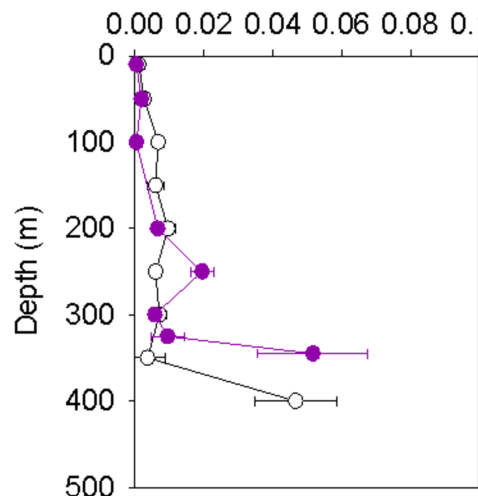
1070 **Table 1:** Parameters of the Boundary scavenging profile model

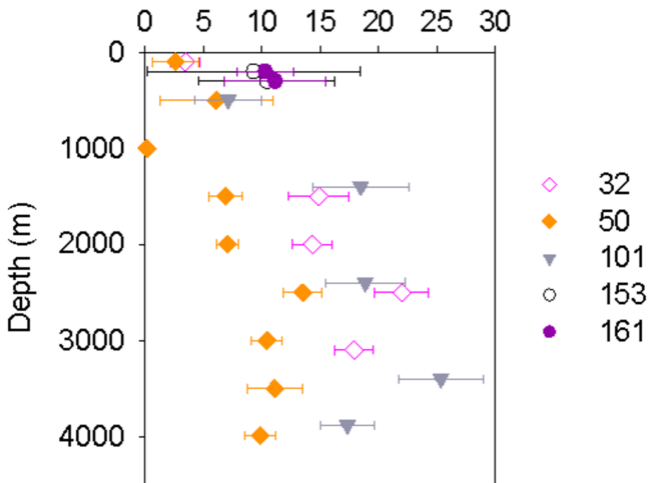
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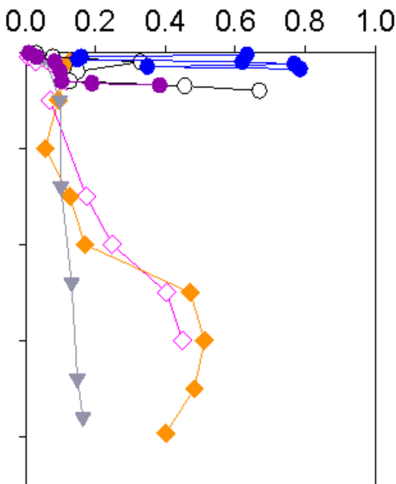
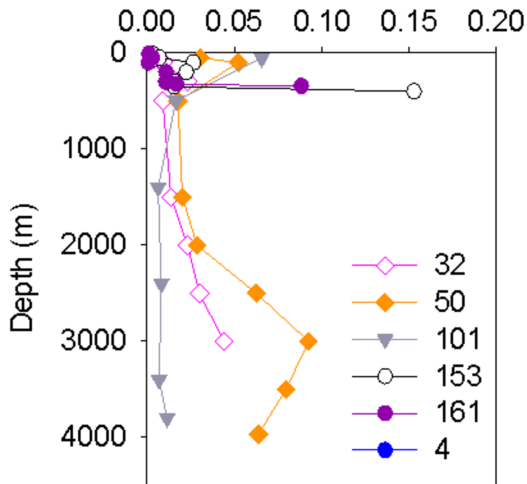


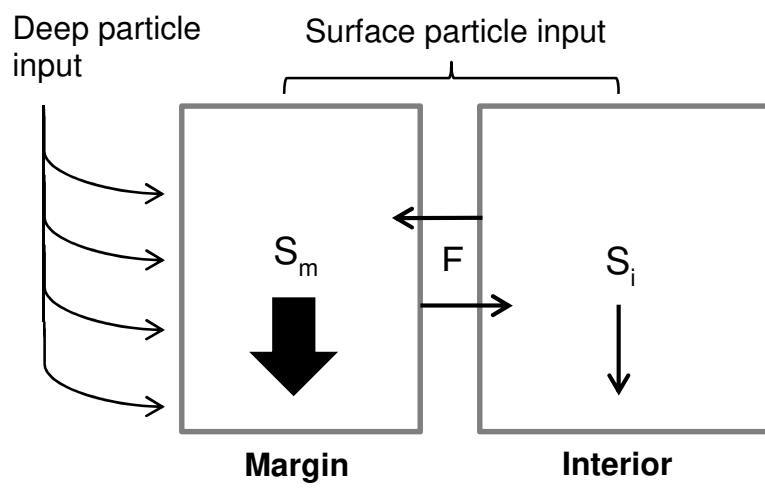


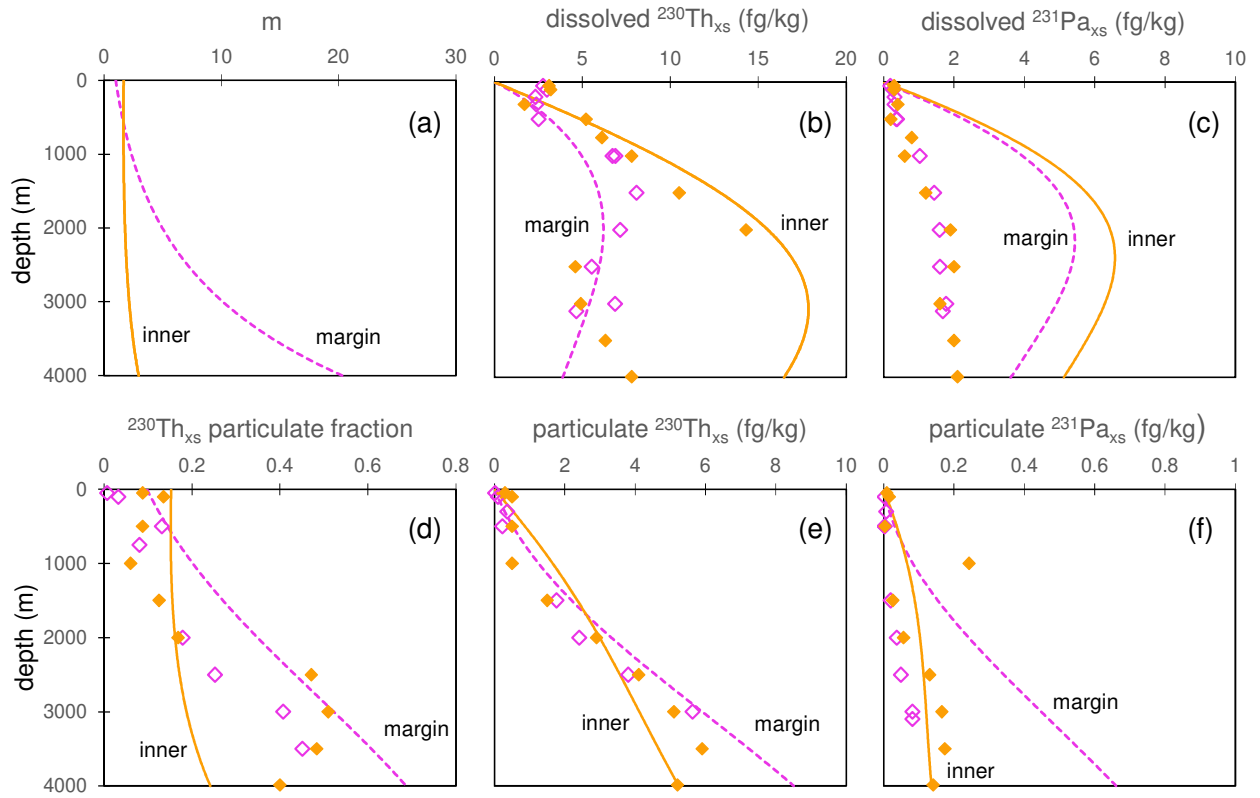
Particulate  $^{231}\text{Pa}_{\text{xs}}$  (fg/kg)Particulate  $^{230}\text{Th}_{\text{xs}}$  (fg/kg)Particulate  $^{232}\text{Th}$  (pg/kg)

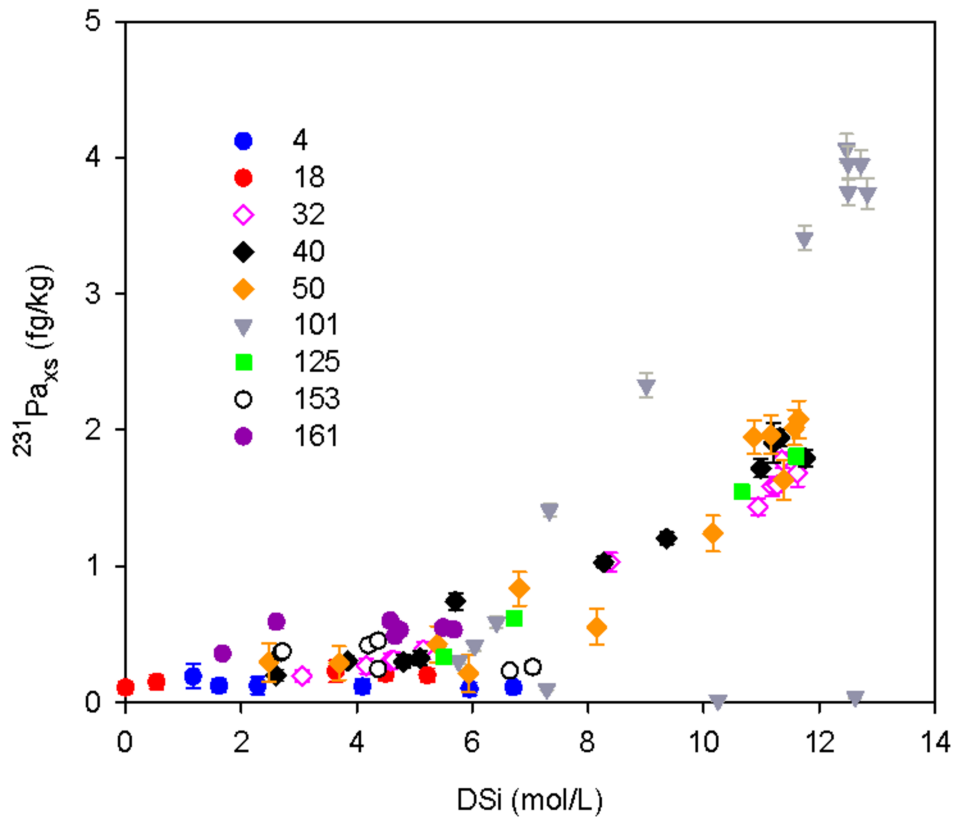
$F_{Th/Pa}$ 



Particulate fraction of  $^{231}\text{Pa}_{\text{xs}}$ Particulate fraction of  $^{230}\text{Th}_{\text{xs}}$ 







**Table 1.** Parameters of the Boundary scavenging profile model

| <b>Parameter</b>                           | <b>Ocean interior</b>  | <b>Ocean margin</b>    |
|--|------------------------|------------------------|
| V (m <sup>3</sup> )                        | 6.5 x 10 <sup>15</sup> | 1.5 x 10 <sup>15</sup> |
| F (m <sup>3</sup> /y)                      | 1.3 x 10 <sup>14</sup> | 1.3 x 10 <sup>14</sup> |
| τ (y)                                      | 50                     | 10                     |
| k  | 0.02                   | 0.1                    |
| S (m/y)                                    | 300                    | 600                    |
| K <sup>230</sup> Th                        | 0.11                   | 0.11                   |
| K <sup>231</sup> Pa                        | 0.0075                 | 0.0075                 |
| P <sup>230</sup> Th (fg/m <sup>3</sup> /y) | 0.056                  | 0.056                  |
| P <sup>231</sup> Pa (fg/m <sup>3</sup> /y) | 0.025                  | 0.025                  |
| μ (y <sup>-1</sup> )                       |                        | 0.5                    |
| m (0)                                      | 1.5                    | 1                      |