

1 **Abstract**

2 The Southern Ocean is known as the largest High-Nutrient, Low-Chlorophyll (HNLC) region
3 of the global ocean due to iron limitation. However, a large phytoplankton bloom develops
4 annually downstream of the Kerguelen Islands, a bloom which is sustained partly by iron
5 released from the sediments deposited onto the margins. In the framework of the KEOPS-2
6 project, we used radium isotopes (^{224}Ra , $T_{1/2} = 3.66$ d; ^{223}Ra , $T_{1/2} = 11.4$ d; ^{228}Ra , $T_{1/2} = 5.75$ y)
7 to provide information on the origin of iron fertilization and on the timescales of the transfer
8 of sediment-derived inputs (including iron and other micronutrients) towards offshore waters.
9 Significant ^{224}Ra and ^{223}Ra activities were found in the near vicinity of the Kerguelen Islands,
10 in agreement with the short half-lives of these isotopes. Significant ^{224}Ra and ^{223}Ra activities
11 were also detected up to 200 km downstream of the islands and more unexpectedly in
12 offshore waters south of the Polar Front. These observations thus clearly indicate i) that the
13 sediment-derived inputs are rapidly transferred towards offshore waters (on timescales in the
14 order of several days up to several weeks) and ii) that the Polar Front is not a physical barrier
15 for the chemical elements released from the sediments of the Kerguelen Plateau. The Ra
16 dataset suggests that iron and other micronutrients released by the shallow sediments of the
17 Kerguelen margins may contribute to fuel the phytoplankton bloom downstream of the
18 islands, despite the presence of the Polar Front. However, the heterogeneous distribution of
19 the ^{224}Ra and ^{223}Ra activities in surface waters suggests that this supply across the front is not
20 a continuous process, but rather a process that is highly variable in space and time.

21

22 **Key words:** Southern Ocean, Iron fertilization, Radium isotopes, GEOTRACES

23

24 1 Introduction

25 The Southern Ocean is recognized as the major High-Nutrient, Low-Chlorophyll (HNLC)
26 region of the global ocean. Despite high nutrient concentrations, the phytoplankton growth
27 was shown to be limited by the very low iron concentrations in surface waters of the Southern
28 Ocean (De Baar et al., 1995; Martin et al., 1990). Dissolved iron is, however, supplied to the
29 surface waters in several locations of the Southern Ocean where iron is released by the
30 sediment margins but this natural iron fertilization remains spatially limited (Tagliabue et al.,
31 2014). Consequently, high phytoplankton biomass can be found offshore the Antarctic
32 continental shelf (Arrigo et al., 2008; Moore and Abbott, 2002) or in the vicinity of
33 subantarctic islands (Blain et al., 2001; Korb et al., 2004; Pollard et al., 2007).

34 One of the largest phytoplankton blooms is observed offshore the Kerguelen Islands, in
35 the Indian sector of the Southern Ocean (Blain et al., 2001). This phytoplankton bloom
36 extends more than 1000 km downstream of the Kerguelen Islands and shows two main
37 features: (i) a plume that extends northeastward of the islands and north of the Polar Front
38 (PF) that shows high mesoscale and temporal variability, and (ii) a larger bloom
39 southeastward of the islands and south of the PF (Blain et al., 2001, 2007). The two areas are
40 separated by a narrow band of relatively low chlorophyll concentration associated with the PF
41 that follows the inner shelf edge between 200 and 500 m isobaths (Park and Gamberoni,
42 1997; Park et al., 1998b). While Park et al. (2008a) suggest that the northward geostrophic
43 flow associated with the PF may possibly block any southward penetration of lithogenic
44 inputs released by the Kerguelen Islands the numerous eddies and meanders formed along the
45 PF may contribute to transport chemical elements between the northern Kerguelen Plateau
46 and offshore waters.

47 The KEOPS-2 (Kerguelen ocean and plateau compared study) project aimed at
48 understanding the circulation patterns off the Kerguelen Islands and the mechanisms of iron
49 fertilization in that area. The KEOPS-2 cruise was conducted during austral spring 2011
50 eastward of the Kerguelen Islands. Natural radio-tracers such as radium isotopes (^{223}Ra , $T_{1/2} =$
51 11.4 d ; ^{224}Ra , $T_{1/2} = 3.66\text{ d}$; ^{228}Ra , $T_{1/2} = 5.75\text{ y}$) have already been proved to be powerful tools
52 to track the origin and fate of chemical elements - including iron and other micronutrients -
53 that are released by the sediments deposited on the margins (Annett et al., 2013; van Beek et
54 al., 2008; Charette et al., 2007; Dulaiova et al., 2009; Sanial et al., 2014). In this work, we
55 refer to these latter inputs as “sediment-derived inputs”. Radium isotopes are produced by the
56 decay of particle-bound thorium isotopes in the sediments and are delivered to the open ocean
57 by diffusion and advection processes. Thus, a water mass that interacts with shallow
58 sediments deposited onto the margins is potentially enriched in radium and in other elements
59 that also diffuse out of the sediments (e.g. iron, other micronutrients ...). While iron may then
60 be removed from the water column by biotic or abiotic processes, radium behaves as a
61 conservative tracer. Radium is only affected by radioactive decay and mixing in such a way
62 that the water body keeps the signature of its contact with the sediments. The radium
63 signature of a given water mass may then be transferred by diffusion and advection towards
64 offshore waters. The presence of significant Ra activities in offshore waters thus indicates that
65 the water body has interacted with shallow sediments. Alternatively, vertical mixing may also
66 transport Ra towards surface waters. Because radium isotopes decay, they can be used as
67 chronometers to estimate the time elapsed since the water body left the margin, which in turn
68 gives information on how quickly the microelements released by the shallow sediments may
69 be transferred to offshore waters (Moore, 2000). In this work, we examined the distribution of
70 ^{223}Ra , ^{224}Ra and ^{228}Ra in surface waters downstream of the Kerguelen Islands in order (i) to
71 investigate the origin and dispersion of the sediment-derived inputs, including iron and (ii) to
72 determine the apparent ages of offshore waters that provide information on the timescales of

73 the transfer of water and associated chemical elements between the margins and offshore
74 waters. In addition to the Ra distribution in surface waters, we report several vertical profiles
75 of ^{223}Ra , ^{224}Ra and ^{228}Ra that provide constraints on the vertical transport of chemical
76 elements associated with vertical mixing.

77 2 **Material and Methods**

78 2.1 The KEOPS-2 project

79 The KEOPS-2 cruise took place east of the Kerguelen Islands (northern Kerguelen
80 Plateau) between 14 October and 23 November 2011 on board the *R/V Marion Dufresne*
81 (IPEV: Institut Polaire Français – Paul Emile Victor and TAAF: Terres Australes et
82 Antarctiques Françaises). The KEOPS-2 project was designed to study the mechanisms of
83 natural iron fertilization off Kerguelen Islands and its impact on ecosystems and
84 biogeochemical cycles. The KEOPS-2 project was labelled as a GEOTRACES process study
85 and followed up a first KEOPS project conducted in 2005 in the area of the Southern
86 Kerguelen Plateau (Blain et al., 2007).

87 2.2 Study area

88 The Kerguelen Plateau, located in the Indian sector of the Southern Ocean, constitutes
89 one of the few physical barriers for the eastward-flowing Antarctic Circumpolar Current
90 (ACC). Various studies provide a detailed description of the general ocean circulation patterns
91 around the Kerguelen Plateau (Charrassin et al., 2004; Park and Gambéroni, 1995; Park et al.,
92 1998, 2008b, 2009). An important oceanographic feature of the area is the presence of the
93 Polar Front (PF), which is commonly characterized by the northernmost position of a
94 subsurface temperature minimum bounded by the 2 °C isotherm (Belkin and Gordon, 1996;
95 Park and Gamberoni, 1997; Park et al., 1993). A strong eastward current associated with this

96 front is deflected to the north at 71 °E following the eastern shelf slope of the Kerguelen
97 Plateau between the 200 and 500 m isobaths and forms a cyclonic meander that turns
98 southward at 75 °E (Belkin and Gordon, 1996; Orsi et al., 1995; Park and Gamberoni, 1997;
99 Park et al., 1993; Pollard et al., 2002). Numerous eddies are generated along the PF eastward
100 of the Kerguelen Plateau, that can, in some cases, be identified on the satellite composite
101 images of sea surface chlorophyll. The location of the stations investigated in this study is
102 shown on Fig. 1.

103 2.3 Radium data

104 2.3.1 Sample collection

105 Surface seawater samples were collected at 7 m depth using a clean pump specially
106 designed by IPEV for the KEOPS-2 cruise. Large volumes of surface seawater were collected
107 (250-900 L) and stored in large plastic tanks. We used a CTD (SBE-19plus, Seabird®) and a
108 rosette system equipped with 22x12-L Niskin bottles to collect seawater samples from various
109 depths throughout the water column. Three samples were also collected directly on two
110 beaches of the Kerguelen Islands (Baie du Morbihan: samples KER-1 and Baie des
111 Baleiniers: samples BaieB-1 and BaieB-2). Seawater samples were then passed by gravity
112 through PVC cartridges filled with “Mn-fibers” (MnO₂-impregnated acrylic fiber), following
113 (Moore, 2008). The flow rate was fixed at $\leq 0.5 \text{ L min}^{-1}$ to provide 100 % extraction
114 efficiency (Moore, 2008; van Beek et al., 2010). The Mn-fibers were then rinsed with MilliQ
115 water and partially dried before analysis.

116 2.3.2 Sample analysis

117 The Mn-fibers were analyzed using a Radium Delayed Coincidence Counter (RaDeCC;
118 (Charette et al., 2001; Moore and Arnold, 1996; Moore, 2008)). Three counting sessions are

119 necessary to determine both excess ^{224}Ra and excess ^{223}Ra activities in the samples. The first
120 counting was performed on board the research vessel during the cruise and provides the total
121 ^{224}Ra and ^{223}Ra activities. The Mn-fibers were analyzed again 3 weeks after sampling to
122 determine the ^{224}Ra activities supported by ^{228}Th and then after 3 months to determine the
123 ^{223}Ra activities supported by ^{227}Ac (Moore, 2000). The ^{224}Ra activities are corrected for the
124 ^{224}Ra supported by ^{228}Th and the ^{223}Ra activities are corrected for the ^{223}Ra supported by
125 ^{227}Ac . The ^{224}Ra and ^{223}Ra activities discussed hereafter thus refer to these excess ^{224}Ra and
126 ^{223}Ra activities. Uncertainties for both isotopes were calculated following Garcia-Solsona et
127 al. (2008) and were reported with one-sigma confidence interval.

128 Activities of ^{228}Ra were then determined using the low-background gamma detectors
129 placed at the LAFARA underground laboratory in the French Pyrénées (van Beek et al., 2010,
130 2013). Mn-fibers were either ashed at 820 °C for 16 h (Charette et al., 2001) and analyzed
131 using a well-type germanium detector or compressed and analyzed using a semi-planar
132 detector. Cross-calibrations between the two detectors were made to avoid any bias in the
133 determination of the Ra activities. Each sample was analyzed for ca. 120 hours to allow the
134 quantification of the low ^{228}Ra activities present in Southern Ocean waters (Kaufman et al.,
135 1973). ^{228}Ra activities were determined using ^{228}Ac peaks (338, 911 and 969 keV). All radium
136 activities are reported in disintegration per minute per 100 L of seawater (dpm/ 100 L). The
137 uncertainties reported for gamma counting consist in the error associated with counting
138 statistics (one sigma).

139 2.4 Physical data

140 2.4.1 Color data

141 High resolution maps ($1/25^\circ \times 1/25^\circ$) of chlorophyll concentration (mg/m^3) were
142 constructed by a 10-day weighted mean of MODIS and MERIS measurements. These satellite

143 products were delivered 3 times a week in near-real time during the cruise from Ssalto/Duacs
144 and CLS (Collecte Localisation Satellites, Toulouse, France) with support from CNES
145 (Centre National d'Etudes Spatiales, France). These images were used to define the sampling
146 strategy in the investigated area.

147 2.4.2 Surface drifters

148 Drifters provided by the US National Ocean and Atmospheric Administration
149 (NOAA) Global Drifter Program (GDP) were also released. The drogue is centered at 30 m
150 depth. These drifters thus provide information on the mean currents in the surface mixed layer
151 and on the dispersion of water masses due to eddy activities. Successive positions of the
152 drifter were transmitted to the R/V Marion Dufresne four times a day by the NOAA GDP
153 center. The time-irregular positions of the drifter were interpolated into a regular time step of
154 12 minutes and a low-pass filter of 48 hours was then applied to filter all tidal currents and
155 inertial oscillations.

156 2.4.3 Lagrangian particle analysis

157 The Lagrangian particle analysis was based on total surface currents, which are the sum
158 of the absolute geostrophic currents (deduced from altimeter product) and Ekman currents
159 (daily mean). The Ekman component is deduced from the European Centre for Medium-
160 Range Weather Forecasts (ECMWF) wind stress analysis applying a regional Ekman model,
161 specifically adjusted for the Kerguelen area. The altimeter current products were produced by
162 Ssalto/Duacs and distributed by AVISO, with support from CNES. Total surface currents
163 were delivered every day with a $1/8^\circ \times 1/8^\circ$ resolution. Details of the mapping technique are
164 given by (Dibarboure et al., 2011).

165 2.4.4 Lagrangian Model

166 The altimetry-derived velocities providing the geostrophic mesoscale velocity at the
167 ocean surface were analyzed in near-real time with a Lagrangian model. This model was
168 inspired by Mongin et al. (2009) who reconstructed the extension of the Kerguelen
169 chlorophyll plume with a transport scheme based on altimetry. The model created thousands
170 of virtual surface drifters released on the shelf break of Kerguelen (2000 m isobaths; apparent
171 age = 0). The trajectories were constructed by backward-in-time integration of the altimetric
172 velocity field and were stopped when a hit over the Kerguelen shelf break was detected
173 (indicating a trajectory coming from the shelf), or when a maximum integration time—set to
174 120 days—was reached (indicating no interaction with the shelf on the past 120 days). This
175 model was applied successfully by Sanial et al. (2014) to highlight the key role played by
176 surface horizontal transport in defining the extension of the spring-time chlorophyll plume in
177 the Crozet area.

178 **3 Results**

179 3.1 Hydrological context during the KEOPS-2 cruise

180 The KEOPS-2 cruise lasted almost two months (October-November 2011). During that
181 period, the phytoplankton bloom developed off the Kerguelen Islands (Fig. 2). The satellite
182 composite images of sea surface chlorophyll reveal a complex shape of the phytoplankton
183 bloom that may be associated with the complex hydrography of the area. High concentration
184 of chlorophyll first appeared close to the Kerguelen Islands (October 2011) before spreading
185 out in offshore waters until covering a large part of the study area at the end of November
186 2011. East of the Kerguelen Islands, a narrow band of low chlorophyll concentration is
187 associated with the northward branch of the PF that splits the phytoplankton bloom into two
188 parts.

189 The PF also delimits two surface water masses characterized by a strong contrast in
190 temperature and salinity; the Antarctic Surface Water (AASW) is located south of the PF, and
191 the SubAntarctic Surface Water (SASW) is located north of the PF (Emery and Meincke,
192 1986). The potential temperature-salinity diagrams of the water masses investigated in this
193 study are shown in Fig. 3. The SASW is identified only at station F-L, suggesting that this
194 station is located north of the PF. The Winter Water (WW), a typical feature of the Antarctic
195 Zone that is characterized by a subsurface temperature minimum layer around 200 m depth
196 (Park et al., 1998a, 2008b, 2014) is found on all the vertical profiles reported here, except for
197 station F-L, thus confirming its location north of the PF. Below the WW, three water masses
198 can be identified: the Upper Circumpolar Deep Water (UCDW), the Lower Circumpolar Deep
199 Water (LCDW) and the Antarctic Bottom Water (AABW) (Park et al., 1993, 2008b). Note
200 that the AABW is only found on the F-L profile (commonly observed below 2600 m in this
201 area; (Park et al., 2008b)).

202 3.2 Radium distribution in surface waters

203 The radium activities reported in this study are shown in Table 1 and fall in the range
204 of previous radium data reported for surface waters near islands of the Southern Ocean
205 (Annett et al., 2013; Charette et al., 2007; Dulaiova et al., 2009; Hanfland, 2002; Kaufman et
206 al., 1973; Sanial et al., 2014; van Beek et al., 2008). The highest ^{223}Ra , ^{224}Ra and ^{228}Ra
207 activities are found in seawater samples collected at shallow stations near the Kerguelen
208 Islands (bathymetry < 200 m; Fig. 4). The radium activities then gradually decrease offshore.
209 Several samples, however, display significant ^{224}Ra activities in samples collected offshore
210 (Fig. 4a): stations UW-21-23-34 and TEW-7 located along the PF; stations UW-32, E-1 and
211 TEW-5 south of the PF; and station TNS-1 north of the PF. A greater number of offshore
212 stations exhibit significant ^{223}Ra activities, which agrees with the longer half-life of the ^{223}Ra
213 isotope (Fig. 4b). The stations displaying significant ^{224}Ra activities also display significant

214 ^{223}Ra activities. The radium activities are especially high at station TNS-2 located north of the
215 PF and at stations E-1 and G-1, located south of the PF. Station G-2 was visited twice and
216 showed high ^{223}Ra and ^{224}Ra activities at both visits. Station A3 located on the southern
217 Kerguelen Plateau was also visited twice. Significant ^{223}Ra and ^{224}Ra activities were
218 determined in the water sample collected at station A3-1 during the first visit at station A3
219 (note however that these activities are low) but were both below the detection limit at station
220 A3-2 (second visit at station A3). In contrast, the ^{228}Ra activities are similar at the two visits
221 of A3 (Table 1). All surface samples display significant ^{228}Ra activities up to ca. 300
222 kilometers offshore from the Kerguelen Islands (i.e. station TEW-8). Relatively high values
223 are observed at stations TNS-2 and UW-32 located north and south of the PF, respectively
224 (Fig. 4c). Station R-2, which was chosen as the reference station for typical HNLC waters east
225 of the Kerguelen Islands, shows significant ^{223}Ra , ^{224}Ra and ^{228}Ra activities in surface waters.

226 3.3 Vertical distribution of Ra isotopes

227 The study of the vertical distribution of Ra isotopes allows us to provide constraints on
228 the vertical transport of Ra associated with vertical mixing. Consequently, these profiles help
229 us to define the origin of the Ra enrichments observed in surface waters off the Kerguelen
230 Islands (lateral versus vertical supply of Ra). The major water masses, identified with the
231 potential temperature-salinity diagrams throughout the water column are reported for each
232 profile. The shallow Ra profiles (stations TEW-3, G-1 and A3-2) are shown on Fig. 5 and the
233 deep profiles (stations F-L, E-4W and E-1) are shown on Fig. 6.

234 The ^{223}Ra and ^{224}Ra activities are usually higher in samples collected near the seafloor
235 and are below the detection limit at intermediate depths (Table 2; Fig. 5 and 6). Significant
236 ^{223}Ra and ^{224}Ra activities are observed in surface and/or subsurface waters several kilometers
237 offshore from the islands, in particular at stations G-1 and E-1 located south of the PF and at

238 station F-L located north of the PF. The vertical profiles of ^{223}Ra and ^{224}Ra are quite unique at
239 station F-L. Although i) this station is located far from the Kerguelen Islands and ii) the
240 bottom is at 2670 m depth, the ^{223}Ra and ^{224}Ra activities are relatively high throughout the
241 water column (Fig. 6). Significant ^{228}Ra activities were found in the different water columns
242 investigated in this study. The ^{228}Ra activities at stations TEW-3 and G-1 are relatively high
243 and uniform throughout the water column. The ^{228}Ra activities at station A3 are uniform in
244 the upper 250 m and then increase with increasing water depth. The vertical ^{228}Ra profiles at
245 the deep stations (F-L, E-1 and E-4W) exhibit an increasing trend with increasing depth
246 reflecting the diffusion of radium out of the sediments. This latter pattern is also especially
247 marked at station A3 on the southern Kerguelen Plateau (Fig. 5).

248 **4. Discussion**

249 4.1 Origin of the radium enrichments in surface waters

250 The relatively high radium activities (^{223}Ra , ^{224}Ra and ^{228}Ra) observed in surface
251 waters east of the Kerguelen Islands may be explained either by the vertical transport or
252 diffusion that supplies radium to surface waters or by the lateral advection of waters that have
253 recently interacted with shallow sediments (Blain et al., 2001; Park et al., 2008a, van Beek et
254 al., 2008).

255 When considering solely the ^{228}Ra vertical profiles - that show in most cases an increase
256 of ^{228}Ra activities with increasing depth - it cannot be excluded that the vertical mixing
257 contributes to increase radium activities in surface waters. However, the ^{224}Ra and ^{223}Ra
258 vertical profiles – that show higher Ra activities in the upper and in the deep water column but
259 Ra activities below the detection limit in the mid water column - clearly indicate that the
260 higher ^{224}Ra and ^{223}Ra activities in surface waters cannot be explained by vertical mixing. The

261 ^{224}Ra and ^{223}Ra enrichments in surface waters are thus more likely explained by the lateral
262 advection of waters that have recently interacted with shallow sediments.

263 The northward advection of a water mass that has interacted with the shallow
264 sediments deposited on the shelves of Heard Island has been identified as a pathway for the
265 micronutrients that sustain the phytoplankton bloom on the southern Kerguelen Plateau
266 (Chever et al., 2010; van Beek et al., 2008; Zhang et al., 2008). The presence of a chlorophyll
267 plume that expands northward of the southern Kerguelen Plateau may also support the
268 existence of this northward advection (Fig. 2). The observation of significant ^{224}Ra and ^{223}Ra
269 activities in surface waters at station A3-1 confirms this circulation pattern and suggests that
270 the transit time of the waters that interacted with the shallow margins of Heard Island may be
271 <1 month between Heard Islands and station A-3. This is in agreement with the Ra data
272 obtained in 2005 during the KEOPS-1 project, where significant ^{224}Ra and ^{223}Ra activities
273 were also found in surface waters at station A3 (van Beek et al., unpublished data). When the
274 waters move further north towards the area investigated in this study, the ^{224}Ra and ^{223}Ra
275 activities will then continue to decay. Two drifters released during the KEOPS-2 cruise at
276 station A3 allow us to provide constraints on the transit time between the southern Kerguelen
277 Plateau and the studied area (eastward of Kerguelen at around 49 °S). A first drifter
278 recirculated around station A3 nearly 20 days before it moved slowly northward. It took
279 approximately 60-75 days for the drifter to reach the investigated area located eastward of the
280 Kerguelen Islands (Fig. 7). It took approximately 53-65 days for the second drifter to reach
281 the area eastward of Kerguelen. Such transit times agree with the estimate of Park et al.
282 (2008b) during the KEOPS-1 project (i.e. several months between Heard Islands and the
283 eastern flank of the Kerguelen Islands). With such a transit time, a water body that interacted
284 with the shallow margins of Heard Island should not contain any remaining short-lived
285 radium isotopes when reaching the eastern flank of the Kerguelen Islands. As a consequence,

286 the ^{224}Ra and ^{223}Ra activities found in offshore waters east of the Kerguelen Islands, south of
287 the PF are best explained by diffusion or advection of Ra via waters that recently interacted
288 with the shallow sediments of the northern Kerguelen Plateau. This scenario, however,
289 implies that the Ra isotopes (and potentially other chemical elements such as iron) were
290 transferred offshore across or via the PF. High dissolved and particulate trace element
291 concentrations (Fe, Ni and Co) were also found east of the PF confirming that chemical
292 elements may be transported offshore across or via the PF (Qu  rou   et al. 2014; van der
293 Merwe et al., 2014). Among the potential mechanisms allowing surface waters to be
294 transported eastward across the PF, one can invoke either i) the wind stress (eastward winds
295 are especially strong in that region) or ii) eddies that form along the PF and that could
296 promote the passage of chemical elements across the front.

297 However, a contribution of surface waters originating from the southern Kerguelen
298 Plateau may not be completely excluded. In contrast to ^{224}Ra and ^{223}Ra that both disappear
299 due to radioactive decay along the northward transport, ^{228}Ra with a longer half-life would
300 remain in these waters. The ^{228}Ra activities observed eastward of the Kerguelen Islands may
301 thus be partly explained by an advective transport of waters originating from the south. It
302 cannot be excluded, therefore, that the northward advection originating from the southern
303 plateau contributes to the natural fertilization of the investigated area, in addition to the input
304 of chemical elements across the PF that was shown by the short lived isotopes.

305 South of the Kerguelen Islands (i.e. along the Polar Front at stations UW-23 and UW-24
306 or south of the Polar Front e.g. at stations UW-15, UW-16, R-2; Figure 4), it cannot be
307 completely excluded that the observed radium enrichments are partly explained by an input of
308 radium associated with the Leclaire Rise located west of the Kerguelen Islands at ca 350 m
309 depth (Weis and Frey, 2002). Station R-2, which is located east of the Leclaire Rise south of
310 the Polar Front, shows significant ^{223}Ra and ^{224}Ra activities in surface waters. Although these

311 activities are relatively low (0.016 and 0.057 dpm/ 100 L, respectively), they suggest that the
 312 waters downstream of the Leclaire Rise may be impacted by this topographic feature.
 313 However, sample UW-14 collected in surface waters lying above this rise does not show
 314 significant ^{223}Ra and ^{224}Ra activities and only low ^{228}Ra activity, which suggest that vertical
 315 mixing may not efficiently transport radium released by the shallow sediments towards
 316 surface waters above this topographic feature. Note that the influence of the Leclaire Rise on
 317 the chemical element concentrations downstream of the rise is also observed in Fe and other
 318 trace metal (REE, Mn, Al) concentrations, but only in waters lying in the 200-500 m depth
 319 interval (Bowie et al., 2014; van der Merwe et al., 2014; Grenier et al., in prep.).

320 4.2 Timescales of the offshore transport of surface waters

321 Once released into the water column, radium isotopes are subject to dilution, mixing
 322 and radioactive decay. The decay of short-lived radium isotopes in offshore waters provides
 323 information of how quickly chemical elements (including micronutrients) also released by the
 324 sediments are diluted and dispersed into the ocean (Moore, 2000). The presence of ^{224}Ra and
 325 ^{223}Ra in offshore waters thus indicates that the waters have recently been in interaction with
 326 the sediments. In contrast, when both ^{224}Ra and ^{223}Ra activities are below the detection limit,
 327 this suggests that the water bodies have not been in contact with the sediments over the past 2
 328 months (this is represented in light gray in Fig. 8). The water samples that display significant
 329 ^{223}Ra activity but no ^{224}Ra (represented in dark gray in Fig. 8) suggest that the interaction
 330 between the water body and the sediment occurred between 1 month (^{224}Ra activities < DL)
 331 and 2 months ago (significant ^{223}Ra activities). When both the ^{224}Ra and ^{223}Ra activities were
 332 significant, apparent ages could be calculated following (Moore, 2000) :

$$333 \quad t = \ln \frac{\left[\frac{^{224}\text{Ra}}{^{223}\text{Ra}} \right]_i}{\left[\frac{^{224}\text{Ra}}{^{223}\text{Ra}} \right]_{\text{obs}}} * \frac{1}{\lambda_{224} - \lambda_{223}} \quad (1)$$

334 where $(^{224}\text{Ra}/^{223}\text{Ra})_i$ is the initial ratio in source waters, $(^{224}\text{Ra}/^{223}\text{Ra})_{\text{obs}}$ is the ratio for a given
335 water sample, λ_{224} and λ_{223} are the decay constants of ^{224}Ra and ^{223}Ra , respectively. The
336 assumptions inherent to this equation can be found in Moore (2000) and are: (1) the ^{223}Ra and
337 ^{224}Ra activities are constant in the source region (i.e. a constant initial $^{224}\text{Ra}/^{223}\text{Ra}$ ratio is
338 assumed), (2) the $^{224}\text{Ra}/^{223}\text{Ra}$ ratio changes are only due to radioactive decay and (3) open
339 ocean waters contain no excess ^{223}Ra and ^{224}Ra .

340 In this study, we only reported the apparent ages deduced from the $^{224}\text{Ra}/^{223}\text{Ra}$ ratios
341 because we showed that both the ^{224}Ra and ^{223}Ra determined eastward of Kerguelen originate
342 from the shallow sediments of the Kerguelen Islands (see section 4.1). Apparent ages were
343 thus calculated using an initial $^{224}\text{Ra}/^{223}\text{Ra}$ ratio that was obtained by averaging the
344 ratios found in coastal water samples collected as close as possible to the radium source term
345 in the Baie du Morbihan (samples KER-1 and UW-36) and Baie des Baleiniers (samples
346 Baie-B1 and Baie-B2; Fig. 1). In contrast, we cannot exclude that ^{228}Ra has various origins
347 (Kerguelen Islands and/or Heard Island). The use of the $^{224}\text{Ra}/^{228}\text{Ra}$ or $^{223}\text{Ra}/^{228}\text{Ra}$ ratios to
348 derive apparent ages is thus compromised because it is not possible to determine a single
349 initial ratio in this case.

350 Several offshore samples display a young apparent age (4-8 days), suggesting a rapid
351 transport of radium between the shallow waters of the northern Kerguelen Plateau and
352 offshore. Station TNS1 located north of the PF is reached after 5 days. This observation
353 agrees with the circulation pattern in this area, with waters flowing eastward and that may
354 interact with the shallow northern Kerguelen Plateau (Park et al., 2014). This is also in
355 agreement with the drifters launched during the KEOPS 2 project that also highlighted such
356 advection along the PF (Fig. 7) (Zhou et al., 2014). Station UW-21 located ca. 50 km
357 offshore, station E1 and station UW-32 located ca. 200 km offshore also show relatively
358 young apparent ages (4, 5 and 6 days, respectively). Because all these stations are located

359 south of the PF, this suggests that the sediment-derived inputs may be rapidly transferred
360 towards offshore waters across the PF. Stations R-2 located south of the PF also displays a
361 young apparent age. At station Kerfix located close to station R-2, Jeandel et al. (1998)
362 reported westward currents associated with a recirculation pattern that may transport chemical
363 elements originating from the Kerguelen Plateau. The Ra signal may then be transported
364 eastward, as suggested by the significant ^{223}Ra activities also observed east of station R-2,
365 south of the PF (Fig. 4 and 8). Alternatively, the Leclaire Rise located west of R-2 may
366 impact the surface waters, thus leading to a young age for this water sample. Because the
367 ^{224}Ra and ^{223}Ra activities found at station A3-1 were attributed to the northward advection on
368 the southern Plateau, the apparent age at station A3-1 was calculated assuming that the initial
369 $^{224}\text{Ra}/^{223}\text{Ra}$ ratio off Heard Island is similar to that off Kerguelen Islands (Fig. 8). This
370 hypothesis may be correct since the geological contexts of the two islands are similar. The
371 apparent age thus calculated provides an estimate for the transit time of surface waters above
372 the southern Kerguelen Plateau between Heard Island and station A3. However, during the
373 second visit at station A3 (A3-2), the ^{224}Ra and ^{223}Ra activities were below the detection limit.
374 This may highlight the temporal variability in the circulation patterns in this area: the transit
375 time of surface waters between Heard Island and station A3 may thus vary with time, ranging
376 from one week to 1-2 months. On such timescales, the ^{228}Ra activities do not significantly
377 decay, which would explain why similar ^{228}Ra activities were found during the two visits at
378 A3. Finally, the spatial variability in the distribution of the apparent ages in offshore waters
379 suggests that the passage of chemical elements across the Polar Front is a sporadic process,
380 which may contribute to explain the mosaic structure of the phytoplankton bloom. Future
381 studies in the area could aim to track more precisely the sedimentary sources of radium (and
382 other chemical elements) and to quantify the radium fluxes out of the sediments using e.g. the
383 method developed by Cai et al. (2012).

384 4.3 Lagrangian particle analysis

385 To provide additional constraints on the origin of the Ra signal in offshore waters,
386 Lagrangian analyses derived from total surface currents were conducted at several stations
387 (Fig. 9). A two month backward analysis - to account for the life time of ^{223}Ra - was
388 performed starting from the sampling date for targets that were centered on the station
389 locations.

390 The Lagrangian analyses for the southern stations A3-1, A3-2, UW-35 and G-1 are
391 reported on Fig. 9. The backward trajectories provide a similar pattern and indicate a southern
392 origin for the surface waters found at these stations. This pattern agrees with the trajectories
393 of the two drifters released *in situ* at station A3 (Fig. 7). Waters that have interacted with the
394 shallow shelves of the southern Kerguelen Plateau (Heard Island) may thus reach the
395 investigated area. In particular, this northward advection may explain the significant Ra
396 activities determined at stations G-1 and A3-1. Note that the distance covered by the
397 backward trajectories over the 2 months is short, thus reflecting the relatively slow currents in
398 this area.

399 Lagrangian analyses were also performed for several northern stations: E-1, UW-31,
400 UW-32, TEW-7 and F-L. The trajectories are represented in shades of red on Fig. 9. Stations
401 F-L, TEW-7 and UW-31 are located relatively close to each other, east of the Kerguelen
402 Islands in the area of the southern branch of the cyclonic meander formed by the PF. Their
403 backward trajectories display a similar feature and all point to the same origin, which is the
404 northern Kerguelen Plateau. This suggests that chemical elements originating from the
405 Kerguelen Plateau may be transported offshore via the PF. The transit time given by the
406 Lagrangian analysis is approximately one month between the coast of the Kerguelen Islands
407 and the investigated stations. With a transit time of approximately one month, the ^{224}Ra

408 activities should have disappeared due to radioactive decay - or should be close to the
409 detection limit - while the ^{223}Ra activities should have significantly decayed. As a
410 comparison, the ^{224}Ra and ^{223}Ra activities are below the detection limit at stations F-L and
411 UW-31, whereas significant ^{223}Ra and ^{224}Ra activities were found at station TEW-7. Such
412 discrepancy between the investigated stations may highlight the spatial variability of the
413 circulation patterns in this area or that the Ra activities are close to the detection limits. Both
414 ^{224}Ra and ^{223}Ra activities are also significant at station E-1 located in the center of the
415 cyclonic meander formed by the PF. The Lagrangian analysis suggests that the surface waters
416 at station E-1 originate from the southwest. These waters flow northwards before reaching the
417 PF area and then follow the eastern shelf of the northern Kerguelen Plateau. When passing
418 close to the PF, these waters may receive significant Ra inputs (and potentially other
419 sediment-derived inputs) that could be transported via or across the front in this area. This Ra
420 signal may then be transferred to station E-1, as suggested by the backward trajectories. This
421 hypothesis is also supported by the study conducted by Zhou et al. (2014) who identified a
422 north-eastward drift of surface waters originating from the Kerguelen Plateau. Finally, the
423 backward trajectories at station UW-32 - that also displayed significant ^{223}Ra and ^{224}Ra -
424 highlight the spatial variability in that area: while several trajectories originate from the south,
425 several other trajectories follow the PF and the shelves of the northern Kerguelen Plateau,
426 where these waters could also potentially receive sediment-derived inputs.

427 4.4 Comparison of the apparent radium ages with an altimetry-based Lagrangian model

428 The timescale of the offshore transport of surface waters was also investigated using an
429 altimetry-based Lagrangian model (Fig. 10). The color bar indicates the time (number of
430 days) elapsed since the water body left the 2000 m-isobath. A color code similar to that of
431 Fig. 8 was used. A color palette from red to yellow highlights the rapid offshore transport of
432 the surface waters (surface waters < 6 days). The dark gray coding illustrates surface waters

433 that left the 2000 m-isobath less than 1 month ago. Finally, surface waters that left the 2000
434 m-isobath more than 1 month ago are represented in light gray. As a comparison, the radium
435 apparent ages are reported on the map using the same color code.

436 Young ages can be found close to the 2000 m-isobath, along the PF. Surface waters < 1
437 month follow the cyclonic meander formed by the PF, while waters older than 2 months are
438 found in the center of the meander. Note that the altimetry-derived Lagrangian analysis may
439 misplace structures with errors of ~10 km (e.g. d' Ovidio et al., 2010), which is comparable to
440 the width of the structures visible in the map. It may thus be difficult to compare
441 quantitatively the altimetry-derived ages with the ages determined *in situ*. Nevertheless, two
442 important considerations can be made: (i) the order of magnitude of the satellite-derived and
443 *in situ* ages are consistent in the region; (ii) considering a west-east transect from Kerguelen,
444 both estimations indicate a transition from young to old and then again young ages, which is
445 consistent with the existence of a retentive recirculation region centered at about 73 °W, 49
446 °S.

447 **5 Conclusion**

448 The observation of short-lived Ra isotopes (^{223}Ra and ^{224}Ra) in surface waters east of the
449 Kerguelen Islands, south of the PF clearly indicates that these waters have recently interacted
450 with shallow sediments. Neither the shallow margins of Heard Islands - located hundreds
451 kilometers south of the study area - nor the vertical mixing of deep waters that interacted with
452 bottom sediments can account for the short-lived radium enrichments found in surface waters.
453 The ^{223}Ra and ^{224}Ra activities south of the PF are thus best explained by waters that interacted
454 with the shallow margins of the Kerguelen Islands. This finding implies that chemical
455 elements can be transported across or via the PF. Among the potential mechanisms allowing
456 surface waters to be transported eastward across the PF, one can invoke either i) the wind

457 stress (eastward winds are especially strong in that region) or ii) eddies that form along the PF
458 and that may promote the transport of surface waters and associated chemical elements. The
459 spatial variability observed in the ^{223}Ra and ^{224}Ra distribution in surface waters south of the
460 PF suggests that the input of waters and associated chemical elements across the PF –
461 potentially driven by wind stress or eddies - act as sporadic pulses that may highly vary in
462 both space and time. This pathway may thus constitute a mechanism that contribute to
463 fertilize the phytoplankton bloom with iron and other micronutrients east of the Kerguelen
464 Islands, south of the PF. This finding shows that the PF may not act as a strong barrier for
465 surface waters and associated chemical elements, a finding that may also apply to other
466 frontal systems of the world's ocean.

467 **Author contribution**

468 P. van Beek and B. Lansard performed the sample collection on board the R/V Marion
469 Dufresne. The sample analysis was done on the ship by P. van Beek and B. Lansard and in the
470 laboratory by V. Sanial, and M. Souhaut. F. d'Ovidio developed the model code and E.
471 Kestenare performed the CTD analysis and the simulations for the Lagrangian analysis. M.
472 Zhou provided the drifter data. S. Blain is PI of the KEOPS-2 project and helped interpret the
473 data. V. Sanial interpreted the data and prepared the manuscript.

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484 Polaire Paul-Emile Victor).

485 **Figure captions**

486 **Fig. 1.** Location of stations investigated for Ra analysis. Solid circles represent surface
487 seawater samples. Circles show the locations where vertical profiles were made. KP is the
488 abbreviation for Kerguelen Plateau.

489 **Fig. 2.** Satellite composite images of sea surface chlorophyll *a* (mg m^{-3}) at successive dates
490 between the beginning of the KEOPS-2 cruise (19/10/2011) and the end of the cruise
491 (23/11/2011). The location of the water samples collected for radium analysis within the
492 different time intervals is also reported (solid circles).

493 **Fig. 3.** Potential temperature-salinity diagrams at stations where radium analyses were
494 performed. F-L station is plotted in bold. TEW-3, G-1, E-1, E4-W and A3-2 stations are
495 plotted in gray. The main water masses are reported on the figure: Antarctic Surface Water
496 (AASW), SubAntarctic Surface Water (SASW), Winter Water (WW), Upper Circumpolar
497 Deep Water (UCDW), Lower Circumpolar Deep Water (LCDW) and Antarctic Bottom Water
498 (AABW).

499 **Fig. 4.** ^{224}Ra (A), ^{223}Ra (B) and ^{228}Ra (C) distributions in surface waters off the Kerguelen
500 Islands. Radium activities are expressed in dpm/ 100 L. White circles indicate samples with
501 Ra activity below the detection limit. A schematic view of the Polar Front (PF) is shown.

502 **Fig. 5.** Vertical profiles of ^{223}Ra , ^{224}Ra and ^{228}Ra activities (dpm/ 100 L) at the shallow
503 stations. The main water masses are indicated: Winter Water (WW), Upper Circumpolar Deep
504 Water (UCDW). The bottom depth is denoted by the horizontal lines.

505 **Fig. 6.** ^{223}Ra , ^{224}Ra and ^{228}Ra activities (dpm/ 100 L) at the deep stations. The major water
506 masses are indicated: Winter Water (WW), Upper Circumpolar Deep Water (UCDW), Lower

507 Circumpolar Deep Water (LCDW), Antarctic Bottom Water (AABW). The bottom depth is
508 denoted by the horizontal lines.

509 **Fig. 7.** Trajectories of the drifters launched eastward of the Kerguelen Islands during the
510 KEOPS-2 project. The trajectories of the two drifters released at station A3 are reported in
511 color. The equivalent transit time of the two drifters is reported in days along their trajectory.
512 The other drifter trajectories are represented in light grey.

513 **Fig. 8.** Apparent ages of surface waters determined using the $^{224}\text{Ra}/^{223}\text{Ra}$ ratios. The offshore
514 apparent ages were estimated using an initial $^{224}\text{Ra}/^{223}\text{Ra}$ ratio that was obtained by averaging
515 the ratios found at stations located on the northern Kerguelen Plateau (< 200 m water depth).
516 When both ^{224}Ra and ^{223}Ra were significant, apparent ages could be determined (colored
517 symbols). The samples displaying an apparent age between 1 and 2 months are shown in dark
518 gray ($^{224}\text{Ra} < \text{DL}$ but significant ^{223}Ra activities). Water samples displaying an apparent age >
519 2 months are shown in light grey ($^{224}\text{Ra} < \text{DL}$ and $^{223}\text{Ra} < \text{DL}$). A schematic view of the Polar
520 Front (PF) is represented.

521 **Fig. 9.** Lagrangian particle analysis derived from total surface currents (considering absolute
522 geostrophic plus Ekman currents). Solid circles represent the location of the stations. The
523 targets for Lagrangian analysis were centered on and around the station locations (to account
524 for spatial variability). The sampling date is indicated in brackets. Two month-backward
525 trajectories are shown. The first month is represented in bold.

526 **Fig. 10.** Ages of surface waters (in days) derived from an altimetry Lagrangian-based model.
527 *In-situ* ages derived from radium isotopes are represented by circles. The colorbar indicates
528 the time elapsed since the water body left the 2000-m isobath.

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704

705 **Table 1:** Dissolved ^{223}Ra , ^{224}Ra and ^{228}Ra activities determined in surface samples collected off the
706 Kerguelen Islands. The ^{223}Ra and ^{224}Ra activities are excess radium activities (see Methods for details).
707 Activities are expressed in disintegration per minute per 100 L (dpm/ 100 L). <DL = Below the
708 Detection Limit.

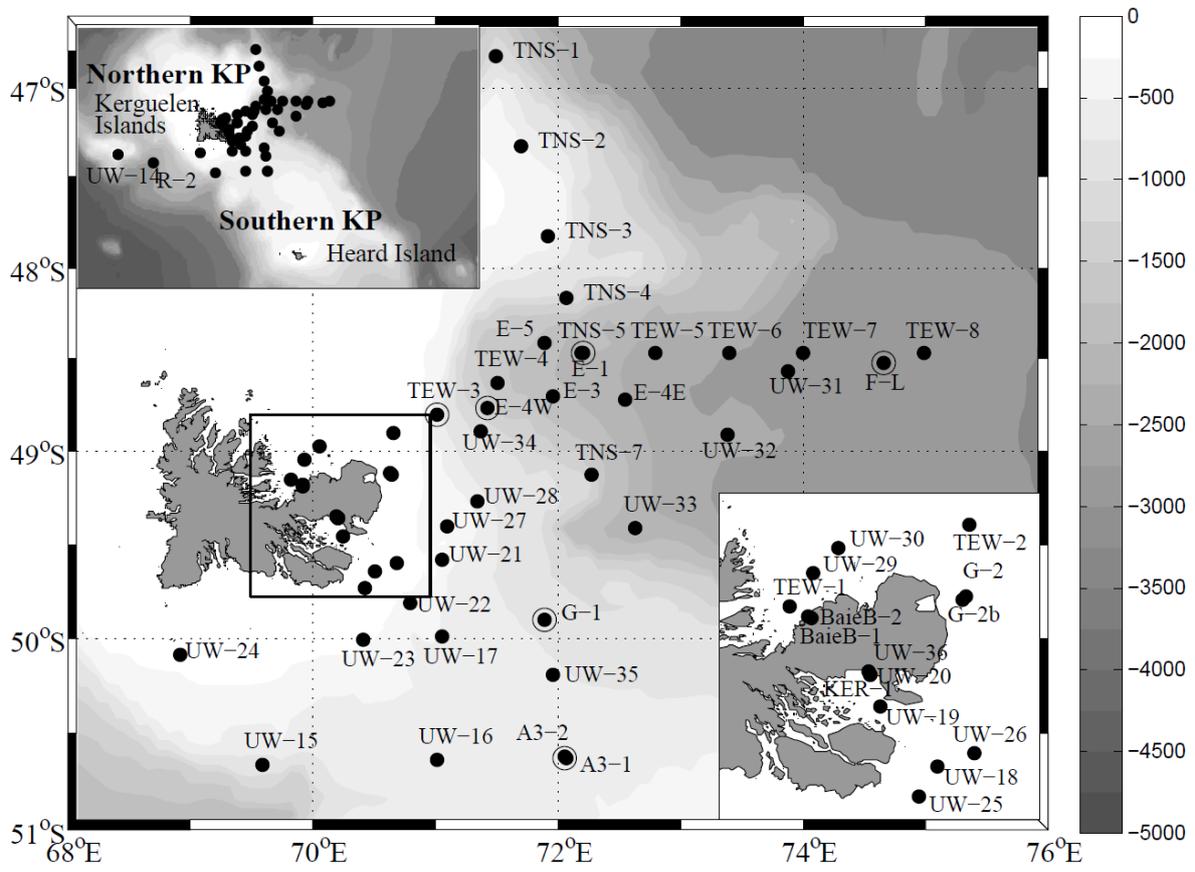
709 **Table 2:** Dissolved ^{223}Ra , ^{224}Ra and ^{228}Ra activities determined in seawater samples collected in the
710 water column using Niskin bottles. The ^{223}Ra and ^{224}Ra activities are excess radium activities (See
711 Methods for details). Activities are expressed in disintegration per minute per 100 L (dpm/ 100 L). The
712 number of counts detected using the RaDeCC is also reported in the Table (cnts). <DL = Below the
713 Detection Limit.

| Station | Sampling Date | Volume (L) | Depth (m) | Bottom Depth (m) | ²²³ Ra | | ²²⁴ Ra | | ²²⁸ Ra | | ²²³ Ra/ ²²⁸ Ra | | ²²⁴ Ra/ ²²⁸ Ra | | ²²⁴ Ra/ ²²³ Ra | |
|---------|------------------|------------|-----------|------------------|-------------------|---------|-------------------|---------|-------------------|---------|--------------------------------------|---------|--------------------------------------|--------|--------------------------------------|---------|
| | | | | | (dpm/ 100 L) | | (dpm/ 100 L) | | (dpm/ 100 L) | | | | | | | |
| UW-14 | 17/10/2011 12:10 | 900 | 7 | 342 | < LD | | < LD | | 0.06 ± 0.019 | | | | | | | |
| UW-15 | 18/10/2011 05:43 | 900 | 7 | 712 | 0.008 | ± 0.006 | < LD | | 0.03 | ± 0.010 | 0.24 | ± 0.19 | | | | |
| UW-16 | 18/10/2011 09:50 | 900 | 7 | 560 | 0.007 | ± 0.007 | < LD | | 0.05 | ± 0.013 | 0.13 | ± 0.13 | | | | |
| UW-17 | 19/10/2011 22:03 | 500 | 7 | 676 | < LD | | < LD | | 0.17 ± 0.035 | | | | | | | |
| UW-18 | 19/10/2011 01:30 | 250 | 7 | 104 | 0.062 | ± 0.018 | 0.213 | ± 0.062 | 0.49 | ± 0.079 | 0.13 | ± 0.04 | 0.44 | ± 0.15 | 3.45 | ± 1.41 |
| UW-19 | 19/10/2011 01:30 | 250 | 7 | 160 | 0.047 | ± 0.016 | 0.143 | ± 0.052 | 0.47 | ± 0.139 | 0.10 | ± 0.05 | 0.31 | ± 0.14 | 3.03 | ± 1.51 |
| UW-20 | 19/10/2011 04:15 | 250 | 7 | | 0.090 | ± 0.020 | 0.808 | ± 0.079 | 1.21 | ± 0.082 | 0.07 | ± 0.02 | 0.67 | ± 0.08 | 8.98 | ± 2.15 |
| UW-21 | 24/10/2011 10:46 | 700 | 7 | 597 | 0.011 | ± 0.005 | 0.036 | ± 0.024 | 0.11 | ± 0.026 | 0.10 | ± 0.05 | 0.33 | ± 0.24 | 3.38 | ± 2.69 |
| UW-22 | 24/10/2011 12:10 | 450 | 7 | 253 | 0.023 | ± 0.007 | < LD | | 0.20 ± 0.039 | | 0.11 ± 0.04 | | | | | |
| UW-23 | 24/10/2011 14:05 | 450 | 7 | 233 | 0.008 | ± 0.006 | 0.116 | ± 0.025 | 0.04 | ± 0.013 | 0.19 | ± 0.16 | 2.92 | ± 1.16 | 15.17 | ± 11.93 |
| UW-24 | 24/10/2011 23:37 | 450 | 7 | 171 | < LD | | < LD | | 0.11 ± 0.020 | | | | | | | |
| UW-25 | 28/10/2011 03:40 | 700 | 7 | 116 | 0.060 | ± 0.011 | 0.218 | ± 0.055 | 0.08 | ± 0.017 | 0.72 | ± 0.19 | 2.62 | ± 0.84 | 3.64 | ± 1.11 |
| UW-26 | 28/10/2011 04:35 | 700 | 7 | 130 | 0.043 | ± 0.010 | 0.065 | ± 0.033 | 0.19 | ± 0.023 | 0.23 | ± 0.06 | 0.34 | ± 0.18 | 1.50 | ± 0.84 |
| UW-27 | 28/10/2011 06:00 | 500 | 7 | 393 | 0.019 | ± 0.007 | < LD | | 0.06 ± 0.016 | | 0.31 ± 0.14 | | | | | |
| UW-28 | 28/10/2011 06:55 | 700 | 7 | 650 | 0.020 | ± 0.008 | < LD | | 0.07 ± 0.015 | | 0.29 ± 0.13 | | | | | |
| UW-29 | 31/10/2011 07:44 | 450 | 7 | 100 | 0.031 | ± 0.011 | 0.118 | ± 0.045 | 0.53 | ± 0.038 | 0.06 | ± 0.02 | 0.22 | ± 0.09 | 3.81 | ± 2.02 |
| UW-30 | 31/10/2011 08:17 | 450 | 7 | 100 | 0.083 | ± 0.014 | 0.201 | ± 0.039 | 0.58 | ± 0.039 | 0.14 | ± 0.03 | 0.35 | ± 0.07 | 2.44 | ± 0.62 |
| UW-31 | 05/11/2011 12:20 | 700 | 7 | | < LD | | < LD | | 0.17 ± 0.027 | | | | | | | |
| UW-32 | 08/11/2011 08:50 | 700 | 7 | 4561 | 0.013 | ± 0.008 | 0.035 | ± 0.046 | 0.24 | ± 0.040 | 0.05 | ± 0.04 | 0.15 | ± 0.19 | 2.82 | ± 4.10 |
| UW-33 | 08/11/2011 13:17 | 450 | 7 | 1664 | < LD | | < LD | | 0.09 ± 0.030 | | | | | | | |
| UW-34 | 09/11/2011 01:50 | 500 | 7 | 1118 | 0.017 | ± 0.011 | 0.124 | ± 0.078 | 0.10 | ± 0.020 | 0.16 | ± 0.12 | 1.23 | ± 0.80 | 7.45 | ± 6.84 |
| UW-35 | 17/11/2011 16:41 | 700 | 7 | 554 | < LD | | < LD | | 0.16 ± 0.026 | | | | | | | |
| UW-36 | 21/11/2011 05:25 | 500 | 7 | 21 | 0.098 | ± 0.016 | 0.411 | ± 0.098 | 1.07 | ± 0.054 | 0.09 | ± 0.02 | 0.38 | ± 0.09 | 4.18 | ± 1.22 |
| UW-37 | 22/11/2011 08:35 | 900 | 7 | 3720 | < LD | | < LD | | 0.13 ± 0.016 | | | | | | | |
| TNS-1 | 23/10/2011 11:55 | 500 | 7 | 2280 | 0.015 | ± 0.005 | 0.046 | ± 0.033 | 0.07 | ± 0.022 | 0.21 | ± 0.10 | 0.65 | ± 0.50 | 3.11 | ± 2.46 |
| TNS-2 | 23/10/2011 07:00 | 700 | 7 | 520 | 0.023 | ± 0.006 | < LD | | 0.31 ± 0.064 | | 0.07 ± 0.02 | | | | | |
| TNS-3 | 23/10/2011 03:23 | 700 | 7 | 540 | 0.006 | ± 0.004 | < LD | | 0.15 ± 0.050 | | 0.04 ± 0.03 | | | | | |
| TNS-4 | 22/10/2011 19:30 | 700 | 7 | 1800 | 0.015 | ± 0.005 | < LD | | 0.08 ± 0.017 | | 0.20 ± 0.08 | | | | | |
| TNS-5 | 22/10/2011 11:55 | 700 | 7 | 2060 | 0.021 | ± 0.005 | 0.070 | ± 0.030 | 0.17 | ± 0.050 | 0.13 | ± 0.05 | 0.42 | ± 0.22 | 3.26 | ± 1.62 |
| TNS-7 | 21/10/2011 20:25 | 700 | 7 | 1864 | < LD | | < LD | | 0.03 ± 0.012 | | | | | | | |
| TEW-1 | 31/10/2011 05:56 | 700 | 7 | 92 | 0.014 | ± 0.011 | 0.131 | ± 0.045 | 0.84 | ± 0.051 | 0.02 | ± 0.01 | 0.16 | ± 0.05 | 9.31 | ± 7.86 |
| TEW-2 | 31/10/2011 10:40 | 450 | 7 | 85 | 0.039 | ± 0.011 | 0.153 | ± 0.045 | 0.28 | ± 0.027 | 0.14 | ± 0.04 | 0.55 | ± 0.17 | 3.89 | ± 1.56 |
| TEW-3 | 31/10/2011 17:35 | 500 | 7 | 557 | 0.008 | ± 0.007 | < LD | | 0.10 ± 0.018 | | 0.08 ± 0.07 | | | | | |
| TEW-4 | 01/11/2011 03:45 | 500 | 7 | 1596 | < LD | | < LD | | 0.08 ± 0.025 | | | | | | | |
| TEW-5 | 01/11/2011 16:05 | 450 | 7 | 2290 | < LD | | 0.128 | ± 0.044 | 0.18 | ± 0.033 | | | 0.71 | ± 0.28 | | |
| TEW-6 | 02/11/2011 00:05 | 450 | 7 | 2400 | 0.011 | ± 0.006 | < LD | | 0.08 ± 0.019 | | 0.13 ± 0.08 | | | | | |
| TEW-7 | 02/11/2011 05:15 | 700 | 7 | 2510 | 0.020 | ± 0.009 | 0.147 | ± 0.062 | 0.16 | ± 0.019 | 0.13 | ± 0.06 | 0.93 | ± 0.40 | 7.37 | ± 4.51 |
| TEW-8 | 02/11/2011 17:40 | 900 | 7 | 2800 | < LD | | < LD | | 0.17 ± 0.024 | | | | | | | |
| E-1 | 29/10/2011 10:55 | 900 | 7 | 2065 | 0.021 | ± 0.005 | 0.070 | ± 0.022 | 0.10 | ± 0.022 | 0.23 | ± 0.08 | 0.73 | ± 0.28 | 3.26 | ± 1.29 |
| E-3 | 03/11/2011 16:15 | 900 | 7 | 1915 | 0.009 | ± 0.005 | < LD | | 0.03 ± 0.016 | | 0.33 ± 0.25 | | | | | |
| E-4W | 12/11/2011 06:15 | 900 | 7 | 1385 | 0.010 | ± 0.007 | < LD | | 0.14 ± 0.034 | | | | | | | |
| E-4E | 12/11/2011 08:37 | 500 | 7 | 2210 | < LD | | < LD | | 0.12 ± 0.021 | | | | | | | |
| E-5 | 18/11/2011 00:25 | 900 | 7 | 1920 | < LD | | < LD | | 0.11 ± 0.008 | | | | | | | |
| A3-1 | 19/10/2011 19:15 | 900 | 7 | 528 | 0.015 | ± 0.003 | 0.034 | ± 0.024 | 0.12 | ± 0.041 | 0.12 | ± 0.05 | 0.28 | ± 0.21 | 2.26 | ± 1.62 |
| A3-2 | 19/10/2011 19:15 | 900 | 7 | 531 | < LD | | < LD | | 0.12 ± 0.024 | | | | | | | |
| G-1 | 09/11/2011 05:20 | 900 | 7 | 592 | 0.023 | ± 0.007 | < LD | | 0.05 ± 0.020 | | 0.46 ± 0.23 | | | | | |
| G-2 | 09/11/2011 14:10 | 500 | 7 | 67 | 0.089 | ± 0.015 | 0.412 | ± 0.052 | 0.36 | ± 0.046 | 0.25 | ± 0.05 | 1.15 | ± 0.21 | 4.64 | ± 0.98 |
| G-2b | 21/11/2011 11:00 | 500 | 7 | 67 | 0.130 | ± 0.017 | 0.568 | ± 0.067 | 0.75 | ± 0.066 | 0.17 | ± 0.03 | 0.75 | ± 0.11 | 4.36 | ± 0.77 |
| F-L | 06/11/2011 13:33 | 900 | 7 | 2670 | < LD | | < LD | | 0.17 ± 0.021 | | | | | | | |
| R-2 | 26/10/2011 02:40 | 900 | 7 | 2531 | 0.016 | ± 0.009 | 0.057 | ± 0.028 | 0.11 | ± 0.016 | 0.15 | ± 0.09 | 0.53 | ± 0.27 | 3.49 | ± 2.51 |
| KER-1 | 19/10/2011 04:45 | 87.8 | 1 | 3 | 0.302 | ± 0.048 | 2.053 | ± 0.125 | 1.62 | ± 0.160 | 0.19 | ± 0.03 | 1.27 | ± 0.15 | 6.80 | ± 1.16 |
| BaieB-1 | 31/10/2011 05:00 | 99.3 | 1 | 3 | 0.219 | ± 0.032 | 2.332 | ± 0.118 | 0.88 | ± 0.089 | 0.25 | ± 0.04 | 2.65 | ± 0.30 | 10.67 | ± 1.67 |
| BaieB-2 | 40847.20833 | 65.2 | 1 | 3 | 0.012 | ± 0.008 | 0.256 | ± 0.024 | 2.57 | ± 0.183 | 0.005 | ± 0.003 | 0.10 | ± 0.01 | 20.64 | ± 13.83 |

| Station and Depth (m) | Volume (L) | Bottom Depth (m) | ²²³ Ra (dpm/ 100 L) | ²²³ Ra cnts | ²²⁴ Ra (dpm/ 100 L) | ²²⁴ Ra cnts | ²²⁸ Ra (dpm/ 100 L) | ²²⁸ Ra cnts |
|--------------------------|---------------|---------------------|-----------------------------------|---------------------------|-----------------------------------|---------------------------|-----------------------------------|---------------------------|
| E-1 | | | | | | | | |
| 182 | 256 | 2065 | < LD | | 0.041 ± 0.031 | 134 | 0.03 ± 0.01 | 44 |
| 508 | 263 | 2065 | 0.019 ± 0.012 | 38 | < LD | | < LD | |
| 1013 | 262 | 2065 | 0.010 ± 0.009 | 24 | < LD | | 0.07 ± 0.015 | 78 |
| 1623 | 256 | 2065 | 0.058 ± 0.013 | 74 | < LD | | 0.22 ± 0.034 | 134 |
| 2069 | 274 | 2065 | 0.170 ± 0.024 | 180 | 0.045 ± 0.037 | 344 | 0.23 ± 0.033 | 141 |
| TEW-3 | | | | | | | | |
| 101 | 259 | 557 | < LD | | < LD | | 0.29 ± 0.068 | 125 |
| 303 | 257 | 557 | 0.039 ± 0.013 | 37 | 0.065 ± 0.052 | 129 | 0.28 ± 0.037 | 154 |
| 557 | 252 | 557 | 0.014 ± 0.015 | 33 | 0.077 ± 0.056 | 213 | 0.37 ± 0.098 | 68 |
| F-L | | | | | | | | |
| 101 | 257 | 2670 | 0.008 ± 0.007 | 17 | < LD | | 0.20 ± 0.130 | 49 |
| 183 | 260 | 2670 | < LD | | 0.086 ± 0.022 | 246 | 0.19 ± 0.054 | 35 |
| 405 | 258 | 2670 | 0.016 ± 0.009 | 24 | < LD | | 0.06 ± 0.030 | 22 |
| 907 | 258 | 2670 | 0.039 ± 0.012 | 46 | 0.103 ± 0.060 | 107 | 0.12 ± 0.044 | 28 |
| 1825 | 122 | 2670 | 0.064 ± 0.017 | 32 | 0.128 ± 0.077 | 239 | 0.54 ± 0.130 | 49 |
| 2723 | 124 | 2670 | 0.142 ± 0.049 | 123 | 0.265 ± 0.154 | 346 | 0.93 ± 0.097 | 290 |
| G-1 | | | | | | | | |
| 10 | 269 | 592 | < LD | | < LD | | 0.38 ± 0.042 | 187 |
| 53 | 251 | 592 | 0.020 ± 0.009 | 30 | 0.051 ± 0.049 | 107 | 0.31 ± 0.039 | 157 |
| 130 | 255 | 592 | < LD | | < LD | | 0.26 ± 0.066 | 43 |
| 303 | 260 | 592 | < LD | | < LD | | 0.38 ± 0.081 | 56 |
| 455 | 234 | 592 | < LD | | < LD | | 0.29 ± 0.074 | 44 |
| 576 | 223 | 592 | 0.088 ± 0.021 | 84 | 0.075 ± 0.058 | 75 | 0.36 ± 0.077 | 48 |
| E-4W | | | | | | | | |
| 94 | 261 | 1385 | 0.020 ± 0.014 | 31 | < LD | | 0.34 ± 0.079 | 50 |
| 192 | 260 | 1385 | < LD | | < LD | | 0.30 ± 0.041 | 135 |
| 608 | 253 | 1385 | < LD | | < LD | | 0.29 ± 0.043 | 151 |
| 1013 | 123 | 1385 | < LD | | 0.133 ± 0.046 | 93 | 0.43 ± 0.116 | 36 |
| 1383 | 123 | 1385 | 0.057 ± 0.021 | 25 | 0.071 ± 0.046 | 145 | 0.90 ± 0.164 | 59 |
| A3-2 | | | | | | | | |
| 101 | 258 | 531 | < LD | | < LD | | 0.10 ± 0.042 | 25 |
| 152 | 246 | 531 | < LD | | < LD | | 0.16 ± 0.029 | 115 |
| 233 | 258 | 531 | < LD | | < LD | | 0.18 ± 0.060 | 33 |
| 303 | 124 | 531 | < LD | | < LD | | 0.49 ± 0.125 | 39 |
| 404 | 110 | 531 | < LD | | < LD | | 0.68 ± 0.156 | 44 |
| 518 | 246 | 531 | 0.081 ± 0.019 | 58 | < LD | | 0.70 ± 0.102 | 82 |
| TEW-1 | | | | | | | | |
| 82 | 258.5 | 92 | 0.053 ± 0.015 | 49 | 0.125 ± 0.059 | 334 | 0.88 ± 0.083 | 197 |
| TEW-8 | | | | | | | | |
| 20 | 269 | 2800 | 0.011 ± 0.009 | 22 | < LD | 335 | 0.14 ± 0.032 | 69 |
| G-2 | | | | | | | | |
| 50 | 229.6 | 67 | 0.094 ± 0.022 | 65 | 0.737 ± 0.097 | 389 | 0.79 ± 0.083 | 167 |

715 Fig. 1.

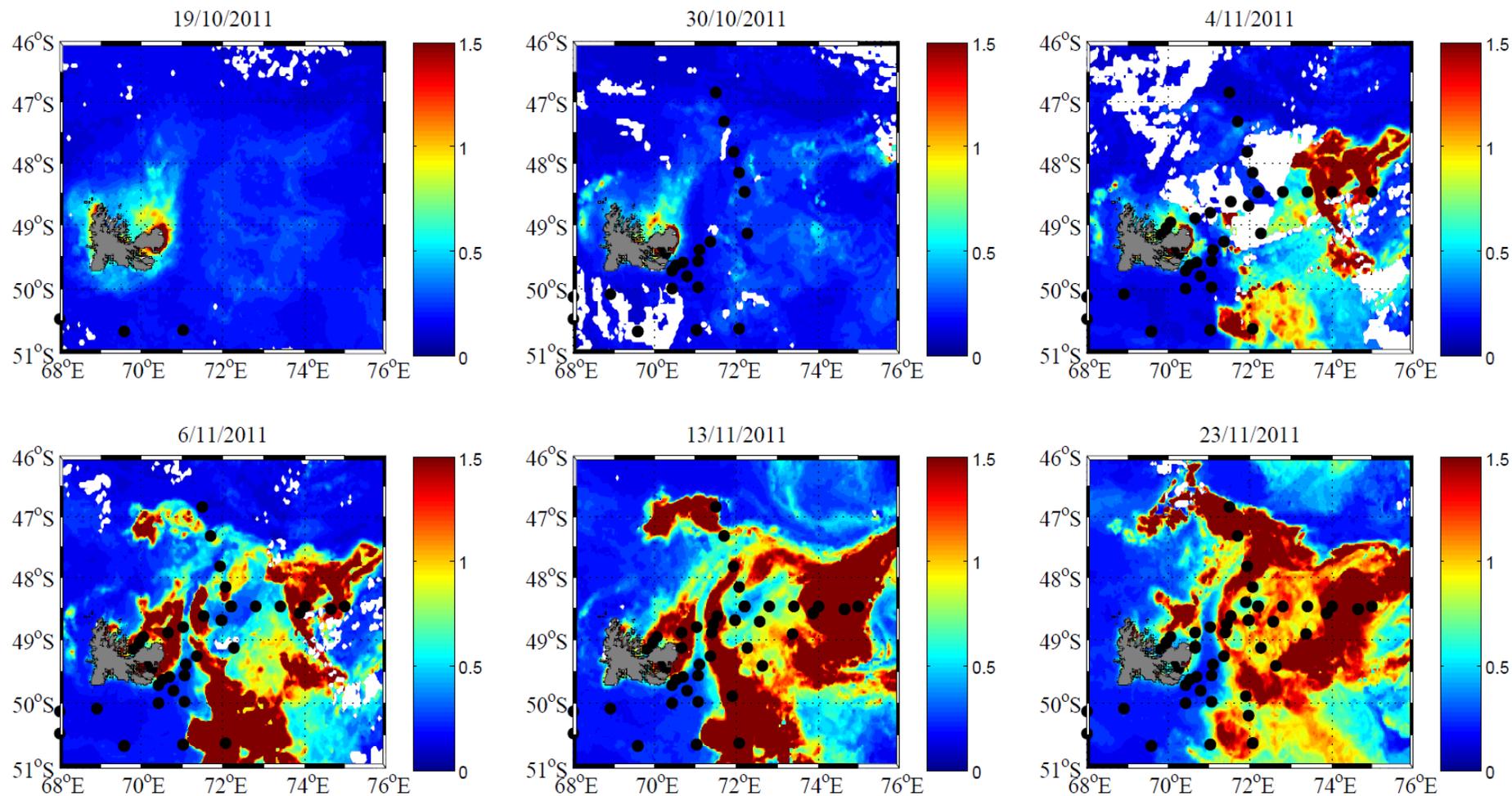
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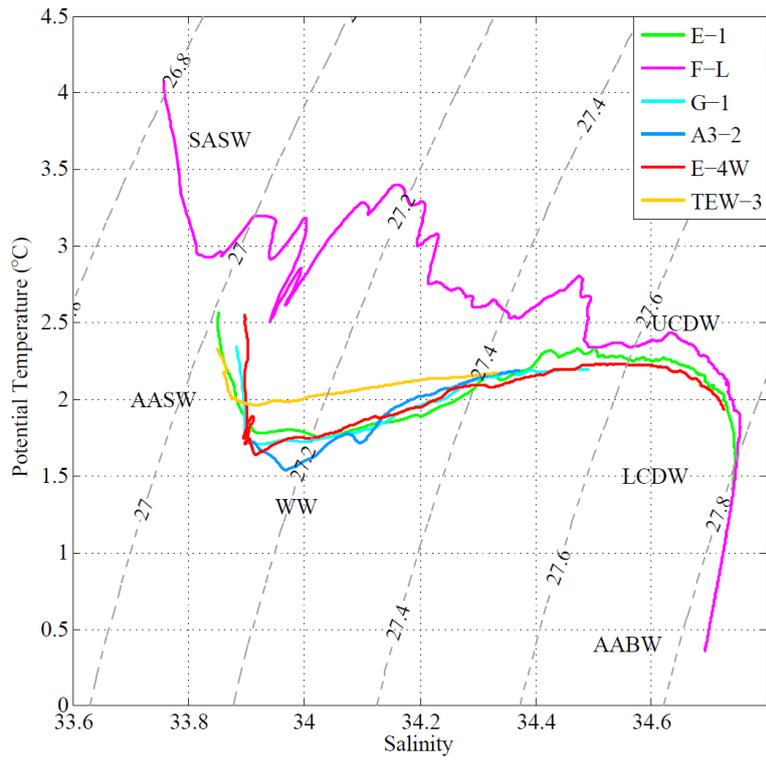
719 Fig. 2.



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722 Fig. 3.



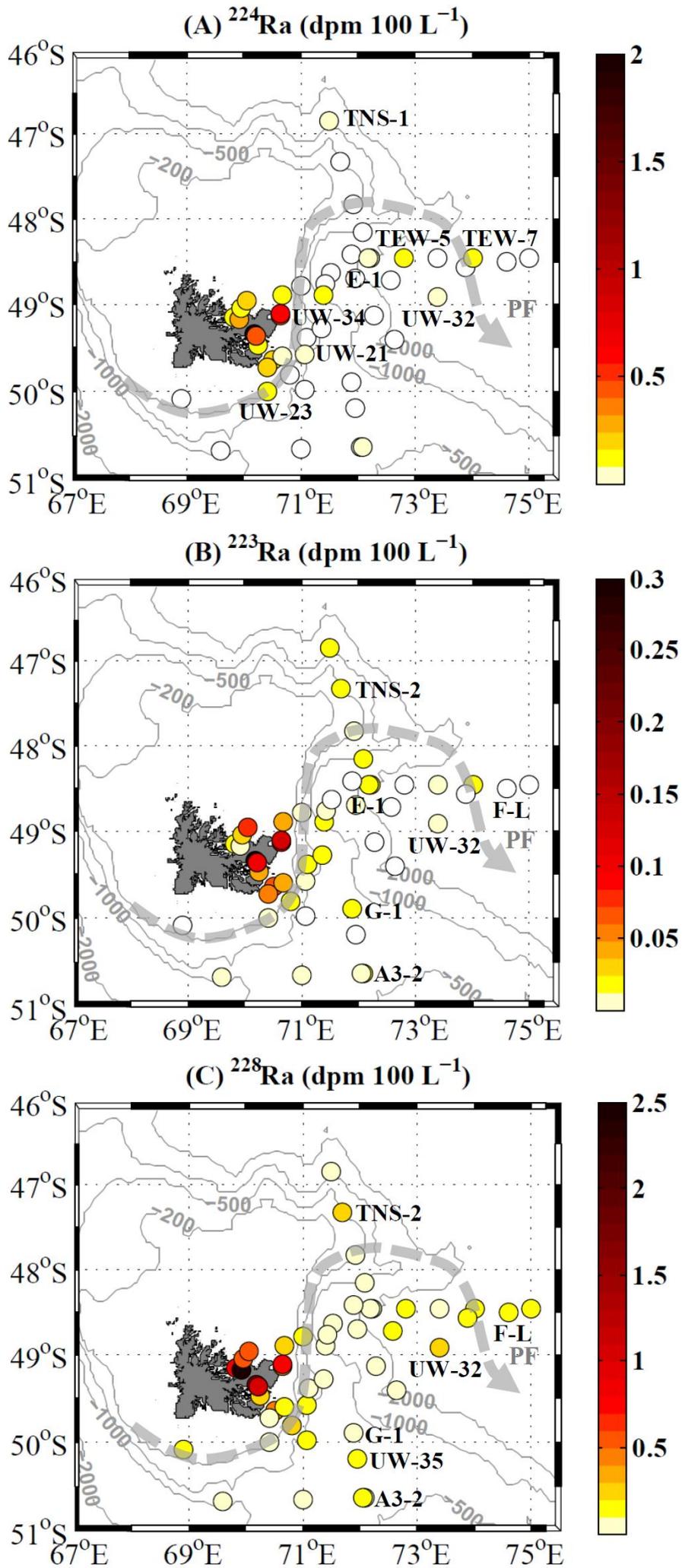
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725 Fig. 4.

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728 Fig. 5.

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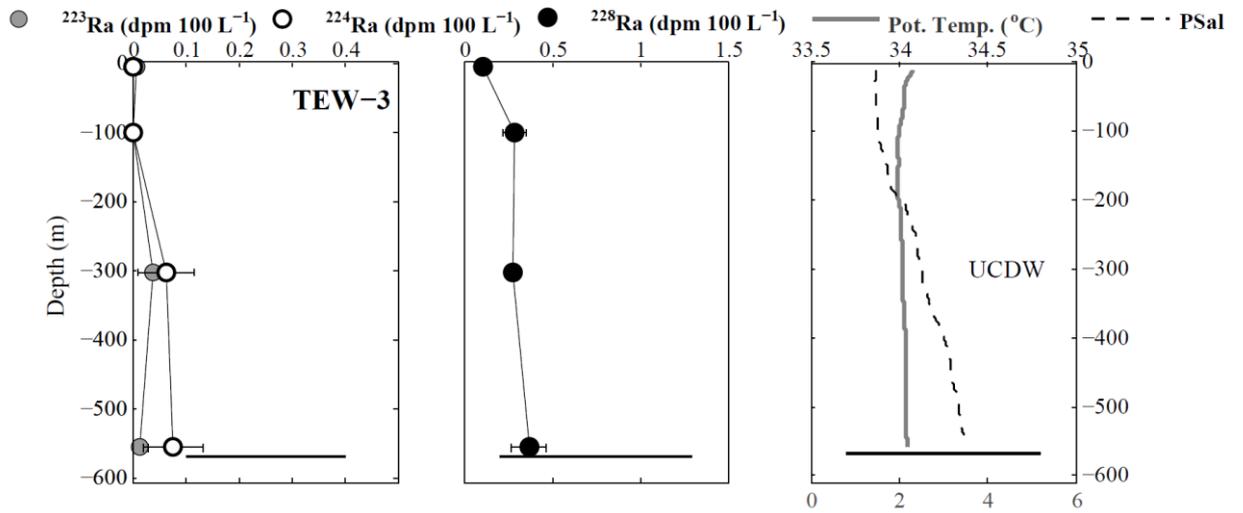
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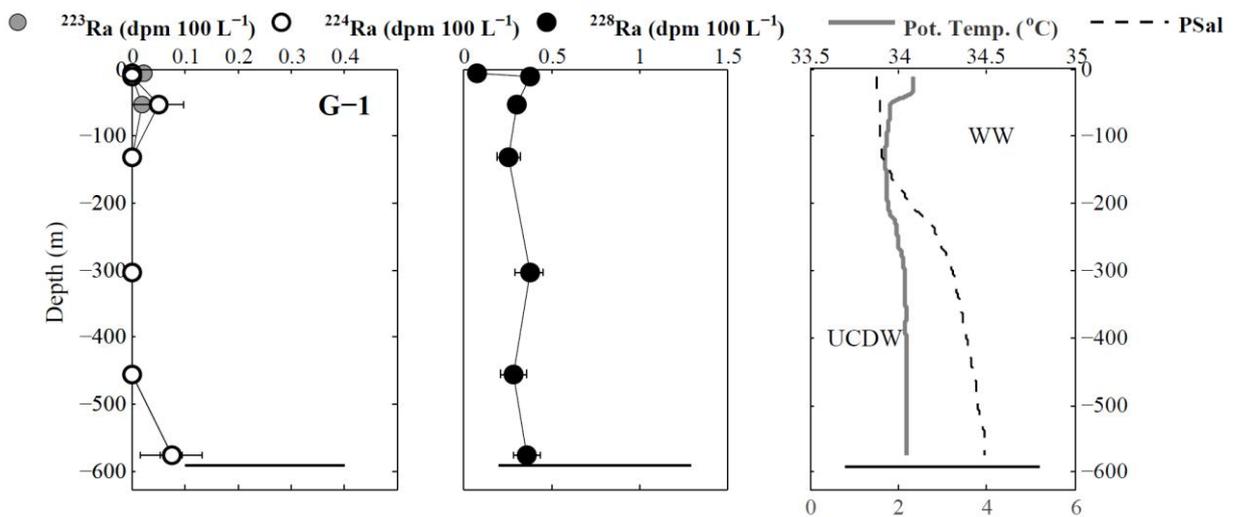
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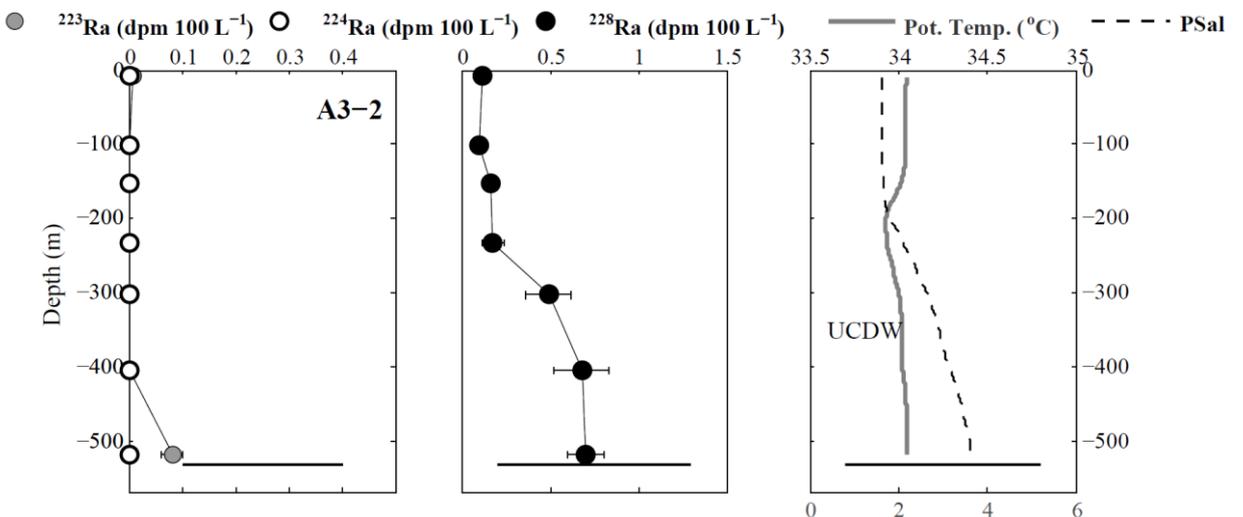
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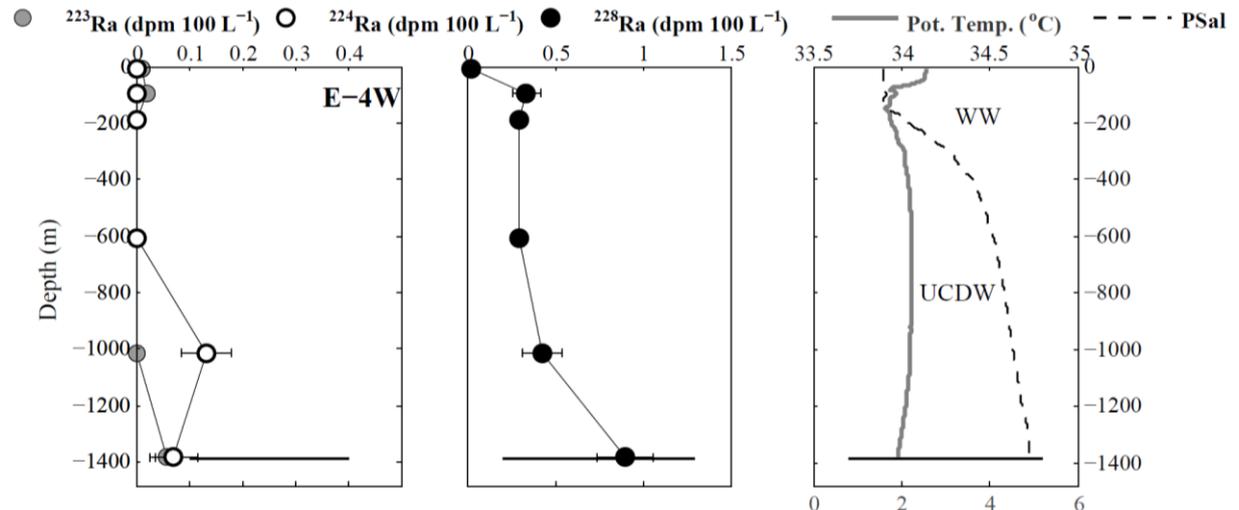
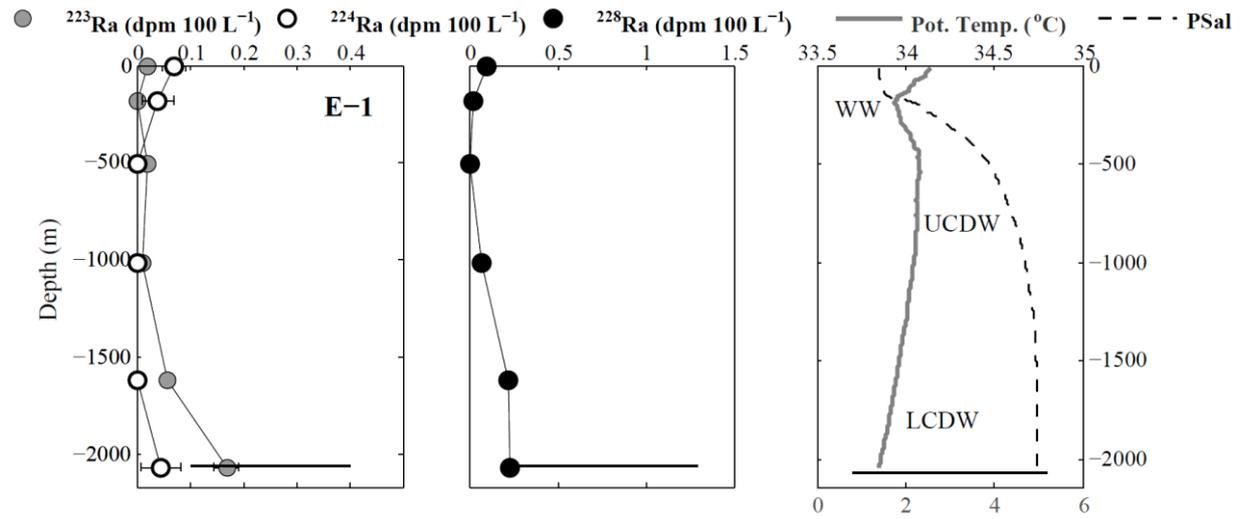
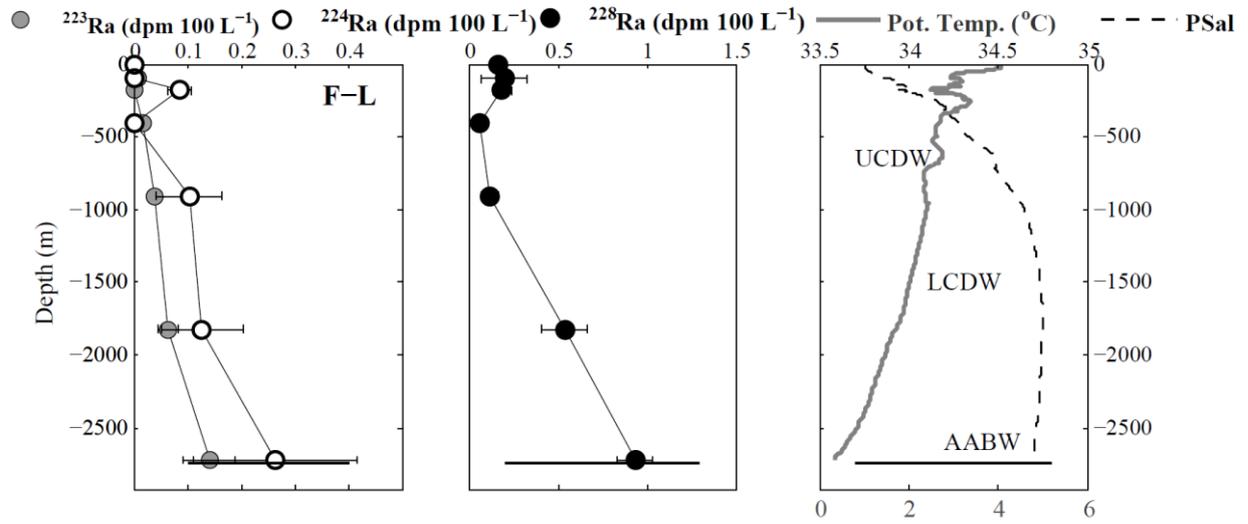
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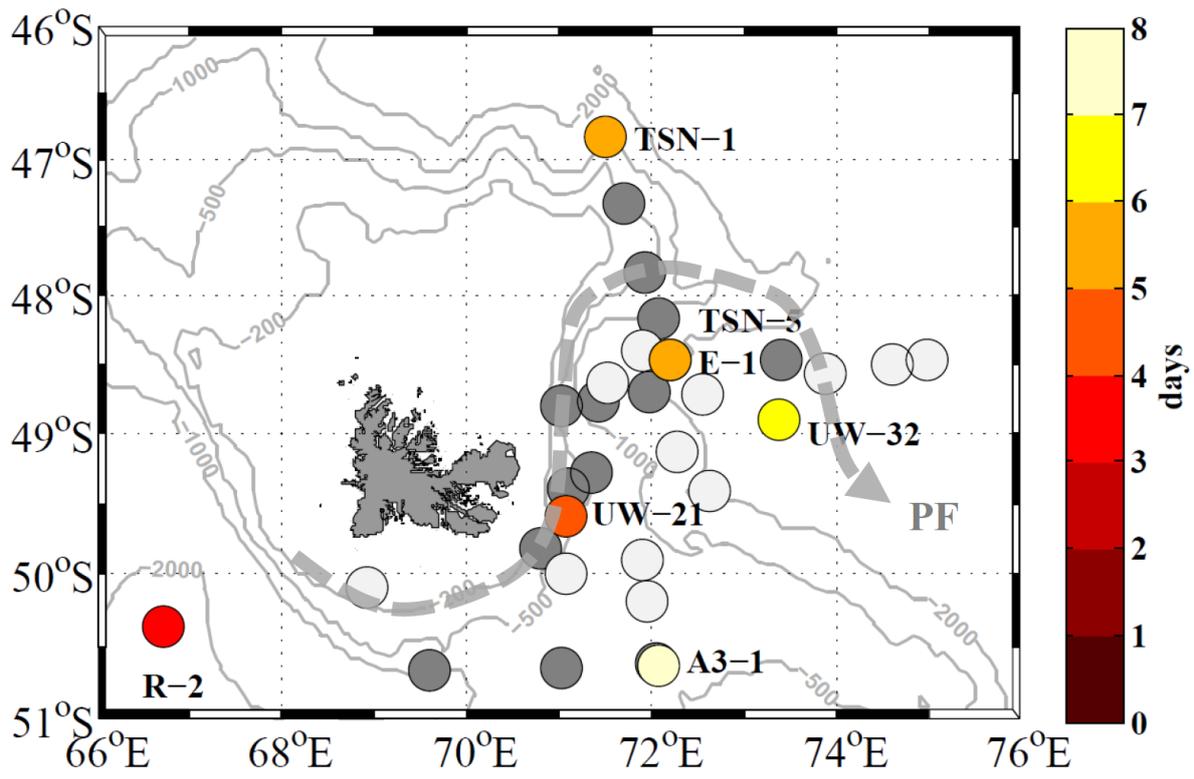
749 Fig. 6.

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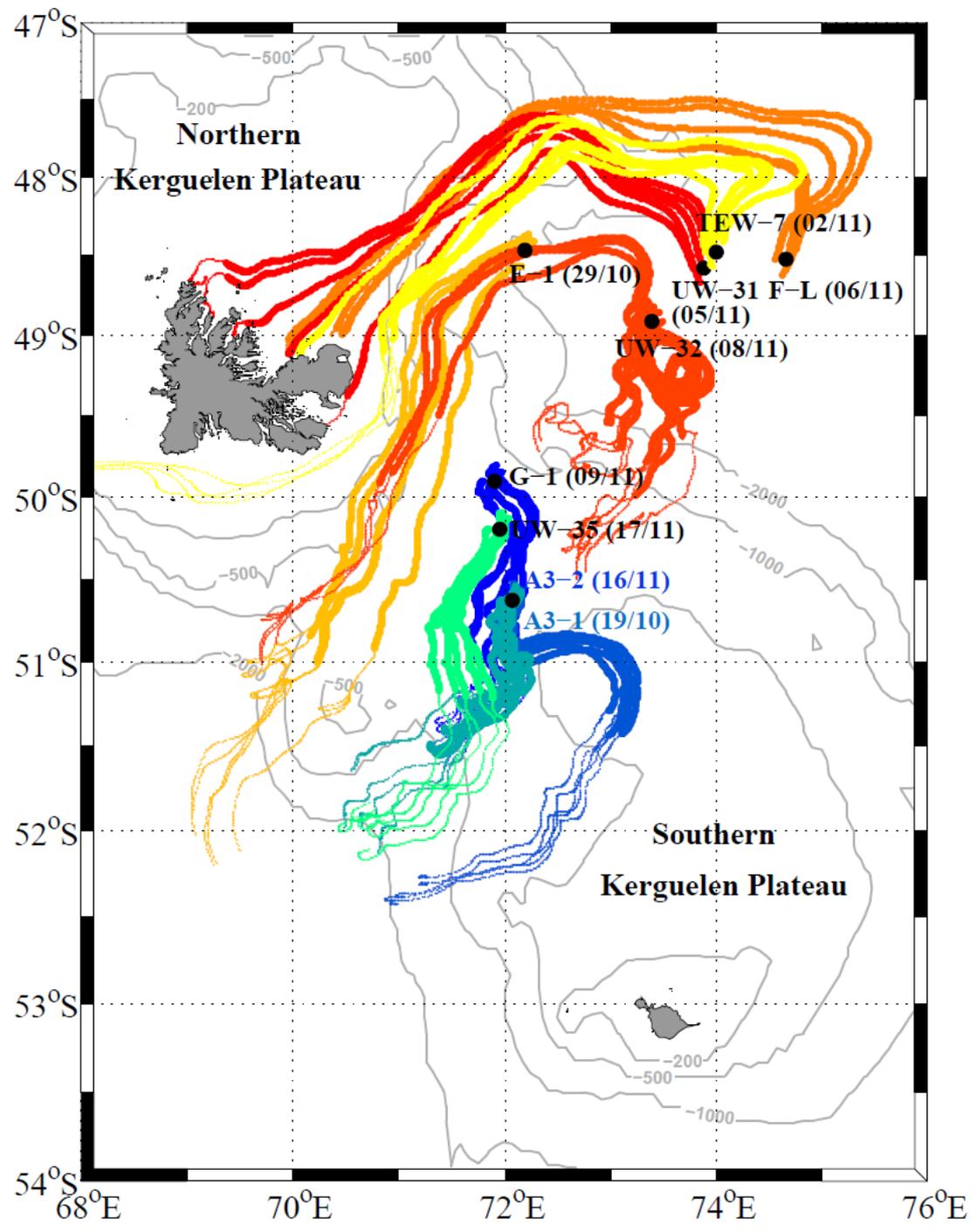
755 Fig. 8.



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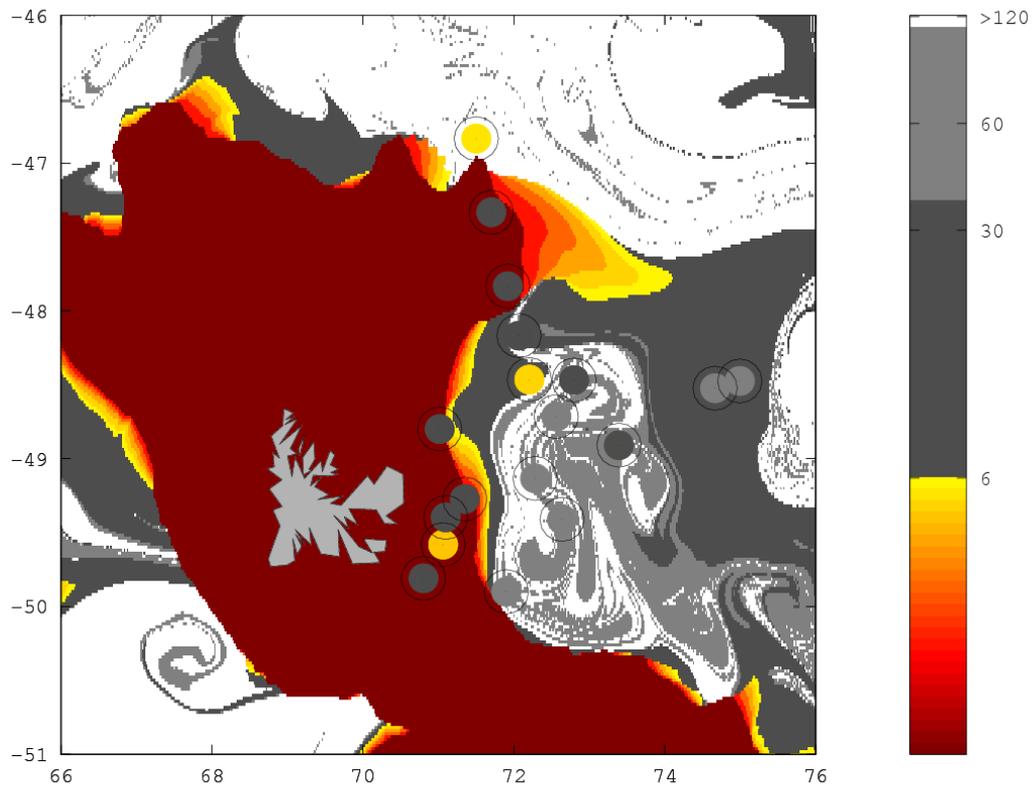
758 Fig. 9.



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761 Fig. 10



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