

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

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35 **Abstract**

36 ^{231}Pa , ^{230}Th and ^{232}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}Pa
39 and ^{230}Th in the Arctic Ocean deviates from the linear increase expected from reversible
40 scavenging. Higher ^{232}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}Th distributions in the Nansen Basin. Modeled dissolved ^{231}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}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}Pa and ^{230}Th are produced at a
58 constant rate by the decay of homogeneously distributed ^{235}U and ^{234}U , respectively. Both
59 ^{231}Pa and ^{230}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}Pa and ^{230}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}Pa
70 and ^{230}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}Pa and ^{230}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}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}Pa , ^{230}Th , and ^{232}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}Pa and
92 ^{230}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[®] bottles mounted on a General Oceanic[®]
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}Pa , ^{230}Th and ^{232}Th ,
109 5 L of water were filtered directly from the Niskin bottles into sampling containers using
110 Acropak500[™] cartridges (0.45 μm 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 μm pore size) were
115 collected on Supor[®] 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}U , ^{234}U , ^{231}Pa , ^{232}Th and
121 ^{230}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}Pa , ^{230}Th and ^{232}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}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 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}Pa , ^{229}Th and ^{236}U , followed by total dissolution in a 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, HNO_3 and HCl followed by spiking, chemical
141 separation and analysis of ^{231}Pa , ^{230}Th and ^{232}Th at Laboratoire des Sciences du Climat et de
142 l'Environnement (LSCE).

143 ^{231}Pa , ^{230}Th and ^{232}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}Th
157 (average = 15 ± 6 pg), 0.07 and 8.42 fg for ^{230}Th (average = 1.02 ± 1.80 fg) and 0.05 and 0.26

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

160 For particles, procedural blanks were prepared in the same way as for samples by
161 using acid cleaned filters (0.5N 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}Th (average = 4.3 ± 1.2 pg), 0.02 and 0.36 fg for ^{230}Th (average = 0.15 ± 0.03 fg) and
164 0.004 and 0.7 fg for ^{231}Pa (average = 0.27 ± 0.34 fg). These blanks were equivalent to 0.1 –
165 16 % of the measured ^{232}Th , 0.1 – 19 % of the measured ^{230}Th and 0.1 – 89 % of the measured
166 ^{231}Pa

167 All measured ^{231}Pa and ^{230}Th concentrations were corrected for the in-growth of ^{231}Pa
168 and ^{230}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}Pa , ^{230}Th and ^{232}Th concentrations were measured (Tab S1). It was
173 initiated because the first analysis of ^{230}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}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}Pa and
178 ^{230}Th measurements. The ^{232}Th replicates can vary by as much as 30 % indicating small ^{232}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}Pa , ^{230}Th
184 and ^{232}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}Pa , ^{230}Th and ^{232}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°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 (~ 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 ~ 100
221 and ~ 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°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 ~ 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}Th concentrations

237 Concentrations of dissolved ^{232}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}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}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}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}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}Pa and ^{230}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}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}Pa and ^{230}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}Pa , ^{230}Th (fg/kg) and
 262 ^{232}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}U (through ^{234}Th , its parent isotope), ^{230}Th and ^{231}Pa (through ^{231}Th , its
271 parent isotope) by α -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}Pa and ^{230}Th at stations 50 and 125
276 measured by Valk et al., (2018).

277 λ_{230} , λ_{231} , λ_{235} and λ_{238} are the decay constants for ^{230}Th ($\lambda_{230} = 9.16 \times 10^{-6} \text{ y}^{-1}$), ^{231}Pa
278 ($\lambda_{231} = 2.11 \times 10^{-5} \text{ y}^{-1}$), ^{235}U ($\lambda_{235} = 9.85 \times 10^{-10} \text{ y}^{-1}$) and ^{238}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}Th (230.033 g/mol) ^{231}Pa (231.036 g/mol) and ^{232}Th (232.038 g/mol). The concentrations of
281 ^{231}Pa and ^{230}Th can be converted to radioactivity units ($\mu\text{Bq/kg}$) by using the conversion factors
282 0.5724 and 1.3110, respectively. ^{232}Th concentrations can be converted to pmol/kg using the
283 atomic mass of ^{232}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}Pa , between 2.3 and 72 % for particulate ^{231}Pa ,
287 between 0.3 and 34 % for dissolved ^{230}Th and between 1.0 and 58 % for particulate ^{230}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}Pa was similar compared to the stations located in the Nansen Basin, while
308 concentrations of dissolved ^{230}Th in the Nansen Basin (St. 32, 40, 50) were similar or higher
309 compared to the distribution of dissolved ^{230}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 ~ 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 ~ 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}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}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 1st 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}U and ^{234}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:

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

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}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}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}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}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}Th close to the seafloor were
426 accompanied with elevated concentrations of particulate $^{230}\text{Th}_{\text{xs}}$ and ^{232}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}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}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}Th (up to ~2400 pg/kg) and elevated
453 concentrations of dissolved ^{232}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}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}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}Th . In general,
508 dissolved ^{232}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}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}Th concentrations towards the seafloor
512 suggests enhanced bottom scavenging. The lowest dissolved ^{232}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}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}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}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}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 (m^3)

574 (Jakobsson, 2002). These 2 boxes exchange a total flux of water F (m^3/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). μ is an arbitrary parameter aimed to produce
 602 an exponential increase of the particle concentration in the deep waters. μ 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}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}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}Th and $^{231}\text{Pa}_{\text{xs}}$ are produced by *in-situ* decay of U at a constant rate P . The
 615 produced ^{230}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}Th (75 000 y) and ^{231}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}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}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}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}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}Th particulate fraction with depth and for the non-linear dissolved ^{230}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}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}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}Pa (compared to ^{230}Th).

716 Extremely low dissolved $^{231}\text{Pa}_{\text{xs}}$ concentrations (0.02 ± 0.005 fg/kg) were observed in
717 surface waters (10 – 100 m) of station 101 (Fig. 2). As the high ^{232}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}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}Pa
736 for biogenic silica (BSi) (e.g. Chase et al., 2002). We propose that ^{231}Pa is scavenged by BSi
737 formed in surface waters. As these particles settle through the water column, BSi dissolves
738 and releases ^{231}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}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}Th is now applied to ^{231}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}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}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}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}Pa gradient between the inflow and outflow supports a net export
766 of ^{231}Pa into the Atlantic.

767 Finally, boundary scavenging along the slopes of the Eurasian Basin can reduce the
768 dissolved ^{231}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}Pa in the Arctic Ocean.

772 Roy-Barman (2009) proposed a balanced Arctic budget of ^{231}Pa between the inner
773 ocean and the margin without export of ^{231}Pa through the Fram Strait. However, it was based
774 on very low $F_{\text{Th}/\text{Pa}}$ values (~ 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 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}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}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}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}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 (≈ 10), while higher $F_{\text{Th/Pa}}$ (≈ 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}Pa distributions were largely overestimated. These results suggest that advection
851 of dissolved ^{231}Pa out the Atlantic is an important sink for the Arctic ^{231}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}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.

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1033 **Figures and tables**

1034

1035 **Figure 1:** Samples for the analysis of dissolved and particulate ^{231}Pa , ^{230}Th and ^{232}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}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}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}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.

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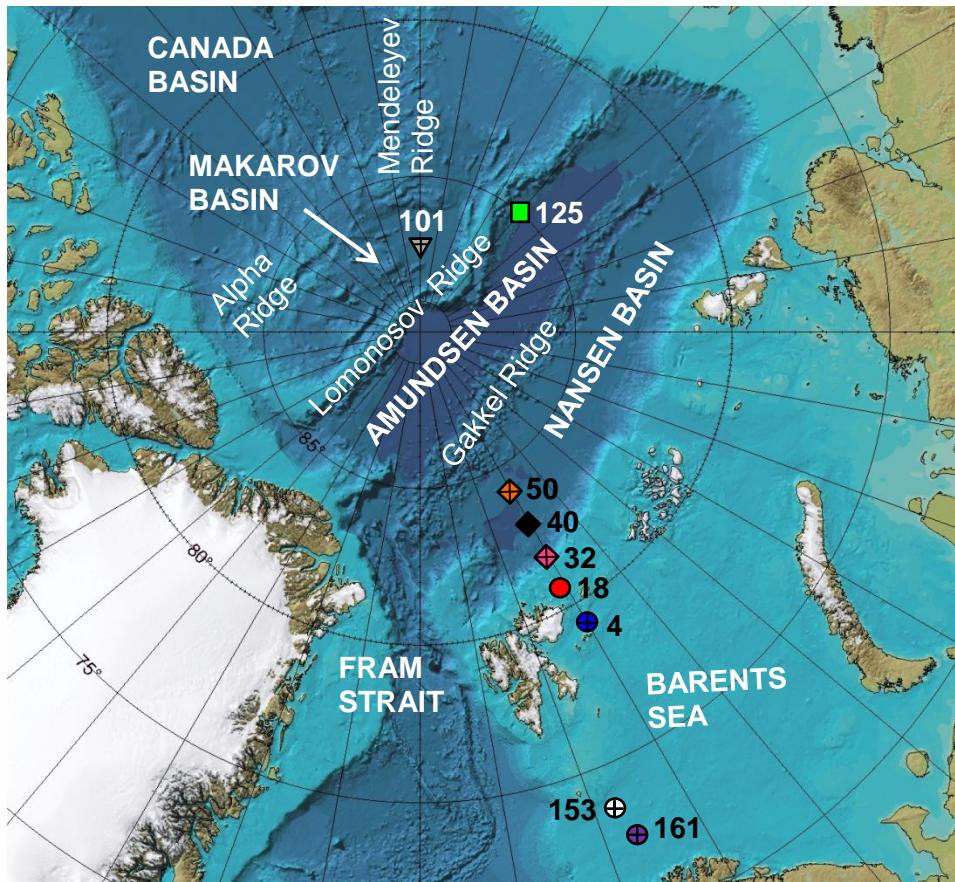
1066 **Figure 8:** Dissolved $^{231}\text{Pa}_{\text{xs}}$ versus DSi measured during PS94 (Van Ooijen et al., 2016).
1067 Diamonds: Nansen Basin, squares: Amundsen basin, triangles: Makarov Basin and circles:
1068 shelf stations.

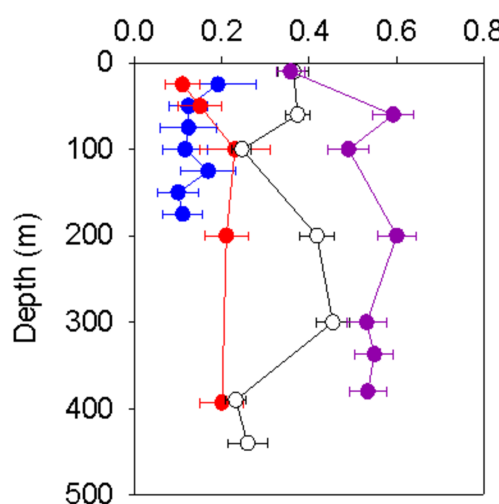
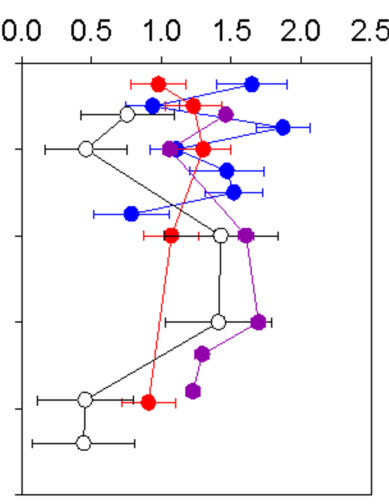
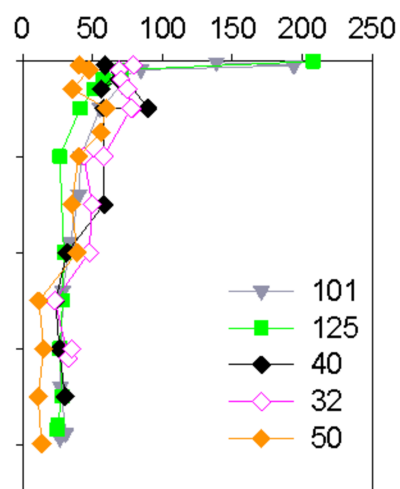
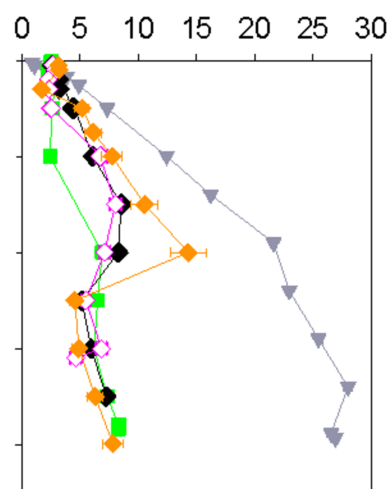
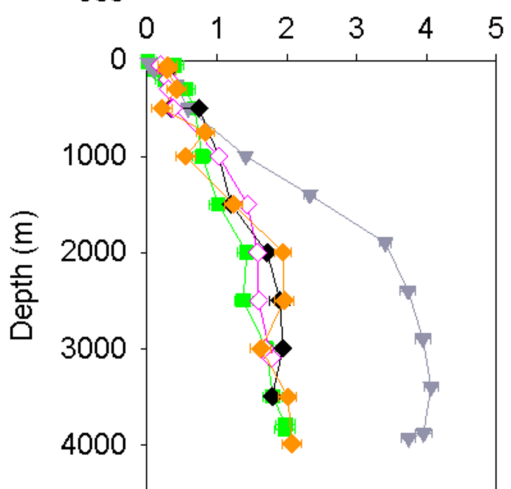
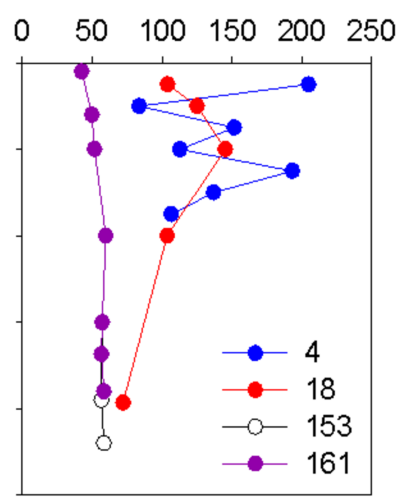
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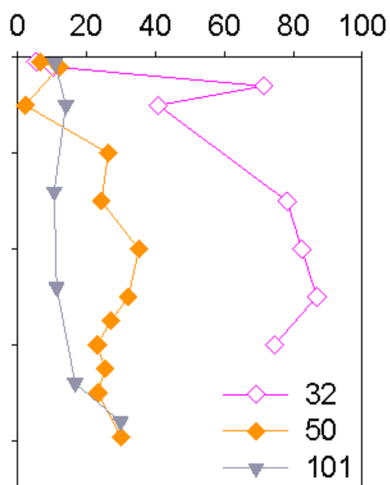
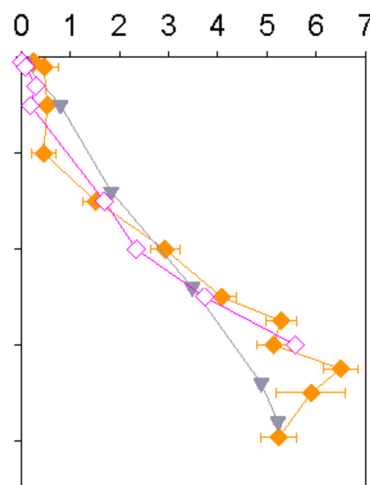
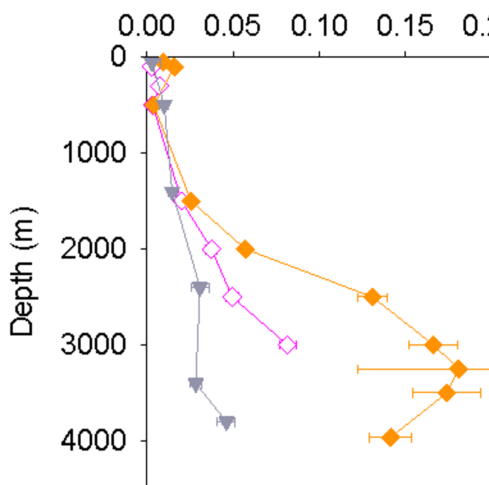
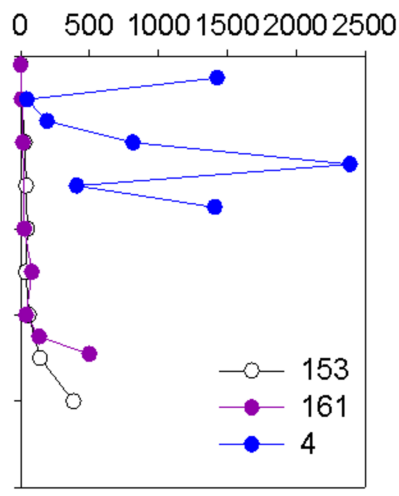
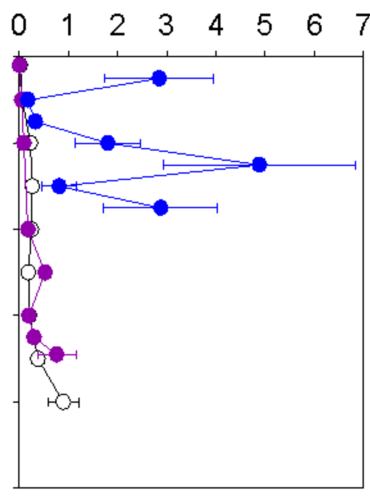
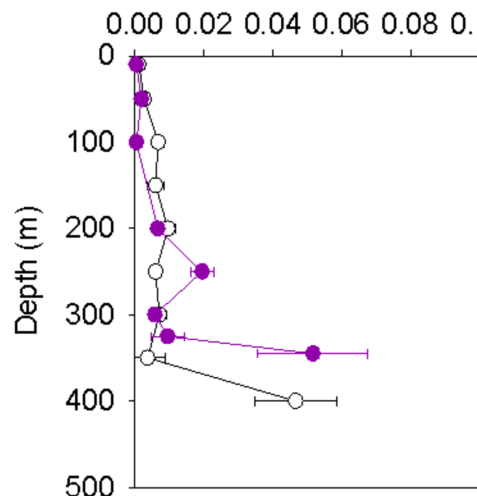
1070 **Table 1:** Parameters of the Boundary scavenging profile model

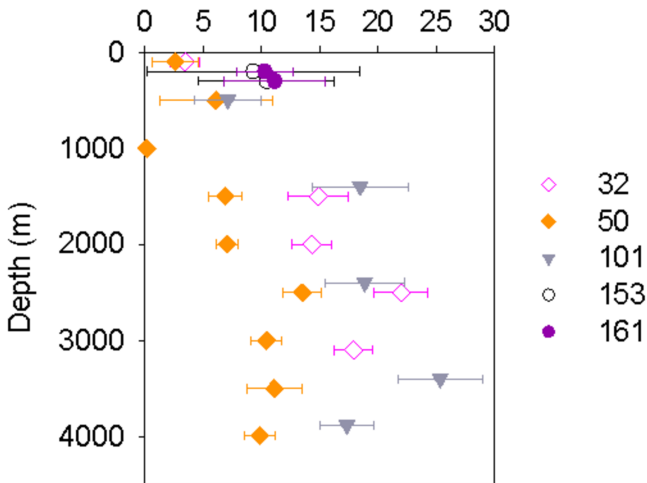
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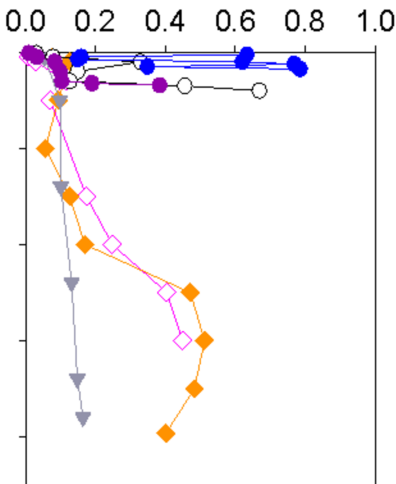
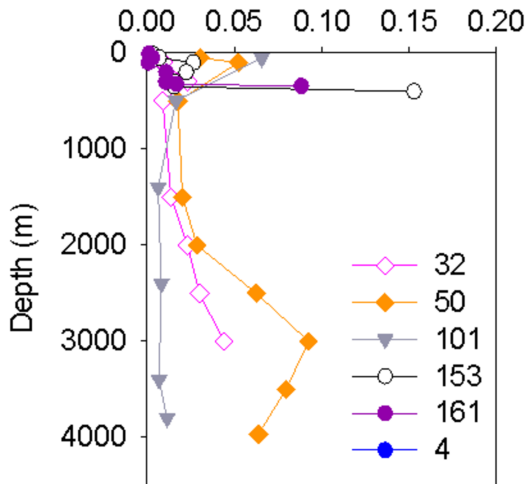
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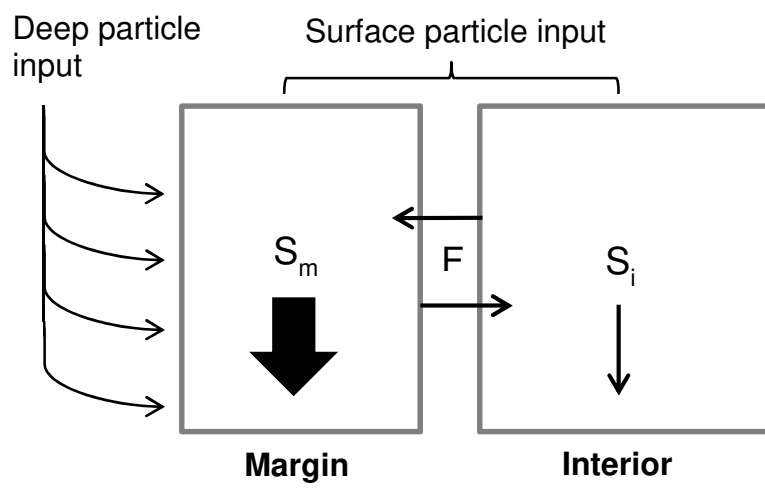


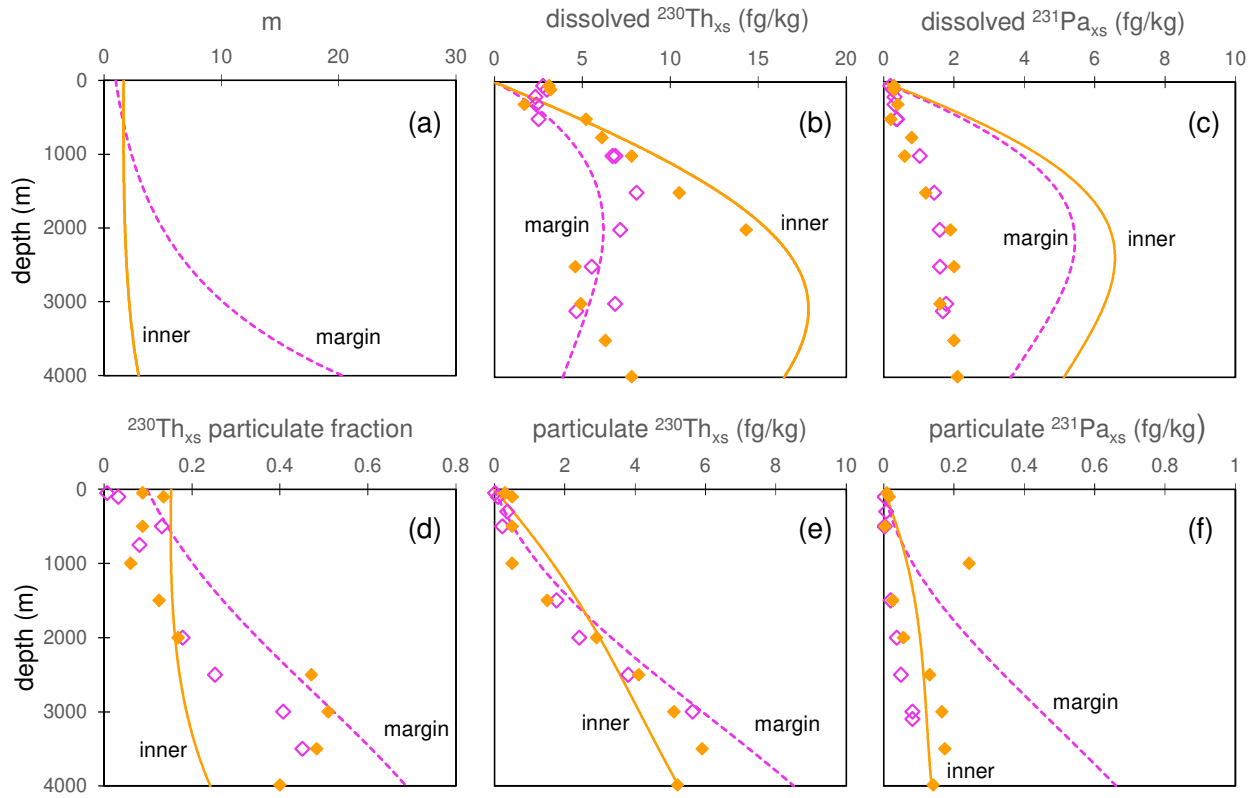
Dissolved $^{231}\text{Pa}_{\text{xs}}$ (fg/kg)Dissolved $^{230}\text{Th}_{\text{xs}}$ (fg/kg)Dissolved ^{232}Th (pg/kg)

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

$F_{Th/Pa}$ 

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





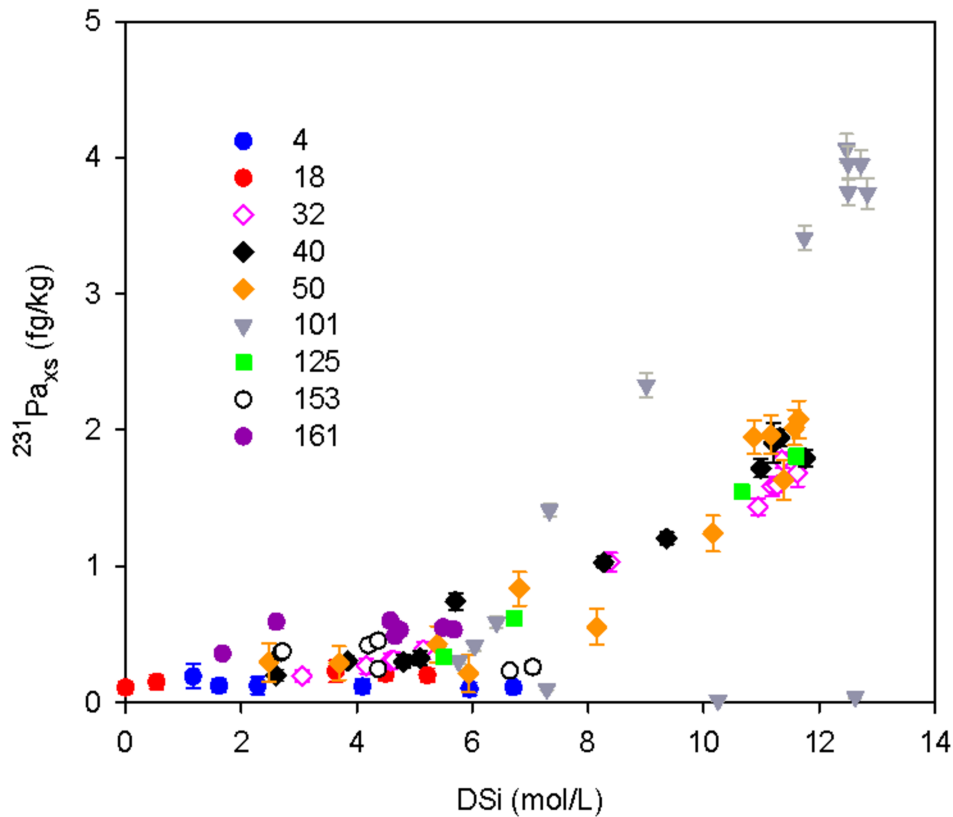


Table 1. Parameters of the Boundary scavenging profile model

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