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1 **High particulate organic carbon export during the decline of a vast diatom bloom**
2 **in the Atlantic sector of the Southern Ocean**

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20 **Abstract**

21 Carbon fixation by phytoplankton plays a key role in the uptake of atmospheric
22 CO₂ in the Southern Ocean. Yet, it still remains unclear how efficiently the particulate
23 organic carbon (POC) is exported and transferred from ocean surface waters to depth
24 during phytoplankton blooms. In addition, little is known about the processes that
25 control the flux attenuation within the upper twilight zone. Here, we present results of
26 downward POC and particulate organic nitrogen fluxes during the decline of a vast
27 diatom bloom in the Atlantic sector of the Southern Ocean in summer 2012. We used
28 thorium-234 (²³⁴Th) as a particle tracer in combination with drifting sediment traps
29 (ST). Their simultaneous use evidenced a sustained high export rate of ²³⁴Th at 100 m
30 depth in the weeks prior to and during the sampling period. The entire study area, of

31 approximately 8000 km², showed similar vertical export fluxes in spite of the
32 heterogeneity in phytoplankton standing stocks and productivity, indicating a
33 decoupling between production and export. The POC fluxes at 100 m were high,
34 averaging 26 ± 15 mmol C m⁻² d⁻¹, although the strength of the biological pump was
35 generally low. Only <20% of the daily primary production reached 100 m, presumably
36 due to an active recycling of carbon and nutrients. Pigment analyses indicated that
37 direct sinking of diatoms likely caused the high POC transfer efficiencies (~60%)
38 observed between 100 and 300 m, although faecal pellets and transport of POC linked
39 to zooplankton vertical migration might have also contributed to downward fluxes.

40 *1. Introduction*

41 The Southern Ocean is an important sink for atmospheric CO₂ accounting for
42 about 15-20% of the global oceanic uptake (Gruber et al., 2009; Takahashi et al., 2002),
43 with a significant contribution from phytoplankton that fix CO₂ to organic carbon
44 (Hauck et al., 2013). However, the strength and efficiency of the biological pump (i.e.
45 export efficiency and transfer efficiency, respectively), as well as their controlling
46 factors are poorly understood, especially during phytoplankton blooms (Buesseler and
47 Boyd, 2009).

48 The export efficiency (i.e. the fraction of production that is exported from the
49 upper ocean, usually taken as the base of the euphotic zone or 100 m) is low in much of
50 the global ocean (<5-10%), but it is typically about 50% for blooms at high latitudes
51 (Buesseler, 1998). However, previous studies in the Southern Ocean have reported
52 lower export efficiencies during high-productive events (Jacquet et al., 2011; Planchon
53 et al., 2015; Rutgers van der Loeff et al., 1997; Savoye et al., 2008). Furthermore, iron-
54 fertilised blooms in the Southern Ocean show high variability in the fraction of

55 production being exported from the ocean surface. As an example, during SOFeX-South
56 (66°S) the export efficiency was low (<10%, Buesseler et al., 2005), while during
57 EIFEX it was approximately 60%, indicating a very strong biological pump (Smetacek
58 et al., 2012). A number of factors may explain the differences observed in export
59 efficiencies, including phytoplankton community composition and study time frame.
60 Diatoms are key exporters of carbon from the ocean surface to deep waters and
61 sediments, and hence play an essential role in reducing the CO₂ content in the
62 atmosphere (Smetacek, 1999). Previous studies have shown that very low macronutrient
63 concentrations, specifically of silicic acid, prevent the development of diatom blooms in
64 benefit of flagellates, resulting in low particulate organic carbon (POC) export fluxes
65 from the ocean surface (Jacquet et al, 2011; Martin et al., 2013). Further, diatoms in the
66 Southern Ocean contribute differently to POC export according to their life cycle
67 strategy in which the degree of silicification is relevant (Assmy et al., 2013; Quéguiner,
68 2013). Moreover, since export lags production, the time scale of the studies may often
69 be too short to quantitatively estimate the strength of the biological pump during bloom
70 events (Buesseler et al., 2004; Charette and Buesseler, 2000).

71 The transfer efficiency (i.e. the fraction of shallow export that is transferred to
72 depth) indicates the attenuation of the flux that takes place within a certain depth range.
73 While it is known that the attenuation of POC fluxes is sharpest in the upper twilight
74 zone, i.e. 100-300 m below the euphotic zone depth, our understanding of the processes
75 affecting sinking particles throughout this layer is still poor (Buesseler and Boyd, 2009).
76 The vertical flux of organic matter throughout the water column is dominated by large
77 particles such as marine snow and faecal pellets (Ebersbach et al., 2011; Fowler and
78 Knauer, 1986; Laurenceau-Cornec et al., 2015) that can be attenuated to a large extent
79 by zooplankton and microbial degradation (Giering et al., 2014; Iversen et al., 2010;

80 Kjørbe, 2000; Smith et al., 1992). However, packaging of slowly sinking phytoplankton
81 cells into large faecal pellets may play a key role in increasing the export and transfer
82 efficiencies in the Southern Ocean (Cavan et al., 2015; Le Moigne et al., 2014). Mineral
83 ballasting also appears to alter the efficiency by which POC is transported to depth (e.g.
84 Iversen and Robert, 2015; Klaas and Archer, 2002).

85 Thorium-234 (^{234}Th , half-life = 24.1 d) is widely used as a particle tracer, mainly
86 of POC, since it is particle reactive and its half-life allows studying events occurring
87 over short time scales, ranging from days to weeks, such as phytoplankton blooms (e.g.
88 Buesseler et al., 1992; Rutgers van der Loeff et al., 1997). A deficit of ^{234}Th with
89 respect to its parent ^{238}U is typically found in the upper ocean. Once the ^{234}Th
90 downward flux at a specific depth is quantified, this flux can be converted to POC and
91 particulate organic nitrogen (PON) fluxes by determining the $\text{POC}/^{234}\text{Th}$ and $\text{PON}/^{234}\text{Th}$
92 ratios, respectively, in sinking particles (Buesseler et al., 2006; Cochran and Masqué,
93 2003).

94 This study focuses on the decline of a vast diatom bloom that occurred in the
95 Antarctic Circumpolar Current (ACC) region of the Southern Ocean (around 51°S
96 13°W) during the late austral summer in 2012. Our objectives were to evaluate the
97 export efficiency and transfer efficiency of POC between 100 and 300 m, as well as
98 identify the main mechanisms that had an influence on particle fluxes. Particle fluxes
99 were quantified by means of two different techniques, as highly recommended given the
100 uncertainties associated with each collection method (e.g. Puigcorbé et al., 2015;
101 Turner, 2015). We used the disequilibrium between the natural radionuclides ^{234}Th and
102 ^{238}U to determine the export fluxes of POC and PON in parallel with the use of surface-
103 tethered drifting sediment traps. Export fluxes were related to the evolution of

104 chlorophyll *a* (Chl-*a*) and POC concentrations in the water column, as well as to
105 pigments in sinking particles, following the decline of the bloom during three weeks.
106 Net primary production (NPP) measured during the same cruise (Hoppe et al., this
107 issue) was used to assess the export efficiency.

108 *2. Methods*

109 *2.1. Study area*

110 Samples were collected from 29 January to 17 February 2012 during the ANT-
111 XXVIII/3 expedition in the Atlantic sector of the Southern Ocean (7 January-11 March,
112 2012; R/V Polarstern; Wolf-Gladrow, 2013). The sampling was carried out to study a
113 massive bloom with high spatial and temporal resolution over an area of about 8000
114 km² located between the Antarctic Polar Front (APF) and the Southern Polar Front
115 (SPF; Leach et al., and Strass et al., this issue). Time-series measurements of ²³⁴Th,
116 POC, PON, Chl-*a*, other pigments and NPP were carried out at a station located in the
117 centre of the study area at 51.21°S 12.67°W (hereafter “central station”, indicated by a
118 ‘C’ in front of the station number). The location and sampling dates of the stations
119 sampled for ²³⁴Th, POC and PON fluxes are given in Figure 1 and Table 1.

120 *2.2. Total ²³⁴Th and ²³⁸U*

121 Total ²³⁴Th activities were determined from 4 L seawater samples obtained from
122 Niskin bottles attached to a CTD rosette. Samples were collected from 11-13 depths
123 down to 500-750 m at 14 stations, with the highest resolution in the upper 200 m of the
124 water column. Three seawater profiles were taken at the central station on 3, 12 and 17
125 February (C91, C128, C140, respectively). In addition, replicates of deep samples (2500
126 m) were collected for calibration purposes (Rutgers van der Loeff et al., 2006). The

127 samples were processed according to the MnO₂ co-precipitation technique (Buesseler et
128 al., 2001b), using ²³⁰Th as a chemical yield tracer (Pike et al., 2005). The precipitates
129 were filtered through QMA quartz fibre filters, dried overnight at 50°C and prepared for
130 beta counting. The counting was done on board using low background beta counters
131 (Risø National Laboratories, Denmark). Samples were re-measured after 10 months to
132 quantify the background counts. All filters were processed to determine ²³⁰Th by
133 inductively coupled plasma mass spectrometry (ICP-MS) using ²²⁹Th as an internal
134 standard. Briefly, filters were spiked with ²²⁹Th and precipitates were dissolved in 10
135 mL of 8M HNO₃/10% H₂O₂ solution. Samples were sonicated for 30 minutes before
136 allowing them to rest for at least 6 hours. Prior to the ICP-MS analyses the samples
137 were filtered through Acrodisc 0.2 µm syringe filters and reconstructed with 2.5%
138 HNO₃/0.01% HF. The average chemical recovery of the analytical process was 94 ± 4%
139 (n = 173), and the uncertainty of the ²³⁰Th/²²⁹Th ratios averaged 1.5 ± 0.6% (n = 173).
140 The activity of ²³⁸U was derived from salinity using the relationship given by Owens et
141 al. (2011). The ²³⁴Th activity uncertainties were calculated by propagating uncertainties
142 associated with counting, detector background and calibration, ICP-MS measurements
143 and ²³⁸U activities, and were always <10% (average: 8.9 ± 0.3%, n = 173). All data of
144 total ²³⁴Th and ²³⁸U activities are available at
145 <http://dx.doi.org/10.1594/PANGAEA.848823>.

146 2.3. ²³⁴Th, POC and PON in particles

147 Sinking particles were collected using surface-tethered sediment traps deployed
148 for 1 to 3 days (Table 2). The sediment traps (ST) were attached to a drifting array with
149 a surface buoy equipped with a GPS satellite transmitter, two surface floats and 12
150 buoyancy balls acting as wave breakers in order to reduce the hydrodynamic stress on

151 traps. The ST array was equipped with two sets of four gimbal mounted collection
152 cylinders positioned at nominal depths of 100-120 m and 300-320 m, respectively. At
153 each depth, three cylinders were filled with an unpoisoned brine solution for
154 biogeochemical analyses and one cylinder with a viscous gel to preserve sinking
155 particles in their original shape. This type of array was deployed a total of 11 times,
156 including 7 deployments at the central station (C91, C98, C99, C114, C128, C136,
157 C140) between 3 and 17 February. One of the cylinders for biogeochemical analyses
158 was used after picking off swimmers under a binocular microscope: i) one half was
159 filtered through a pre-combusted QMA filter to analyse ^{234}Th , POC and PON on the
160 same filter as recommended by Buesseler et al. (2006); ii) the other half split was
161 filtered using a pre-combusted GF/F filter to analyse POC and PON. The total POC and
162 PON fluxes collected with the ST were determined as the average of the fluxes obtained
163 from samples i) and ii). Additionally, four Challenger *in-situ* pumps (ISP) were
164 deployed at 100, 150, 300 and 400 m at two stations (C91 and 139) to collect particles
165 using 53- μm pore size nylon mesh screens (Table 2). Particles were subsequently rinsed
166 with filtered seawater and re-filtered through a pre-combusted QMA filter to analyse
167 ^{234}Th , POC and PON after removing swimmers from the filters. The activity of ^{234}Th in
168 particles was first measured on board and re-measured at the home laboratory 10
169 months later as described for the water samples. POC and PON were determined with
170 an EuroVector Elemental Analyser, pre-treating the filters with diluted HCl (Knap et al.,
171 1996). The samples were corrected for POC and PON blanks (1.38 and 0.20 μmol ,
172 respectively), which on average represented about 2% of the POC and PON
173 measurements.

174 2.4. Pigments in sediment traps

175 One fifth of a ST cylinder dedicated to biogeochemical analyses was used to
176 analyse pigments after picking off swimmers. Diluted ST samples were filtered through
177 GF/F filters, under low-vacuum pressure (below 20 kPa). Filtered samples were then
178 immediately shock-frozen in liquid nitrogen and stored at -80°C until further analyses
179 by high performance liquid chromatography (HPLC) at the Alfred-Wegener-Institute in
180 Bremerhaven, Germany. Pigments from ST samples were analysed based on the HPLC
181 method of Barlow et al. (1997) as detailed in Cheah et al. (this issue). The samples were
182 measured using a Waters 600 (Waters, USA) controller combined with a Waters 2998
183 photodiode array detector, and a Water 717plus auto sampler. 100 µL of canthaxanthin
184 was added to each sample as internal standard. Pigments were identified and quantified
185 using the EMPOWER software provided by Waters. Three pigment-based
186 phytoplankton size classes (micro-, nano-, and picophytoplankton) were estimated
187 following the method of Uitz et al. (2009), which has been tested for the Southern
188 Ocean waters. Microphytoplankton corresponded to phytoplankton with size >20 µm,
189 nanophytoplankton between 2-20 µm, and picophytoplankton between 0.2-2 µm.

190 *2.5. Chl-a and POC in the water column*

191 Seawater samples for Chl-a (Chl-a_{sw}) and POC determination were obtained
192 from Niskin bottles attached to the CTD rosette from 5 to 6 depths in the upper 100 m at
193 33 stations. For Chl-a_{sw} analysis, samples were filtered onto GF/F filters at pressure
194 below 20 kPa. Filters were immediately transferred to centrifuge tubes with 10 ml 90%
195 acetone and 1 cm³ of glass beads. The tubes were sealed and stored at -20°C for at least
196 30 min. Chl-a was extracted by placing the centrifuge tubes in a grinder for 3 minutes
197 followed by centrifugation at 0°C. The supernatant was poured in quartz tubes and
198 measured for Chl-a content in a Turner 10-AU fluorometer. The fluorometer was

199 calibrated at the beginning and at the end of the expedition. Chl-a content was
200 calculated using the equation given in Knap et al. (1996) using average parameter
201 values from both calibrations. For POC analysis, samples were filtered onto pre-
202 combusted GF/F filters at pressure not exceeding 20 kPa. Filters were immediately
203 transferred to pre-combusted glass petri dishes and dried overnight at 50°C. Dried filters
204 were stored at -20°C until analysis at the home laboratory using an EuroVector
205 Elemental Analyser. The samples were corrected for POC blanks and the uncertainty of
206 the POC measurements was 1.9% based on three reference standards.

207 *2.6. Satellite Chl-a concentration*

208 Satellite Chl-a concentrations were taken from the merged daily OC-CCI Chl-a
209 data (ESACCI-OC-L3S product, ~4 km, version-2, <http://www.oceancolour.org>) and
210 averaged over the time period of the study sampling. The OC-CCI data product
211 combines the Medium Resolution Imaging Spectrometer (MERIS) on the Envisat
212 satellite, the Moderate Resolution Imaging Spectrometer (MODIS) on the Aqua satellite
213 and the Sea-viewing Wide Field-of-view Sensor (SeaWiFS) on the OrbView-2 satellite
214 to one Chl-a product. This data product improves coverage of satellite Chl-a data in
215 polar regions by a factor of two to three as compared to using the products of one single
216 sensor. More details on the processing steps can be found in the product user guide
217 (OC-CCI, 2015).

218 *3. Results*

219 *3.1. Study area*

220 Detailed hydrographic information can be found in Strass et al. and Leach et al.
221 (this issue). The study area was placed between the APF and the SPF within the ACC

222 and around a central station located at 51.21°S 12.67°W. The main water masses
223 identified were: Winter Water (WW, minimum potential temperature, $\theta_{\min} = 1.1-1.9^{\circ}\text{C}$,
224 100-200 m), Upper Circumpolar Deep Water (UCDW, $\theta_{\max} = 2.1-2.4^{\circ}\text{C}$, 400-500 m)
225 and Antarctic Bottom Water (AABW, $\theta_{\min} \sim -0.2^{\circ}\text{C}$). The mixed layer depth (MLD)
226 varied from 24 to 98 m (average: 67 ± 18 m, $n = 73$, Strass et al., this issue). At the
227 central station the structure of the upper water column varied over time: i) the MLD
228 ranged from ~ 75 to 100 m, and some profiles did not show a homogeneous surface
229 layer; ii) the location of the WW θ_{\min} fluctuated several tens of meters above and below
230 150 m; iii) many profiles showed temperature inversions below the θ_{\min} depth and
231 fluctuations of salinity and density within the depth range 200-300 m during the second
232 half of the sampling period. Moreover, at the central station currents flowing to the NE
233 were intensified over time with speeds averaging from 2 to 10 cm s^{-1} for the top 500 m
234 (Strass et al., this issue).

235 Satellite-derived Chl-a concentrations during the sampling period were, on
236 average, lower than 1.5 mg m^{-3} (Figure 1), whereas at the peak of the bloom, during the
237 first half of January, the Chl-a concentrations were $\sim 3 \text{ mg m}^{-3}$ (Hoppe et al., this issue).
238 The euphotic zone depth ranged between 36 and 66 m (average: 45 ± 7 m, $n = 40$,
239 Cheah et al., this issue). Nutrient concentrations and deficits are described in Hoppe et
240 al. (this issue). On average, macronutrient concentrations within the euphotic zone were
241 $\sim 21 \text{ mmol NO}_3 \text{ m}^{-3}$, $\sim 1.4 \text{ mmol PO}_4 \text{ m}^{-3}$ and $\sim 6.6 \text{ mmol Si(OH)}_4 \text{ m}^{-3}$. High iron
242 concentrations that allowed the development of the massive bloom likely reached the
243 study site via advection through the ACC (L.M. Laglera pers. comm.). Concentrations
244 of dissolved iron ($< 0.2 \mu\text{m}$) were on average $0.12 \pm 0.03 \text{ nM}$ within the top 100 m of the
245 water column (Hoppe et al., this issue), suggesting that dissolved iron was already
246 depleted by phytoplankton activity during our sampling.

247 3.2. ²³⁴Th activity profiles and fluxes

248 The vertical profiles of ²³⁴Th and ²³⁸U activity in the water column are shown in
249 Figure 2. Significant deficits of ²³⁴Th with respect to ²³⁸U were present in surface waters
250 down to 100-170 m at all stations, with ²³⁴Th/²³⁸U ratios averaging 0.67 ± 0.11
251 throughout the upper 100 m. In most cases, the base of these deficits matched well with
252 the base of the Primary Production Zone (PPZ, Figure 2), defined as the depth at which
253 fluorescence reaches 10% of its maximum value (Owens, 2013). The PPZ extended, on
254 average, down to 117 ± 12 m, and the NPP at 100 m (deepest depth for NPP
255 determination) ranged from 0.09 to 0.44 mmol C m⁻³ d⁻¹, confirming that primary
256 production was occurring at least until 100 m (C. Hoppe pers. comm.). Below the PPZ,
257 significant excess of ²³⁴Th (²³⁴Th/²³⁸U ratio >1.1) was detected at stations C91 and 106
258 at one single depth (500 m). Significant ²³⁴Th deficits (²³⁴Th/²³⁸U ratio <0.9) below the
259 PPZ were also found at single depths at stations 93, C128, 137 and 139 (150, 750, 500
260 and 400 m, respectively). The central station showed a larger ²³⁴Th deficit along the top
261 100 m during the second and third visits (C128 and C140: $(90 \pm 6) \cdot 10^3$ and $(86 \pm 6) \cdot 10^3$
262 dpm m⁻², respectively) compared to the first sampling (C91: $(71 \pm 6) \cdot 10^3$ dpm m⁻²).

263 ²³⁴Th fluxes (F_{Th} , in dpm m⁻² d⁻¹) are attributed to scavenging of ²³⁴Th onto
264 particles sinking out of surface waters. Here, we estimated these fluxes using two
265 methods: i) from seawater samples ($F_{Th,SW}$), and ii) directly from sediment traps ($F_{Th,ST}$).
266 The $F_{Th,SW}$ were calculated using a steady state (SS) model, neglecting advective and
267 diffusive fluxes (Buesseler et al., 1992):

268
$$F_{Th} = \lambda_{Th}(A_U - A_{Th}) \quad (1)$$

269 where λ_{Th} is the decay constant of ^{234}Th (0.029 d^{-1}) and $(A_U - A_{Th})$ is the integrated ^{234}Th
 270 deficit in the upper water column (dpm m^{-2}). We obtained the integrated ^{234}Th deficit
 271 with rectangular integration for two different horizon depths (100 and 300 m), allowing
 272 comparison with the ^{234}Th fluxes obtained directly from ST (Table 3).

273 Additionally, since the central station was sampled several times and the activity
 274 of ^{234}Th may have changed over time as a consequence of the sinking of the
 275 phytoplankton bloom (e.g. Savoye et al., 2006), ^{234}Th fluxes were also calculated using
 276 a non-steady state (NSS) model at 100 and 300 m (Table 4). This model assumes that
 277 the same water masses are sampled at all visits and ignores advective and diffusive
 278 fluxes of ^{234}Th (Savoye et al., 2006):

$$279 \quad F_{Th} = \lambda_{Th} \left[\frac{A_U(1-e^{-\lambda\Delta t}) + A_{Th1}e^{-\lambda\Delta t} - A_{Th2}}{1-e^{-\lambda\Delta t}} \right] \quad (2)$$

280 where A_U is the activity of ^{238}U , Δt is the time interval between two visits of a single
 281 station and A_{Th1} and A_{Th2} are the activities of ^{234}Th at the first and second visits,
 282 respectively.

283 $F_{Th,SW}$ and $F_{Th,ST}$ estimates at 100 and 300 m are presented in Tables 3-4 and
 284 Figure 3. Though some traps were placed 20 m deeper than the seawater sampling, we
 285 will still use 100 and 300 m to refer to 100 or 120 m and 300 or 320 m, respectively.

286 The SS estimates of $F_{Th,SW}$ at 100 m were relatively homogeneous over the
 287 entire study area and not significantly different from those at the base of the ^{234}Th
 288 deficit (~ 120 m, Wilcoxon test, $p > 0.05$), ranging from 1960 ± 210 to 3100 ± 180 dpm
 289 $\text{m}^{-2}\text{ d}^{-1}$ (average: 2390 ± 340 $\text{dpm m}^{-2}\text{ d}^{-1}$, $n = 14$). Fluxes at 300 m had greater
 290 uncertainties than those at 100 m, and ranged from 1490 ± 710 to 3870 ± 700 dpm m^{-2}

291 d^{-1} (average: $2650 \pm 610 \text{ dpm m}^{-2} d^{-1}$, $n = 14$). The $F_{Th,SW}$ over the study area showed no
 292 significant differences between 100 and 300 m (t-test, $p > 0.05$) (Figure 3). Focusing on
 293 the central station, $F_{Th,SW}$ increased from C91 to C128 at both 100 and 300 m depth,
 294 reaching ~ 3000 and $4000 \text{ dpm m}^{-2} d^{-1}$, respectively. The $F_{Th,SW}$ at station C140 were not
 295 significantly different from those at station C128 (Table 3). According to the NSS
 296 model, the $F_{Th,SW}$ decreased substantially from C91-C128 to C128-C140, especially at
 297 300 m (Table 4).

298 $F_{Th,ST}$ at 100 m ranged from 1050 ± 90 to $4000 \pm 320 \text{ dpm m}^{-2} d^{-1}$ (average: 1720
 299 $\pm 890 \text{ dpm m}^{-2} d^{-1}$, $n = 10$) (Table 3). A flux reduction of about 40% between 100 to
 300 300 m was found at most stations, except at C128 and C140, where no significant
 301 differences were observed. At the central station, $F_{Th,ST}$ at 100 m were relatively
 302 constant from C91 to C140, with an average of $1330 \pm 170 \text{ dpm m}^{-2} d^{-1}$ ($n = 6$). $F_{Th,ST}$ at
 303 300 m were also similar during the entire sampling time, averaging $900 \pm 200 \text{ dpm m}^{-2}$
 304 d^{-1} ($n = 6$).

305 Globally, the ^{234}Th fluxes estimated using the SS approach and ST were
 306 comparable at 100 m (Wilcoxon test, $p > 0.05$, $F_{Th,SW}/F_{Th,ST} = 1.7 \pm 0.9$, $n = 7$), whereas
 307 at 300 m the $F_{Th,SW}$ were higher than the $F_{Th,ST}$ estimates by an average factor of $2.6 \pm$
 308 1.7 ($n = 7$).

309 3.2.1. Impact of physical transport processes on the $F_{Th,SW}$ estimates

310 The physical transport processes can be parameterised as (e.g. Savoye et al.,
 311 2006):

$$312 \quad V = \pm u \frac{\partial A_{Th}}{\partial x} \pm v \frac{\partial A_{Th}}{\partial y} \pm w \frac{\partial A_{Th}}{\partial z} \pm K_x \frac{\partial^2 A_{Th}}{\partial x^2} \pm K_y \frac{\partial^2 A_{Th}}{\partial y^2} \pm K_z \frac{\partial^2 A_{Th}}{\partial z^2} \quad (3)$$

313 where advective and diffusive components are included along the x, y and z directions.

314 Velocities are denoted by u, v and w, $\frac{\partial A_{Th}}{\partial x}$, $\frac{\partial A_{Th}}{\partial y}$ and $\frac{\partial A_{Th}}{\partial z}$ are the ^{234}Th activity

315 gradients and K_x , K_y and K_z are the diffusion coefficients. Vertical advection needs to

316 be considered in areas of established upwelling, while horizontal advection should be

317 taken into account in ocean margins, where this process likely plays a relevant role

318 (Savoie et al., 2006). During our survey the vertical advection was presumably

319 negligible, while horizontal advection was significant with mean currents of about 7 cm

320 s^{-1} for the top 100 and 300 m (V. Strass pers. comm.). Buesseler et al. (1994) showed

321 that advection is dominant over diffusion in the horizontal transport of ^{234}Th in open

322 waters. Therefore, we focused our attention on the horizontal advection in order to have

323 a first estimate of the importance of the physical transport in the present study or, in

324 other words, to test the accuracy of our ^{234}Th export estimates. We addressed this issue

325 considering: i) the mean u and v velocities over the area 50.67-51.67°S and 11.92-

326 13.50°W for the top 100 and 300 m: u = 5.6 cm s^{-1} , v = 4.3 cm s^{-1} and u = 5.7 cm s^{-1} , v =

327 3.9 cm s^{-1} , respectively (V. Strass pers. comm.); ii) the ^{234}Th activity gradients for the

328 top 100 and 300 m (excluding station 86): $\frac{\partial A_{Th}}{\partial x} = (6.3 \pm 7.9) \cdot 10^{-2}$ dpm m^{-3} , $\frac{\partial A_{Th}}{\partial y} =$

329 $(7.4 \pm 6.1) \cdot 10^{-2}$ dpm m^{-3} and $\frac{\partial A_{Th}}{\partial x} = (2.4 \pm 3.0) \cdot 10^{-1}$ dpm m^{-3} , $\frac{\partial A_{Th}}{\partial y} = (0.9 \pm 2.2) \cdot 10^{-1}$

330 dpm m^{-3} , respectively. In this way, the negligence of horizontal advection would lead to

331 an error on the $F_{Th,SW}$ estimates of 580 ± 440 dpm $m^{-2} d^{-1}$ at 100 m and 1500 ± 1700

332 dpm $m^{-2} d^{-1}$ at 300 m. Thus, the physical processes would have a smaller impact on the

333 $F_{Th,SW}$ estimates at 100 m (~25%) than at 300 m (~50%) at the investigated stations due

334 to the smaller spatial variability of ^{234}Th activities in the upper 100 m.

335 Additionally, we have tested the influence of vertical diffusivity on ^{234}Th export

336 using an intermediate K_z value of $10^{-4} \text{ m}^2 \text{ s}^{-1}$ at 100 m (Strass et al. and Leach et al., this
337 issue), obtaining a maximum contribution of this process of 30-180 $\text{dpm m}^{-2} \text{ d}^{-1}$. Thus,
338 the contribution of the vertical diffusivity to the $F_{\text{Th,SW}}$ estimates at 100 m must have
339 been very small, generally lower than the uncertainties associated with the estimates. It
340 was not possible to reproduce this exercise at 300 m due to the low ^{234}Th resolution
341 around that depth.

342 3.2.2. *SS vs NSS models at the central station*

343 At the central station we determined the ^{234}Th export with both SS and NSS
344 models. However, changes in the upper water column properties occurred during the
345 sampling period, especially evident when station C128 was occupied (see section 3.1
346 and Strass et al. this issue), indicating that different water masses with particular
347 scavenging histories were likely sampled. Furthermore, we have quantified an
348 appreciable impact of advection on the $F_{\text{Th,SW}}$ estimates, especially at 300 m (see section
349 3.2.1). This has led us to discard the use of the NSS approach since it can produce large
350 errors when spatial variability is misinterpreted as temporal variability, improving ^{234}Th
351 export estimates only if sampling is conducted in a Lagrangian framework (Resplandy
352 et al., 2012). Additionally, the results obtained from the NSS model (Table 4) are not
353 consistent with results from the ST, which tend to be more constant. Thus, we consider
354 that the SS model gives the best estimate of ^{234}Th export in this study.

355 3.3. *POC/ ^{234}Th and PON/ ^{234}Th ratios in particles*

356 POC/ ^{234}Th and PON/ ^{234}Th (C/Th and N/Th) ratios in particles from ST (C/Th_{ST}
357 and N/Th_{ST}) and in particles $>53 \mu\text{m}$ collected using ISP (C/Th_{ISP} and N/Th_{ISP}), usually
358 considered to be sinking particles (e.g. Buesseler et al., 2006), are presented in Table 5.

359 C/Th_{ST} and N/Th_{ST} ratios at 100 m averaged 14 ± 3 and $1.7 \pm 0.4 \mu\text{mol dpm}^{-1}$ (n
360 = 10), respectively, and were about 30% lower at 300 m depth (10 ± 3 and 1.1 ± 0.3
361 $\mu\text{mol dpm}^{-1}$, n = 9, respectively). C/Th_{ISP} and N/Th_{ISP} ratios also decreased with depth
362 between 100-150 and 300-400 m, by about 60%. At the central station, C/Th_{ST} and
363 N/Th_{ST} ratios at 100 m decreased by 30% from the beginning until the end of the study,
364 but no significant change was observed at 300 m.

365 ST and ISP ratios (C/Th and N/Th) were in reasonable agreement (within a
366 factor of 1.4), except at station 139 at 100 m depth, where the ISP ratios were about 3-4
367 times higher than the ST ratios. The molar C/N ratio was 8 ± 1 for ST ratios (n = 19)
368 and 7 ± 1 for ISP ratios (n = 8).

369 3.4. POC and PON fluxes

370 We used three methods to estimate POC and PON fluxes (F_C and F_N ,
371 respectively) (Table 5): i) measured directly with the ST (ST method: $F_{C,ST}$ and $F_{N,ST}$),
372 ii) combining the $F_{Th,SW}$ and the ST ratios (SWST method: $F_{C,SWST}$ and $F_{N,SWST}$), and iii)
373 combining the $F_{Th,SW}$ and the ISP ratios (SWISP method: $F_{C,SWISP}$ and $F_{N,SWISP}$). For ii)
374 and iii) we used the SS estimates of F_{Th} and the C/Th (or N/Th) ratios in sinking
375 particles, as:

$$376 F_C = F_{Th}(C/Th) \quad (4)$$

377 The average $F_{C,ST}$ and $F_{N,ST}$ at 100 m were 19 ± 9 and $2 \pm 1 \text{ mmol m}^{-2} \text{ d}^{-1}$ (n =
378 11), respectively, with maxima at station 87. $F_{C,ST}$ and $F_{N,ST}$ at 300 m averaged 9 ± 3 and
379 $1.0 \pm 0.3 \text{ mmol m}^{-2} \text{ d}^{-1}$ (n = 11), respectively, about 60% less than the fluxes at 100 m
380 except at stations C128, C136 and C140. During the occupation of the central station,

381 $F_{C,ST}$ and $F_{N,ST}$ at 100 m decreased by 25% over the study period, while they increased
382 by 30% and 50%, respectively, at 300 m.

383 $F_{C,SWST}$ and $F_{N,SWST}$ at 100 m averaged 30 ± 10 and 4 ± 1 mmol m⁻² d⁻¹ (n = 7),
384 respectively. Fluxes at 300 m averaged 20 ± 10 and 2 ± 1 mmol m⁻² d⁻¹ (n = 6),
385 respectively, about 40% less than the fluxes at 100 m except at stations C128 and C140.
386 $F_{C,SWST}$ and $F_{N,SWST}$ estimates at the central station showed changes <5% with time at
387 100 m, whereas the fluxes at 300 m were more variable, with a maximum $F_{C,SWST}$ of 50
388 ± 10 mmol m⁻² d⁻¹ at C128.

389 The SWISP method was used at the two stations where ISP were deployed.
390 $F_{C,SWST}$ and $F_{N,SWST}$ at station 139 were higher than at C91 by a factor of about 2, at both
391 100-150 m and 300-400 m. Fluxes at 300 m were lower than those at 100 m by 40%
392 (C91) and 70% (139).

393 Overall, the F_C and F_N estimated by using SW samples ($F_{C,SWST}$, $F_{N,SWST}$ and
394 $F_{C,SWISP}$, $F_{N,SWISP}$) were greater than those measured with ST by a factor of 2.0 ± 1.0 and
395 2.3 ± 1.3 (n = 17), respectively, at 100 m, and 2.4 ± 1.0 and 2.5 ± 1.0 (n = 17),
396 respectively, at 300 m.

397 3.5. *Chl-a and POC in the water column*

398 The Chl-a_{sw} inventories down to 100 m were, in most instances, larger than 100
399 mg m⁻² (Figure 4A), with highest inventories (>180 mg m⁻²) at the SE quadrant of the
400 study site, while stations 87 and 139, located to the NW, presented the lowest values
401 (~50 mg m⁻²). At the central station the Chl-a_{sw} inventories ranged from 130 to 200 mg
402 m⁻², decreasing by about 30% from the beginning until the end of sampling, with
403 fluctuations over time. The POC inventories down to 100 m ranged from $10 \cdot 10^3$ to

404 $26 \cdot 10^3 \text{ mg m}^{-2}$, showing a strong positive correlation with Chl- a_{SW} ($p < 0.001$; Pearson
405 correlation coefficient, $r = 0.84$; $n = 20$; Figure 4A).

406 *3.6. Pigments and POC in sediment traps*

407 In contrast to the water column, there was no significant correlation between the
408 fluxes of Chl-a measured with ST (Chl- a_{ST}) and $F_{\text{C,ST}}$, neither at 100 m ($p > 0.05$;
409 Spearman correlation coefficient, $\rho = -0.03$; $n = 11$) nor 300 m ($p > 0.05$, $\rho = 0.26$, $n =$
410 11, Figure 4B). Both Chl- a_{ST} fluxes and $F_{\text{C,ST}}$ decreased with depth (except Chl- a_{ST} flux
411 at station C114), although this decrease was stronger for POC at most stations (Figure 5,
412 Table 5).

413 Maximum Chl- a_{ST} flux was recorded at station C98 ($0.79 \text{ mg m}^{-2} \text{ d}^{-1}$ at 100 m),
414 whereas minimum Chl- a_{ST} flux was observed at station 86 ($0.11 \text{ mg m}^{-2} \text{ d}^{-1}$ at 300 m).
415 Fluxes of total pheopigments (TPheo, i.e. sum of pheophytin-a, pyropheophytin-a,
416 pheophorbide-a, and pyropheophorbide-a pigments) were generally higher at 100 m
417 than at 300 m except at stations C91 and C114 (Figure 5A). Maximum TPheo fluxes
418 were recorded at station 87 (2.67 and $1.27 \text{ mg m}^{-2} \text{ d}^{-1}$ at 100 and 300 m, respectively).
419 Ratios of TPheo/Chl- a_{ST} were generally higher at 100 m than at 300 m except at station
420 C91. TPheo/Chl- a_{ST} ratios above 1 were recorded at most of the investigated stations
421 except at 137, 139, C98, C136 and C140 (Figure 5A). Among the marker pigments,
422 fluxes of fucoxanthin were much higher than other pigments (Figure 5B) and the
423 phytoplankton size classification showed that more than 80% of the phytoplankton were
424 microphytoplankton (Figure 5C).

425 *4. Discussion*

426 *4.1. ^{234}Th fluxes*

427 *4.1.1. Seawater approach vs sediment traps*

428 In this study, the ^{234}Th fluxes were estimated using the seawater approach and
429 sediment traps. The SW approach integrates about 5 weeks before sampling (^{234}Th
430 mean life = 35 days), while ST provide information at the sampling time. In general, as
431 shown in section 3.2, the ^{234}Th fluxes based on these techniques were not significantly
432 different at 100 m. This agreement indicates a sustained export rate in the weeks prior to
433 and during the sampling period, suggesting export fluxes at steady state.

434 At 300 m the $F_{\text{Th,ST}}$ were about 3 times lower than the $F_{\text{Th,SW}}$. A review by
435 Buesseler et al. (2007) suggested that ^{234}Th -bearing particles were under-trapped by a
436 factor of about 2 and suggested some processes that could be sources of trap error, such
437 as under-collection of slow settling particles by hydrodynamic bias, particle
438 solubilisation within the trap before sample processing, and “swimmer” related
439 artefacts. Haskell et al. (2013) reported that ST collected about $\leq 20\%$ of the $F_{\text{Th,SW}}$ in
440 two consecutive years and proposed that the main cause of this mismatch was the
441 under-trapping of small, ^{234}Th -enriched slow sinking particles. Additionally, the active
442 transport of surface-derived particles to hundreds of meters down via zooplankton
443 migration (e.g. Angel, 1989) is another factor that may lead to underestimation of the
444 particle export based on ST. Here, we tried to minimise the sources of error by using
445 two surface floats and 12 buoyancy balls as wave breakers, while the cylinders of the
446 ST were gimbal mounted. In addition, samples were processed within 24 h after
447 collection to decrease the impact of solubilisation and zooplankton swimmers on
448 material, which were picked under a binocular microscope.

449 Buesseler et al. (2007) also suggested that the mismatch between the SW
450 approach and ST could be related to the use of inaccurate assumptions in the ^{234}Th

451 model. The exclusion of physical processes in our study could introduce a significant
452 error on the $F_{Th,SW}$ estimates at 300 m (~50%, see section 3.2.1). Besides, in a late stage
453 of a bloom, the SS assumption may lead to an overestimation of the $F_{Th,SW}$ if higher
454 export occurred before sampling (within the ^{234}Th time scale). However, similar $F_{Th,SW}$
455 were found over the entire study area at both 100 and 300 m (Figure 3), irrespectively of
456 phytoplankton biomass (see section 3.5). This indicates that ^{234}Th fluxes probably did
457 not change significantly during the weeks before our arrival and, therefore, the SS
458 model gives reasonable estimates of ^{234}Th export flux.

459 *4.1.2. Results and comparison with other studies*

460 On average, ^{234}Th fluxes at 100 and 300 m were 2100 ± 700 and 2000 ± 1000
461 $dpm\ m^{-2}\ d^{-1}$, respectively. Hence, the fluxes were high and quite similar over the entire
462 study area, despite the differences observed in Chl-*asw* inventories (see section 3.5) and
463 production rates (Hoppe et al., this issue). However, satellite images show widespread
464 high Chl-*a* concentrations in January 2012 (data not shown). Thus, stations with low
465 phytoplankton biomass and production rates (e.g. stations 139 and C136) but high F_{Th}
466 evidenced a temporal decoupling between production and export. Buesseler et al. (2003,
467 2001a), Rutgers van der Loeff et al. (2011) and Henson et al. (2015) showed that export
468 lags production by up to 1-2 months in the Southern Ocean, which is a time frame
469 compatible with the present study.

470 Our F_{Th} estimates at 100 m are within the range of values for blooms during
471 their peak and/or decline occurring in the Southern Ocean, either in land remote areas
472 (Rutgers van der Loeff et al., 1997) or near islands (Morris et al., 2007; Savoye et al.,
473 2008). In contrast, our estimates are higher than the F_{Th} of $\leq 1000\ dpm\ m^{-2}\ d^{-1}$ reported
474 during the onset of a bloom by Planchon et al. (2015), and during the artificial iron

475 fertilisation experiments SOIREE (Charette and Buesseler, 2000), EisenEx (Rutgers van
476 der Loeff and Vöge, 2001) and LOHAFEX (Martin et al., 2013). However, the SOFeX-
477 South experiment revealed an increase of particle fluxes after fertilisation, which
478 resulted in F_{Th} of $\sim 1800 \text{ dpm m}^{-2} \text{ d}^{-1}$ at the MLD (Buesseler et al., 2005, 2004). The
479 variability within the plankton community structure and different sampling time
480 strategies among these experiments most likely played a crucial role in the variety of
481 results obtained (Boyd et al., 2007; Buesseler et al., 2004).

482 A release of ^{234}Th from sinking particles, either via remineralisation or
483 disaggregation, was evidenced by a 40% decrease of the $F_{Th,ST}$ between 100 and 300 m
484 at the majority of the stations. However, the seawater approach did not show excesses
485 of ^{234}Th in the depth range 100-300 m at any station. As a result, ^{234}Th flux attenuation
486 from 100 to 300 m estimated using the two methods was different at 3 out of 7 stations,
487 taking into account the large uncertainties of the $F_{Th,SW}$ estimates at 300 m (16-48%,
488 Table 3). The potential impact of advection on the $F_{Th,SW}$ estimates at 300 m (see
489 section 3.2.1) together with a low resolution of the seawater sampling between 100-300
490 m (2-3 samples) likely explain most of the disagreement between the ST and SW
491 approaches on this issue. Remineralisation and/or particle break-up was also indicated
492 in previous studies in the Southern Ocean (Buesseler et al., 2005; Planchon et al., 2013;
493 Rutgers van der Loeff et al., 2011; Savoye et al., 2004; Usbeck et al., 2002). In our
494 study, $F_{Th,ST}$ were attenuated between 100 and 300 m at the first 4 visits of the central
495 station, but not at stations C128 and C140 (Table 3), indicating that attenuation of $F_{Th,ST}$
496 in this depth range became negligible at the end of the sampling period. This
497 observation is similar to one of the major findings of the SOFeX-South experiment: the
498 attenuation of F_{Th} with depth disappeared as the bloom progressed (Buesseler et al.,
499 2005). This was attributed to changes in the nature of the sinking particles (less labile or

500 more rapidly sinking) and/or less efficient biological and physical processes responsible
501 for particle break-up and consumption below the bloom.

502 4.2. $POC/^{234}Th$ and $PON/^{234}Th$ ratios in particles

503 C/Th and N/Th ratios were $>10 \mu\text{mol C dpm}^{-1}$ (average: $15 \pm 5 \mu\text{mol C dpm}^{-1}$)
504 and $>1 \mu\text{mol N dpm}^{-1}$ (average: $2 \pm 1 \mu\text{mol N dpm}^{-1}$), respectively, in all samples
505 collected either with ST or ISP ($>53 \mu\text{m}$) at 100 m. Overall, these values were higher
506 than those reported by most studies conducted in the Southern Ocean (Buesseler et al.,
507 2005, 2001a; Coppola et al., 2005; Jacquet et al., 2011; Martin et al., 2013; Morris et
508 al., 2007; Planchon et al., 2013; Rutgers van der Loeff et al., 2011, 1997; Savoye et al.,
509 2008), but similar to others (Cochran et al., 2000; Puigcorb  et al., this issue; Rutgers
510 van der Loeff et al., 2002; Smetacek et al., 2012). Multiple factors may have played a
511 role in shaping the C/Th and N/Th ratios, such as particle source, sinking velocity, Th
512 speciation and remineralisation of C, N and ^{234}Th associated with sinking particles
513 (Buesseler et al., 2006). Buesseler (1998) and Buesseler et al. (2006) hypothesised that
514 high C/Th ratios encountered in productive regimes and high latitude areas may be
515 related to the dominance of sinking of large cells, such as diatoms. This was explained
516 by the fact that diatoms have a high volume relative to surface area (V:SA), and hence
517 high C/Th ratios because C varies as a function of volume, whereas Th adsorption
518 varies as a function of surface area. In our study, the dominance of fucoxanthin, a
519 marker pigment for diatoms, and microphytoplankton in trap material (Figure 5B,C)
520 revealed that diatoms were the major phytoplankton group contributing to vertical
521 fluxes. Additionally, according to microscopic analyses (C. Klaas pers. comm.), the
522 needle-shaped *Pseudo-nitzschia lineola* and the large centric diatom *Dactyliosolen*
523 *antarcticus* dominated the bloom, with significant contributions from *Fragilariopsis*

524 *kerguelensis* as well as other *Pseudo-nitzschia*, *Rhizosolenia* and *Chaetoceros* species.
525 Thus, diatoms with a high V:SA, including *Dactyliosolen antarcticus* and *Chaetoceros*
526 spp. played a relevant role in the bloom, which could be responsible for the high ratios
527 observed.

528 C/Th and N/Th ratios at 300 m were, on average, about 30% lower than at 100
529 m. The reduction of C/Th ratios with depth is commonly observed (Buesseler et al.,
530 2006). It is likely due to the production of ^{234}Th in the entire water column leading to
531 additional adsorption of ^{234}Th onto particles at depth, which compensates its decay, and
532 a preferential remineralisation of POC with respect to ^{234}Th (Rutgers van der Loeff et
533 al., 2002). The same reasons would apply for PON.

534 The C/N ratios averaged 8 ± 1 and 7 ± 1 for ST and ISP samples, respectively,
535 and were within the range of values previously reported during bloom events (Martin et
536 al., 2013; Morris et al., 2007; Savoye et al., 2008), with a diverse composition of the
537 dominant particle type (diatoms: Salter et al., 2007; faecal material: Ebersbach and
538 Trull, 2008, and Ebersbach et al., 2014).

539 4.3. POC and PON fluxes at 100 m

540 POC and PON export fluxes at 100 m averaged $26 \pm 15 \text{ mmol C m}^{-2} \text{ d}^{-1}$ (range:
541 $11\text{-}67 \text{ mmol C m}^{-2} \text{ d}^{-1}$) and $4 \pm 2 \text{ mmol N m}^{-2} \text{ d}^{-1}$ (range: $1.6\text{-}12 \text{ mmol N m}^{-2} \text{ d}^{-1}$, Table
542 5), respectively. Our F_C are high in relation to the average fluxes compiled by Le
543 Moigne et al. (2013) and Maiti et al. (2013). Le Moigne et al. (2013) reported an
544 average F_C of $9 \pm 13 \text{ mmol C m}^{-2} \text{ d}^{-1}$ ($n = 726$; range: $<0\text{-}125 \text{ mmol C m}^{-2} \text{ d}^{-1}$) for the
545 global ocean, and $13 \pm 14 \text{ mmol C m}^{-2} \text{ d}^{-1}$ ($n = 196$; range: $0\text{-}91 \text{ mmol C m}^{-2} \text{ d}^{-1}$) for the

546 Southern Ocean. Maiti et al. (2013) reported a range of F_C from 1 to 50 $\text{mmol C m}^{-2} \text{d}^{-1}$
547 for the Southern Ocean, averaging $11 \pm 8 \text{ mmol C m}^{-2} \text{d}^{-1}$ ($n = 140$).

548 Considering other natural blooms observed in the Southern Ocean, our estimates
549 of F_C are comparable to those reported in KEOPS (average: $23 \pm 4 \text{ mmol C m}^{-2} \text{d}^{-1}$,
550 Savoye et al., 2008), the JGOFS Southern Ocean expedition (range: 20-39 mmol C m^{-2}
551 d^{-1} , Rutgers van der Loeff et al., 1997), and the CROZEX project (average: 14 ± 4
552 $\text{mmol C m}^{-2} \text{d}^{-1}$, Morris et al., 2007). Our estimates of F_N are also similar to the results
553 from KEOPS and CROZEX (Morris et al., 2007; Savoye et al., 2008). In contrast, our
554 F_C are higher than those reported in KEOPS II (average: $7 \pm 2 \text{ mmol C m}^{-2} \text{d}^{-1}$,
555 Planchon et al., 2015) for a bloom during its growing stage. On the other hand, the F_C
556 determined during SOIREE (Charette and Buesseler, 2000), SOFeX-South (Buesseler et
557 al., 2005) and LOHAFEX (Martin et al., 2013) were clearly lower (average $\leq 8 \text{ mmol C}$
558 $\text{m}^{-2} \text{d}^{-1}$) than in this study. In the case of LOHAFEX, it was explained by silica
559 limitation that prevented diatom growth. The low F_C measured during SOIREE and
560 SOFeX-South were likely due to a limited sampling period. However, the artificially
561 induced bloom during EIFEX (Smetacek et al., 2012) led to an extraordinarily high F_C
562 ($\sim 80 \text{ mmol C m}^{-2} \text{d}^{-1}$) with a high export efficiency. Thus, the high export fluxes found
563 in our study were comparable with other studies of the later stages of natural and iron
564 fertilised blooms in the Southern Ocean that were not silica limited.

565 *4.4. Export efficiency*

566 The export efficiency is a useful parameter to illustrate the strength of the
567 biological pump, even though production and export operate at distinct time scales
568 (Buesseler and Boyd, 2009). It should also be noted the temporal mismatch between the
569 measurements of production and export: 24 h incubation for NPP (Hoppe et al., this

570 issue), 15-72 h deployment for ST and a several weeks of integration for the SW
571 approach. We assessed the export efficiency by dividing the F_C at 100 m, either from
572 the SW methods or ST, by the NPP integrated down to 100 m (Hoppe et al., this issue).

573 The export efficiency within the bloom averaged $23 \pm 24\%$ (range: 7-100%;
574 Table 6, Figure 6A), taking into account all the techniques used to estimate the export of
575 POC (ST, SWST, SWISP). The export efficiency was around 10% according to the ST,
576 which is common in the open ocean (Buesseler, 1998), whereas it was around 25%
577 considering the SWST method. Besides these differences, the export efficiency was
578 always $\leq 50\%$ for the ST and SWST methods, contrasting with export efficiencies $>50\%$
579 reported during blooms at high latitudes, mostly characterized by large diatoms
580 (Buesseler, 1998; Smetacek et al., 2012) and a combination of *Phaeocystis* and diatoms
581 (Buesseler et al., 2003; Poulton et al., 2007). Other Southern Ocean studies (Buesseler
582 et al., 2005; Planchon et al., 2015) have also reported export efficiencies of $\sim 10\%$
583 during not yet declining diatom blooms, where low POC export fluxes were measured
584 ($7-8 \text{ mmol C m}^{-2} \text{ d}^{-1}$). In contrast, during our study the bloom was declining, which lead
585 to high downward fluxes. We propose that this relatively low strength of the biological
586 pump was likely related to an active recycling of carbon and nutrients in surface waters.
587 Only for one station (139), which evidenced a temporal decoupling between production
588 and export (lowest Chl- a_{SW} inventories and NPP rates), the export efficiency was $>30\%$
589 according to all techniques. Indeed, we found an inverse relationship between export
590 efficiency and NPP ($p < 0.05$; ST method: $\rho = -0.95$, $n = 8$; SWST method: $\rho = -0.89$, n
591 $= 6$) supporting recent observations (Cavan et al., 2015; Laurenceau-Cornec et al., 2015;
592 Maiti et al., 2013). This relationship could be explained by a combination of temporal
593 decoupling between primary production and export (Henson et al., 2015; Puigcorbé et

594 al., this issue), and other processes such as zooplankton grazing (Cavan et al., 2015),
595 bacterial activity and recycling efficiency (Maiti et al., 2013).

596 *4.5. Transfer efficiency*

597 The upper 300 m of the water column is where intense biological and physical
598 processes transform the size distribution of particles and thus modify the flux of
599 particles to the mesopelagic layer (Guidi et al., 2009). To account for the change of the
600 F_C magnitude within the upper twilight zone, we estimated the transfer efficiency by
601 dividing the F_C at 300 m by those at 100 m.

602 The transfer efficiency averaged $59 \pm 26\%$ (range: 35->100%; Table 6, Figure
603 6B) with a general discrepancy among techniques <30%. Most of the transfer
604 efficiencies were higher than the estimates presented by i) Schlitzer (2002): 40% for the
605 same depth range using an inverse model based on observations of temperature, salinity
606 and nutrient concentrations; and ii) Guidi et al. (2009): <42% when microphytoplankton
607 dominated the phytoplankton community based on a world-ocean analysis. Guidi et al.
608 (2009) showed that maximum values of F_C at 400 m were associated with a dominance
609 of microphytoplankton in the euphotic zone, although the flux experienced a sharp
610 decrease from 100 to 300 m with this community structure. In our study, the transfer
611 efficiency at the central station was lower during the first 4 visits (C91-C114: $42 \pm 6\%$)
612 than during the last 3 (C128-C140: $70 \pm 10\%$) according to the ST results (Table 6).
613 This change coincided with a decrease of the TPheo/Chl- a_{ST} ratio in the trap material
614 from the first group of stations to the second: from 2.5 ± 1.1 to 0.9 ± 0.8 at 100 m and
615 from 3.5 ± 1.7 to 0.3 ± 0.2 at 300 m (Figure 5A). Pheopigments are degraded Chl-a
616 products that are associated with grazing activity and phytoplankton senescence (Wright
617 et al., 2010). Thus, this observation indicates that when the transfer efficiency was

618 higher, the organic material being exported was fresher. Indeed, there is a negative
619 relationship between TPheo/Chl-_{aST} ratios at 100 m and the transfer efficiency of POC
620 from 100 to 300 m derived from ST ($p < 0.01$; $\rho = -0.77$, $n = 10$). This correlation
621 highlights the relevance of the particle composition sinking out from the ocean surface
622 in shaping the efficiency of the biological pump. Buesseler and Boyd (2009) pointed out
623 that very high transfer efficiencies are attributed to direct sinking of algae with low flux
624 attenuation. Here, more than 60% of the Chl-_{aST} and fucoxanthin fluxes at 100 m
625 reached 300 m (Figure 5A,B), which highlights the contribution of diatoms to vertical
626 fluxes in the study area. In particular, the direct sinking of diatoms might be the main
627 explanation for the high transfer efficiencies at stations C136 and C140, since at these
628 locations the high fucoxanthin fluxes at 100 m ($>0.4 \text{ mg m}^{-2} \text{ d}^{-1}$) showed a decrease of
629 only ~10-20% at 300 m, and the TPheo/Chl-_{aST} ratios were <1 (Figure 5A,B). Indeed, a
630 study by Cedhagen et al. (2014) from the same cruise showed that abyssal benthic
631 foraminifera at ~4000 m were feeding on fresh phytodetritus. This further indicates that
632 at least part of the phytoplankton at the surface waters were transported down to the
633 seabed.

634 The lack of relationship between Chl-_{aST} and $F_{C,ST}$ at both 100 and 300 m depth
635 (Figure 4B), however, indicated that reprocessed material or dead phytoplankton also
636 contributed significantly to the export fluxes at most stations. The high fluxes of
637 pheopigments ($>0.6 \text{ mg m}^{-2} \text{ d}^{-1}$) together with TPheo/Chl-_{aST} ratios >1 (Figure 5A) at
638 stations 87, C91, C99 and C114 at 100 and 300 m may be associated with grazing
639 activity and, hence, indicate the contribution of faecal pellets to sinking fluxes.
640 Preliminary results confirmed a significant abundance of faecal pellets in trap material
641 during our survey (Iversen and Klaas, 2013), although this should be validated by
642 quantitative analyses in the future. Specifically, at station C114 the TPheo, Chl-_{aST} and

643 fucoxanthin fluxes increased from 100 to 300 m (Figure 5A,B), which may have been
644 due to zooplankton consumption of diatoms in the euphotic zone and release of faecal
645 pellets between 100 and 300 m. Therefore, production of faecal pellets and zooplankton
646 vertical migration might have modulated the transfer efficiency in this study. Similar
647 conclusions have been reached by Buesseler and Boyd (2009) and Cavan et al. (2015).

648 5. Conclusions

649 We measured downward fluxes of POC and PON during the decline of a vast
650 diatom bloom in a land-remote area of the Atlantic sector of the Southern Ocean. Our
651 main conclusions are:

- 652 1) The simultaneous use of sediment traps and the $^{234}\text{Th}/^{238}\text{U}$ proxy evidenced that
653 ^{234}Th export rates at 100 m were high and constant in the weeks prior to and
654 during the survey.
- 655 2) Stations distributed over an area of 8000 km² showed similar vertical export
656 fluxes in spite of the heterogeneity in phytoplankton standing stocks and
657 productivity, indicating a decoupling between production and export.
- 658 3) The diatom bloom led to high POC export fluxes at 100 m (26 ± 15 mmol C m⁻²
659 d⁻¹), although the export efficiencies were generally low (<20%) in comparison to
660 other diatom blooms at high latitudes. An active recycling of carbon and nutrients
661 in surface waters was likely the reason for the low export efficiencies.
- 662 4) In contrast to the low export efficiencies at 100 m depth, the transfer efficiency of
663 POC between 100 and 300 m was high during the entire study period (~60%),
664 partly driven by the sinking of fresh diatoms. Faecal pellets and the active
665 transport of POC linked to zooplankton vertical migration may have also
666 contributed significantly to vertical fluxes. If we are to understand the efficiency

667 of the biological pump in the Southern Ocean, these processes should be further
668 investigated in the future.

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995 Captions of Figures and Tables

996 Fig. 1: Study area and sampled stations for ^{234}Th , POC and PON analyses. C
997 represents the central station, which was occupied 7 times: C91, C98, C99, C114, C128,
998 C136, C140. See Table 1 for further details regarding sampling dates. The satellite plot
999 represents the mean Chl-a concentration from the OC-CCI Chl-a product version-2
1000 during the sampling period (29 Jan. to 17 Feb. 2012).

1001

1002 Fig. 2: Vertical activity profiles for ^{234}Th (black diamonds) and ^{238}U (dotted line) from
1003 10 to 500-750 m depth. Grey shaded area indicates surface ^{234}Th deficits respect to ^{238}U .
1004 ^{238}U was derived from salinity (Owens et al., 2011). Primary Production Zone (PPZ,
1005 dashed grey line) is defined as the depth at which fluorescence reaches 10% of its
1006 maximum value (Owens, 2013).

1007

1008 Fig. 3: ^{234}Th export fluxes derived from seawater at 100 m (in white) and 300 m (in
1009 grey) assuming steady state conditions. The negligence of horizontal advection would
1010 lead to an error on these estimates of $580 \pm 440 \text{ dpm m}^{-2} \text{ d}^{-1}$ at 100 m and 1500 ± 1700
1011 $\text{dpm m}^{-2} \text{ d}^{-1}$ at 300 m (i.e. $\sim 25\%$ and $\sim 50\%$, respectively, see text for further details).

1012

1013 Fig. 4: POC vs Chl-a: (A) inventories in the upper 100 m of the water column, and (B)
1014 fluxes measured with sediment traps at 100 m (in white) and 300 m (in black).

1015

1016 Fig. 5: Pigment composition of sediment trap samples at 100 and 300 m. (A) Chl- a_{ST}
1017 fluxes, total pheopigment (TPheo) fluxes, and ratios of total pheopigment to Chl- a_{ST}
1018 fluxes (TPheo/Chl- a_{ST}); (B) fucoxanthin (Fuco), 19-hexanoyloxyfucoxanthin (19-Hex),
1019 and zeaxanthin (Zea) fluxes; (C) percentage of phytoplankton size classes:
1020 microphytoplankton (Micro), nanophytoplankton (Nano) and picophytoplankton (Pico).

1021

1022 Fig. 6: (A) POC export fluxes (F_C) at 100 m vs integrated net primary production (NPP)
1023 at 100 m, and (B) POC export fluxes at 300 m vs POC export fluxes at 100 m. Solid
1024 lines indicate the export and transfer efficiencies in (A) and (B), respectively. POC
1025 export fluxes were estimated using three methods: ST (diamonds), SWST (squares) and
1026 SWISP (circles, see text for further details).

1027

1028 Tab. 1: Location and date of the stations sampled for ^{234}Th , POC and PON analyses. C
1029 indicates the central station.

1030

1031 Tab. 2: Sampling of particles using sediment traps and *in-situ* pumps: location, depth,
1032 date, duration of the deployment (sediment traps) and filtered volume (*in-situ* pumps).

1033

1034 Tab. 3: ^{234}Th export fluxes derived from seawater ($F_{\text{Th,SW}}$) assuming steady state
1035 conditions, together with the fluxes derived directly from the sediment traps ($F_{\text{Th,ST}}$) at
1036 different depths.

1037

1038 Tab. 4: ^{234}Th export fluxes derived from seawater ($F_{\text{Th,SW}}$) at 100 and 300 m at the
1039 central station assuming non-steady state conditions. t_2-t_1 is the time interval between
1040 two occupations of the central station.

1041

1042 Tab. 5: POC/ ^{234}Th and PON/ ^{234}Th (C/Th and N/Th) ratios in particles collected using
1043 sediment traps (ST) and *in-situ* pumps (ISP, particles $>53 \mu\text{m}$), and POC and PON

1044 export fluxes (F_C and F_N) estimated using the ST, SWST and SWISP methods at
1045 different depths (see text for further details).

1046 *Calculated from the filter used for POC analysis. The filter used for ^{234}Th and POC
1047 analyses has not been considered because it was likely contaminated with organic
1048 carbon, presenting unusually high C/N molar ratio (14) compared with our overall
1049 sample average (8 ± 1).

1050

1051 Tab. 6: Export and transfer efficiencies using the POC export estimated from the ST,
1052 SWST and SWISP methods (see text for further details).

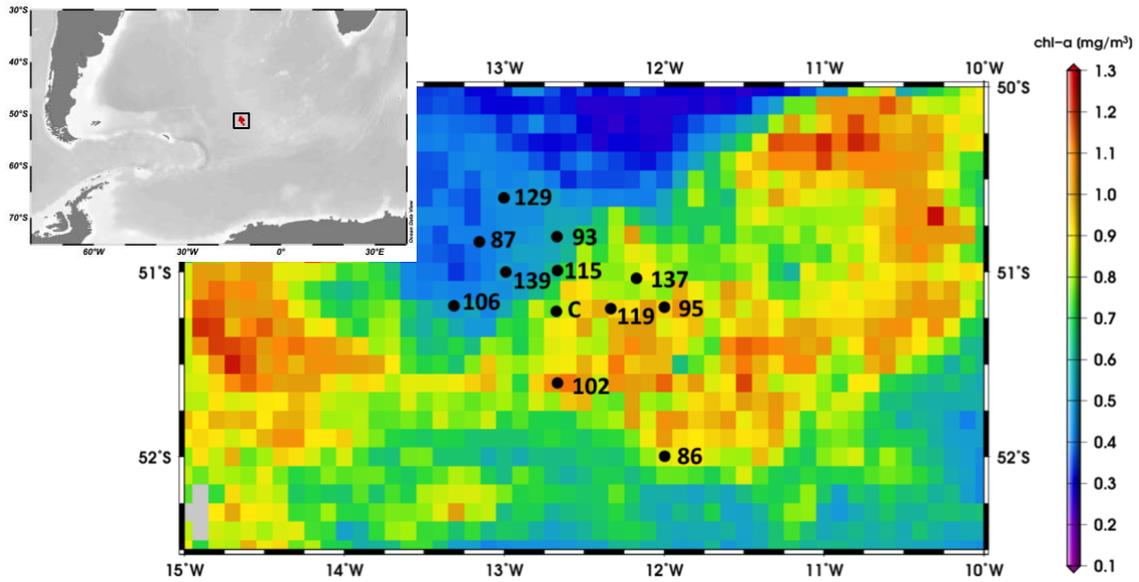
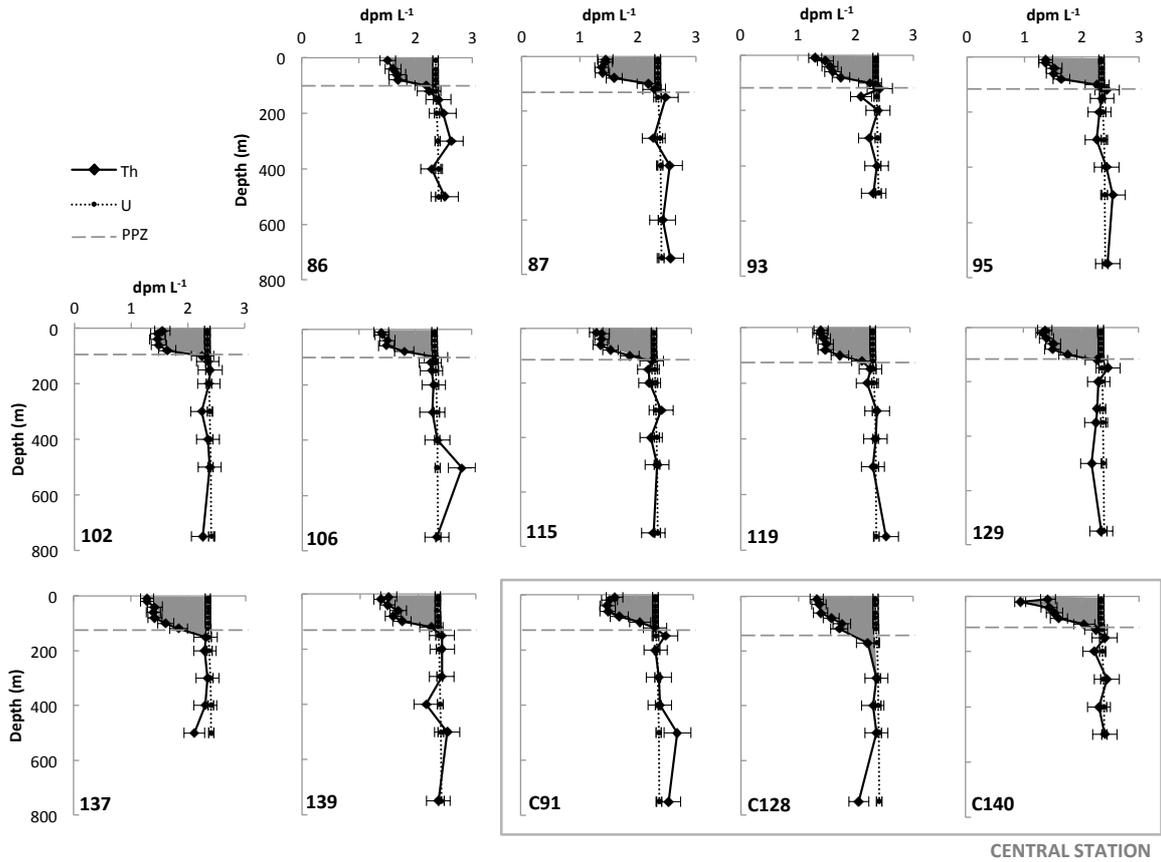


Fig. 1: Study area and sampled stations for ^{234}Th , POC and PON analyses. C represents the central station, which was occupied 7 times: C91, C98, C99, C114, C128, C136, C140. See Table 1 for further details regarding sampling dates. The satellite plot represents the mean Chl-a concentration from the OC-CCI Chl-a product version-2 during the sampling period (29 Jan. to 17 Feb. 2012).



CENTRAL STATION

Fig. 2: Vertical activity profiles for ^{234}Th (black diamonds) and ^{238}U (dotted line) from 10 to 500-750 m depth. Grey shaded area indicates surface ^{234}Th deficits respect to ^{238}U . ^{238}U was derived from salinity (Owens et al., 2011). Primary Production Zone (PPZ, dashed grey line) is defined as the depth at which fluorescence reaches 10% of its maximum value (Owens, 2013).

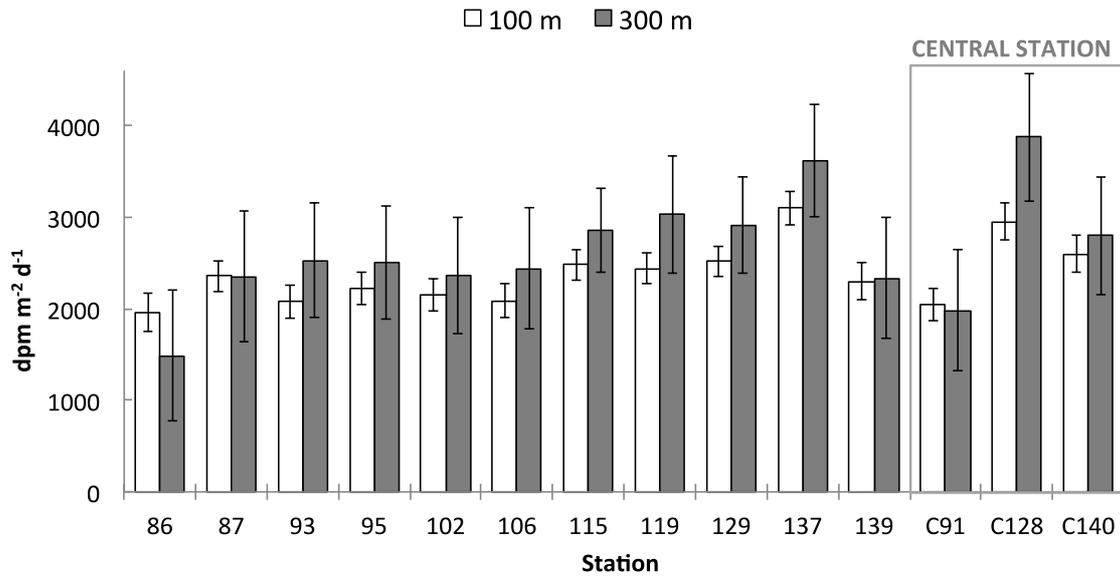


Fig. 3: ²³⁴Th export fluxes derived from seawater at 100 m (in white) and 300 m (in grey) assuming steady state conditions. The negligence of horizontal advection would lead to an error on these estimates of 580 ± 440 dpm m⁻² d⁻¹ at 100 m and 1500 ± 1700 dpm m⁻² d⁻¹ at 300 m (i.e. ~25% and ~50%, respectively, see text for further details).

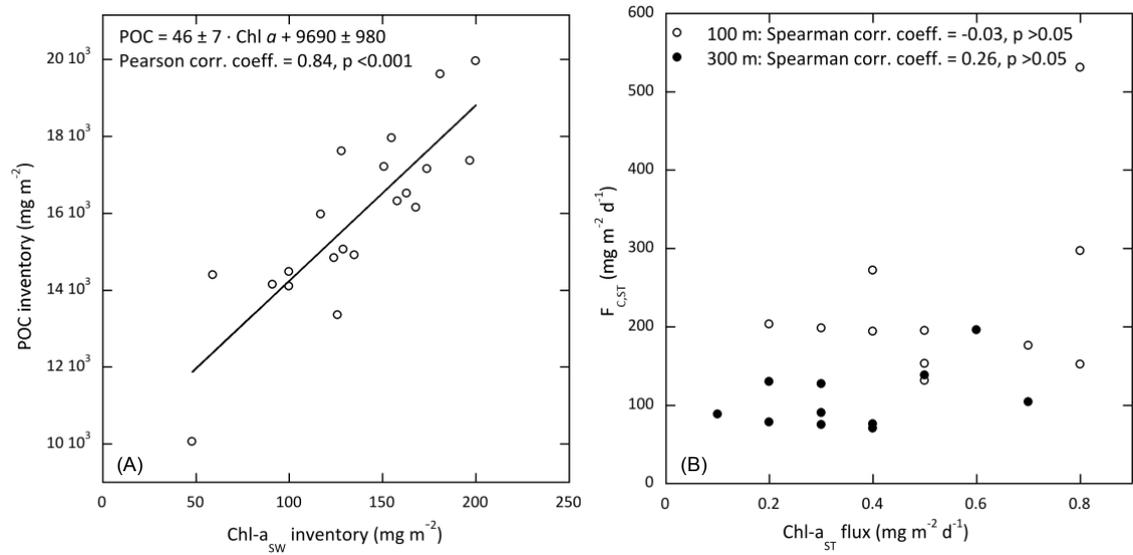


Fig. 4: POC vs Chl-a: (A) inventories in the upper 100 m of the water column, and (B) fluxes measured with sediment traps at 100 m (in white) and 300 m (in black).

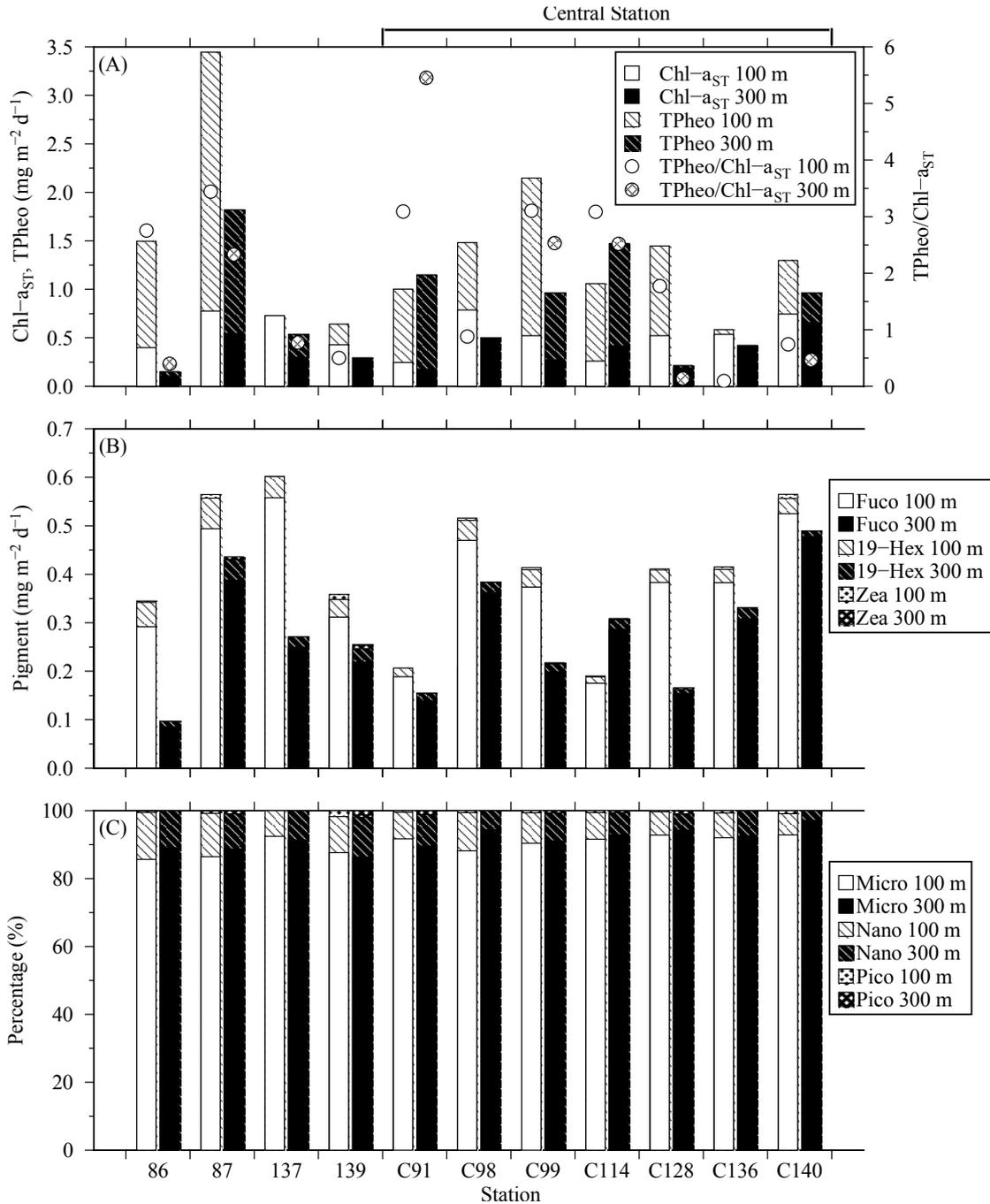


Fig. 5: Pigment composition of sediment trap samples at 100 and 300 m. (A) Chl- a_{ST} fluxes, total pheopigment (TPheo) fluxes, and ratios of total pheopigment to Chl- a_{ST} fluxes (TPheo/Chl- a_{ST}); (B) fucoxanthin (Fuco), 19-hexanoyloxyfucoxanthin (19-Hex), and zeaxanthin (Zea) fluxes; (C) percentage of phytoplankton size classes: microphytoplankton (Micro), nanophytoplankton (Nano) and picophytoplankton (Pico).

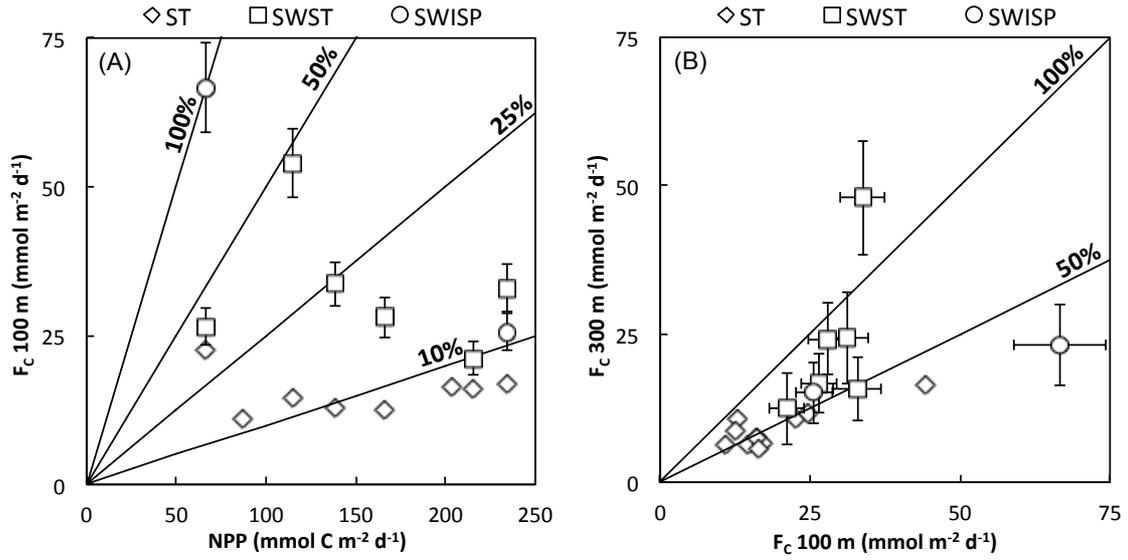


Fig. 6: (A) POC export fluxes (F_c) at 100 m vs integrated net primary production (NPP) at 100 m, and (B) POC export fluxes at 300 m vs POC export fluxes at 100 m. Solid lines indicate the export and transfer efficiencies in (A) and (B), respectively. POC export fluxes were estimated using three methods: ST (diamonds), SWST (squares) and SWISP (circles, see text for further details).

Tab. 1: Location and date of the stations sampled for ^{234}Th , POC and PON analyses. C indicates the central station.

Station	Lon. (°W)	Lat. (°S)	Date (2012)
86	12.00	52.00	29-30 Jan.
87	13.16	50.84	2-3 Feb.
93	12.67	50.81	4 Feb.
95	12.00	51.19	5 Feb.
102	12.67	51.60	7 Feb.
106	13.31	51.18	7 Feb.
115	12.67	50.99	9 Feb.
119	12.33	51.20	10 Feb.
129	13.00	50.60	13 Feb.
137	12.17	51.04	14-15 Feb.
139	12.99	51.00	15-16 Feb.
C91	12.67	51.21	3-5 Feb.
C98	12.67	51.21	5-6 Feb.
C99	12.67	51.21	6-8 Feb.
C114	12.67	51.21	8-11 Feb.
C128	12.67	51.21	12-13 Feb.
C136	12.67	51.21	14-15 Feb.
C140	12.67	51.21	16-17 Feb.

Tab. 2: Sampling of particles using sediment traps and *in-situ* pumps: location, depth, date, duration of the deployment (sediment traps) and filtered volume (*in-situ* pumps).

Station	Lon. (°W)	Lat. (°S)	Deployment sediment traps			Deployment <i>in-situ</i> pumps		
			Depth (m)	Date (2012)	Duration (h)	Depth (m)	Date (2012)	Volume (L)
86	12.00	52.00	100, 300	29 Jan.	23			
87	13.16	50.84	100, 300	2 Feb.	20			
137	12.17	51.04	120, 320	14 Feb.	15			
139	12.99	51.00	120, 320	15 Feb.	19	100, 150, 300, 400	16 Feb.	250-900
C91	12.67	51.21	100, 300	3 Feb.	53	100, 150, 300, 400	3 Feb.	450-1200
C98	12.67	51.21	100, 300	5 Feb.	18			
C99	12.67	51.21	100, 300	6 Feb.	50			
C114	12.67	51.21	100, 300	8 Feb.	72			
C128	12.67	51.21	120, 320	12 Feb.	29			
C136	12.67	51.21	120, 320	14 Feb.	22			
C140	12.67	51.21	120, 320	16 Feb.	18			

Tab. 3: ^{234}Th export fluxes derived from seawater ($F_{\text{Th,SW}}$) assuming steady state conditions, together with the fluxes derived directly from the sediment traps ($F_{\text{Th,ST}}$) at different depths.

Station	Depth (m)	$F_{\text{Th,SW}}$ (dpm $\text{m}^{-2} \text{d}^{-1}$)	$F_{\text{Th,ST}}$ (dpm $\text{m}^{-2} \text{d}^{-1}$)
86	100	1960 \pm 210	1700 \pm 140
	300	1490 \pm 710	970 \pm 80
87	100	2360 \pm 170	4000 \pm 320
	300	2350 \pm 710	1990 \pm 170
93	100	2080 \pm 180	
	300	2530 \pm 630	
95	100	2220 \pm 180	
	300	2510 \pm 620	
102	100	2150 \pm 180	
	300	2360 \pm 640	
106	100	2090 \pm 190	
	300	2440 \pm 660	
115	100	2480 \pm 170	
	300	2860 \pm 470	
119	100	2440 \pm 170	
	300	3020 \pm 640	
129	100	2520 \pm 170	
	300	2910 \pm 520	
137	120	3100 \pm 180	1090 \pm 100
	300-320	3620 \pm 620	620 \pm 60
139	120	2510 \pm 200	2440 \pm 200
	150	2540 \pm 260	
	300-320	2340 \pm 660	1870 \pm 160
	400	2660 \pm 900	
C91	100	2050 \pm 180	1470 \pm 120
	150	2070 \pm 270	
	300	1980 \pm 660	900 \pm 70
	400	1910 \pm 910	
C98	100		1430 \pm 120
	300		790 \pm 70
C99	100		1370 \pm 110
	300		960 \pm 80
C114	100		1200 \pm 100
	300		590 \pm 50
C128	120	2950 \pm 200	1050 \pm 90
	300-320	3870 \pm 700	920 \pm 80
C140	120	2600 \pm 210	1450 \pm 130
	300-320	2800 \pm 650	1220 \pm 110

Tab. 4: ^{234}Th export fluxes derived from seawater ($F_{\text{Th,SW}}$) at 100 and 300 m at the central station assuming non-steady state conditions. t_2-t_1 is the time interval between two occupations of the central station.

Stations	t_2-t_1 (days)	Depth (m)	$F_{\text{Th,SW}}$ (dpm $\text{m}^{-2} \text{d}^{-1}$)
C91-C128	10	100	4290 \pm 880
		300	9600 \pm 3400
C128-C140	4	100	1600 \pm 2000
		300	-5500 \pm 7600

Tab. 5: POC/²³⁴Th and PON/²³⁴Th (C/Th and N/Th) ratios in particles collected using sediment traps (ST) and *in-situ* pumps (ISP, particles >53 μm), and POC and PON export fluxes (F_C and F_N) estimated using the ST, SWST and SWISP methods at different depths (see text for further details).

Station	Depth (m)	C/Th (μmol dpm ⁻¹)		N/Th (μmol dpm ⁻¹)		F _C (mmol m ⁻² d ⁻¹)			F _N (mmol m ⁻² d ⁻¹)		
		ST	ISP (> 53 μm)	ST	ISP (> 53 μm)	ST	SWST	SWISP	ST	SWST	SWISP
86	100	10.8 ± 0.9		1.4 ± 0.1		16	21 ± 3		2.0	2.6 ± 0.4	
	300	8.3 ± 0.7		0.85 ± 0.07		7.3	12 ± 6		0.74	1.3 ± 0.6	
87	100	13 ± 1		1.5 ± 0.1		44	31 ± 3		5.1	3.6 ± 0.4	
	300	10.3 ± 0.9		1.05 ± 0.09		16	24 ± 8		1.7	2.5 ± 0.8	
137	120	17 ± 2		2.2 ± 0.2		15	54 ± 6		1.8	6.7 ± 0.7	
	320					*6.3					
139	100-120	10.6 ± 0.9	29 ± 2	1.2 ± 0.1	5.2 ± 0.4	23	26 ± 3	67 ± 8	2.5	2.9 ± 0.3	12 ± 1
	150		20 ± 2		2.8 ± 0.3			52 ± 7			7 ± 1
	300-320	7.1 ± 0.6	9.9 ± 0.9	0.89 ± 0.07	1.1 ± 0.1	11	17 ± 5	23 ± 7	1.3	2.1 ± 0.6	2.6 ± 0.8
	400		9.0 ± 0.8		1.3 ± 0.1			24 ± 8			3 ± 1
C91	100	16 ± 1	13 ± 1	1.9 ± 0.2	2.1 ± 0.2	17	33 ± 4	26 ± 3	2.0	4.0 ± 0.5	4.2 ± 0.5
	150		12 ± 1		1.7 ± 0.1			25 ± 4			3.5 ± 0.5
	300	7.9 ± 0.6	7.6 ± 0.6	0.90 ± 0.07	1.3 ± 0.1	6.5	16 ± 5	15 ± 5	0.74	1.8 ± 0.6	2.5 ± 0.9
	400		4.3 ± 0.3		0.73 ± 0.06			8 ± 4			1.4 ± 0.7
C98	100	19 ± 2		2.4 ± 0.2		25			3.1		
	300	17 ± 2		1.8 ± 0.2		12			1.2		
C99	100	15 ± 1		1.8 ± 0.1		16			2.0		
	300	9.1 ± 0.8		1.06 ± 0.09		7.5			0.88		
C114	100	14 ± 1		1.9 ± 0.2		16			2.3		
	300	9.8 ± 0.8		1.3 ± 0.1		5.8			0.77		
C128	120	11 ± 1		1.6 ± 0.1		13	34 ± 4		1.7	4.6 ± 0.5	
	320	12 ± 1		1.1 ± 0.1		11	50 ± 10		1.0	4.3 ± 0.9	
C136	120					11					
	320					6.3					
C140	120	11 ± 1		1.3 ± 0.1		13	28 ± 3		1.6	3.5 ± 0.4	
	320	8.6 ± 0.8		1.1 ± 0.1		8.6	24 ± 6		1.1	3.0 ± 0.8	

*Calculated from the filter used for POC analysis. The filter used for ²³⁴Th and POC analyses has not been considered because it was likely contaminated with organic carbon, presenting unusually high C/N molar ratio (14) compared with our overall sample average (8 ± 1).

Tab. 6: Export and transfer efficiencies using the POC export estimated from the ST, SWST and SWISP methods (see text for further details).

Station	Export efficiency (%)			Transfer efficiency (%)		
	ST	SWST	SWISP	ST	SWST	SWISP
86	7.5	10 ± 1		45	60 ± 30	
87				37	80 ± 30	
137	13	47 ± 5		43		
139	34	40 ± 5	100 ± 10	47	60 ± 20	30 ± 10
C91	7.2	14 ± 2	11 ± 1	38	50 ± 20	60 ± 20
C98				47		
C99				46		
C114	8.1			35		
C128	9.2	24 ± 3		84	140 ± 30	
C136	12			58		
C140	7.6	17 ± 2		68	90 ± 20	