1	Carbon export fluxes and export efficiency in the central Arctic during the
2	record sea-ice minimum in 2012: a joint 234 Th/ 238 U and 210 Po/ 210 Pb study
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17	Key Points:
18 19	• First use of ²³⁴ Th/ ²³⁸ U together with ²¹⁰ Po/ ²¹⁰ Pb as proxies for particulate organic carbon export in the Arctic
20	• Low particulate organic carbon fluxes escaping from the euphotic zone during the record

- 21 sea-ice minimum in 2012
- High export efficiency of the biological pump in the central Arctic

23 Abstract

The Arctic sea-ice extent reached a record minimum in September 2012. Sea-ice decline 24 increases the absorption of solar energy in the Arctic Ocean, affecting primary production and 25 the plankton community. How this will modulate the sinking of particulate organic carbon (POC) 26 from the ocean surface remains a key question. We use the $^{234}\text{Th}/^{238}\text{U}$ and $^{210}\text{Po}/^{210}\text{Pb}$ 27 radionuclide pairs to estimate the magnitude of the POC export fluxes in the upper ocean of the 28 central Arctic in summer 2012, covering time scales from weeks to months. The ²³⁴Th/²³⁸U 29 proxy reveals that POC fluxes at the base of the euphotic zone were very low ($2 \pm 2 \text{ mmol C m}^{-2}$ 30 d^{-1}) in late summer. Relationships obtained between the ²³⁴Th export fluxes and the 31 phytoplankton community suggest that prasinophytes contributed significantly to downward 32 fluxes, likely via incorporation into sea-ice algal aggregates and zooplankton-derived material. 33 The magnitude of the depletion of ²¹⁰Po in the upper water column over the entire study area 34 indicates that particle export fluxes were higher before July/August than later in the season. ²¹⁰Po 35 fluxes and ²¹⁰Po-derived POC fluxes correlated positively with sea-ice concentration, showing 36 that particle sinking was greater under heavy sea-ice conditions than under partially ice covered 37 regions. Although the POC fluxes were low, a large fraction of primary production (>30%) was 38 exported at the base of the euphotic zone in most of the study area during summer 2012, 39 indicating a high export efficiency of the biological pump in the central Arctic. 40

41 **1 Introduction**

Climate change is triggering an unprecedented decline in Arctic sea ice. In September 42 2012 the sea-ice cover amounted to less than half of its 1979-2000 baseline [Overland and 43 Wang, 2013]. Such a decrease in ice extent and thickness [Haas et al., 2008] allows more 44 sunlight to be transmitted through the sea ice, increasing the absorption of solar energy in the 45 Arctic Ocean [Nicolaus et al., 2012] and affecting sea-ice and upper-ocean ecosystems 46 [Wassmann, 2011]. Net primary production (NPP) increased by 30% between 1998 and 2012 47 according to a satellite-based study [Arrigo and van Dijken, 2015]. Yet this kind of approach 48 does not take into account the productivity of either under-ice phytoplankton nor sea-ice algae, 49 even though it can be substantial [Gosselin et al., 1997; Fortier et al., 2002; Lee et al., 2010; 50 Arrigo et al., 2012; Fernández-Méndez et al., 2015]. However, light-driven increments in NPP 51 52 will be constrained if nutrient supply to surface waters do not increase considerably by mixing or

⁵³ upwelling [e.g. *Tremblay et al.*, 2015]. Besides this, enhanced NPP does not necessarily mean ⁵⁴ larger export fluxes of particulate organic carbon (POC) to the deep ocean, since the changing ⁵⁵ Arctic scenario favors a phytoplankton community structure based on the smallest cells [*Li et al.*, ⁵⁶ 2009]. Overall, it remains uncertain how the changes in NPP and plankton community will affect ⁵⁷ the sinking of POC from the ocean surface, and in turn contribute to the marine sequestration of ⁵⁸ CO₂ [*Honjo et al.*, 2010; *Anderson and Macdonald*, 2015].

To date, the Arctic Ocean is considered a weak sink for atmospheric CO₂, accounting for 59 ~6% of the global oceanic uptake [Gruber et al., 2009]. An essential component of the ocean 60 carbon sink is the "biological pump" driven by the export of organic particles from the ocean 61 surface to its interior [Falkowski et al., 1998]. During the productive season, the surface 62 downward fluxes of POC are widely heterogeneous in the Arctic, reaching higher values (>30 63 mmol C m⁻² d⁻¹) over the shelves [e.g. Cochran et al., 1995b; Lepore et al., 2007] in comparison 64 to the central Arctic (<5 mmol C m⁻² d⁻¹) [e.g. Moran et al., 1997; Cai et al., 2010]. However, in 65 summer 2012, a widespread deposition of ice algal biomass on the seafloor (>3000 m, median 66 estimate of 750 mmol C m⁻²) was observed in the central Arctic associated with rapid ice melt 67 [*Boetius et al.*, 2013]. 68

69 The export efficiency is defined as the ratio between export and production, which 70 indicates the strength of the biological pump [Buesseler and Boyd, 2009]. A recent model study reports a high annual mean export efficiency of >30% in Arctic waters [Henson et al., 2015]. 71 72 Nevertheless, primary production and export data are very scarce, especially in the interior 73 basins [Gustafsson and Andersson, 2012; Matrai et al., 2013]. Indeed, the temporal mismatch between the measurement of production and export, combined with the existence of a long lag 74 period between both processes in the Arctic (30-40 days), make the assessment of the export 75 efficiency on a seasonal scale difficult [Henson et al., 2015]. 76

The radionuclide pairs ²³⁴Th/²³⁸U and, to a lesser extent, ²¹⁰Po/²¹⁰Pb have been used as proxies of POC export since the 90s [*Buesseler et al.*, 1992; *Shimmield et al.*, 1995], but very few studies have used both pairs together [*Verdeny et al.*, 2009; *Stewart et al.*, 2011; *Wei et al.*, 2011; *Le Moigne et al.*, 2013a]. Several authors have recommended the simultaneous use of ²³⁴Th/²³⁸U and ²¹⁰Po/²¹⁰Pb since they cover different time scales, from weeks to months, respectively, and ²³⁴Th and ²¹⁰Po have different biogeochemical behaviors, providing complementary information on POC export fluxes [*Friedrich and Rutgers van der Loeff*, 2002; *Verdeny et al.*, 2009; *Stewart et al.*, 2011].

In this study, we aim to estimate the magnitude of the POC fluxes at the bottom of the 85 86 euphotic zone and within the upper mesopelagic layer in the central Arctic during the record seaice minimum in 2012, as well as identify mechanisms that control particle export by means of 87 ²³⁴Th/²³⁸U and ²¹⁰Po/²¹⁰Pb. The use of both pairs may shed light on the apparent mismatch 88 between the low ²³⁴Th-based export production estimates [Cai et al., 2010] and the benthic 89 observations of massive sea-ice algae deposits [Boetius et al., 2013] in the central Arctic. It 90 might also give a hint of the trend that POC fluxes may follow as the sea ice continues to decline. 91 To this purpose we: 92

93 1) Quantify the POC export fluxes at the bottom of the euphotic zone, 50, 100 and 150 m on 94 short-term and seasonal scales by using the 234 Th/ 238 U and 210 Po/ 210 Pb pairs.

95 2) Identify potential relationships between sea-ice conditions, phytoplankton community
96 and particle export.

3) Assess the export efficiency combining the export estimates at the bottom of the euphotic
zone with daily, weekly and annual NPP estimates.

- 99 2 Materials and Methods
- 100 2.1 Study area

The sampling was performed from 11 August to 28 September 2012 during the ARK-XXVII/3 expedition in the Eurasian Basin of the central Arctic (2 August-8 October, 2012; R/V Polarstern; *Boetius* [2013]). The survey coincided with a new record low of sea-ice cover since the beginning of satellite imagery in 1978 [*Parkinson and Comiso*, 2013]. The specific locations and dates of the sea-ice stations are given in Figure 1 and Table 1.

106 2.2 Total
234
Th/ 238 U and 210 Po/ 210 Pb

107 Total ²³⁴Th, ²¹⁰Po and ²¹⁰Pb activities were determined from seawater samples collected 108 using Niskin bottles attached to a CTD rosette. 12-depth vertical profiles from 10 to 400 m were 109 taken, with the highest resolution in the upper 150 m of the water column.

110 Total ²³⁴Th activities were determined from 4 L of seawater at nine stations. Additionally,

replicates of deep samples (1500-3000 m) were collected for calibration purposes [Rutgers van 111 der Loeff et al., 2006]. The samples were processed following the MnO₂ co-precipitation 112 technique [Buesseler et al., 2001] using ²³⁰Th as a chemical yield tracer [Pike et al., 2005]. 113 Briefly, the precipitates were filtered through QMA quartz fiber filters, dried overnight at 50 °C 114 and prepared for beta counting. The counting was done on board using low background beta 115 counters (Risø National Laboratories, Denmark). Samples were re-measured after seven months 116 to quantify background activities. ²³⁰Th recoveries were determined in all filters by inductively 117 coupled plasma mass spectrometry (ICP-MS) as described in Roca-Martí et al. [2016]. The 118 average chemical recovery was $94 \pm 4\%$ (n = 107). The parent ²³⁸U activity was derived from 119 salinity using the relationship given by *Owens et al.* [2011]. Stations 4, 5 and 6 had salinities of 120 30.0-32.5 from 10 to 30 m (n = 15), falling below the range used by Owens et al. [2011]. For 121 these samples, we also applied the U-salinity relationship given by Not et al. [2012] determined 122 from sea ice, surface seawater and sea-ice brine samples, covering a wide salinity range (0-135). 123 A difference of only 1.1% in ²³⁸U activity, which is lower than its associated uncertainty (1.9-124 2.3%), was obtained using the two relationships, validating the use of Owens's relationship in the 125 present study. The ²³⁴Th activity uncertainties were always $\leq 6\%$, which include those 126 uncertainties associated with counting, detector background and calibration, and ICP-MS 127 128 measurements.

Total ²¹⁰Po and ²¹⁰Pb activities were determined from 11 L of seawater at seven stations 129 using the cobalt-ammonium pyrrolidine dithiocarbamate (Co-APDC) co-precipitation technique 130 [Fleer and Bacon, 1984]. Samples were immediately acidified after collection with HCl to pH <2 131 and spiked with stable Pb and ²⁰⁹Po as chemical yield tracers. Cobalt nitrate and APDC solutions 132 were added after at least one day of isotope equilibration. Samples were filtered through 0.2 µm 133 membrane filters and stored for later processing at the home laboratory. The filters were digested 134 using concentrated HNO₃ and samples were reconstructed with 1 M HCl. ²¹⁰Po and ²¹⁰Pb were 135 separated by auto-deposition of polonium onto silver discs during six hours [Flynn, 1968]. The 136 silver discs were then counted by alpha spectrometry using passivated implanted planar silicon 137 (PIPS) alpha detectors (Canberra, USA) and silicon surface barrier (SSB) alpha detectors 138 139 (EG&G Ortec, USA). Solutions were re-plated and passed through an anion exchange resin (AG 1-X8) to ensure the complete elimination of polonium from samples [Rigaud et al., 2013]. 140 Samples were re-spiked with ²⁰⁹Po and stored for 9-11 months for later determination of ²¹⁰Pb 141

via ²¹⁰Po ingrowth. At that time samples were plated and counted once more by alpha 142 spectrometry. ²¹⁰Pb and ²¹⁰Po activities at sampling time were calculated applying in-growth, 143 144 decay and recovery corrections following Rigaud et al. [2013]. Two aliquots from each sample were taken before the first and last platings to determine the chemical recovery of stable Pb by 145 inductively coupled plasma optical emission spectrometry (ICP-OES). The average recovery was 146 $87 \pm 9\%$ (n = 83). The activity uncertainties were on average 7% for ²¹⁰Pb and 16% for ²¹⁰Po, 147 which include those uncertainties associated with counting, detector background and ²⁰⁹Po 148 activity. The larger uncertainties of ²¹⁰Po are due to the time elapsed between sampling and the 149 first Po plating (>80 days). All data of total ²³⁴Th, ²³⁸U, ²¹⁰Po and ²¹⁰Pb activities are available at 150 http://doi.pangaea.de/10.1594/PANGAEA.858790. 151

152 2.3 Particulate fraction

Large (>53 µm) particles for analyses of ²³⁴Th, ²¹⁰Po, ²¹⁰Pb, POC and particulate organic 153 nitrogen (PON) were collected using in situ pumps (ISP, Challenger Oceanic, UK). Four ISP 154 were deployed at each station at 25, 50, 100 and 150 m, filtering on average 1500 L. Particles 155 were retained using 53-µm pore size nylon mesh screens and rinsed with filtered seawater. After 156 157 homogenization the sample was subdivided into two aliquots: one was filtered through precombusted QMA filters to analyze ²³⁴Th, POC and PON on the same filter, and the other aliquot 158 was filtered through QMA filters to analyze ²¹⁰Po and ²¹⁰Pb. Swimmers observed by naked eye 159 were picked from all samples. The activity of ²³⁴Th in particles was measured by beta counting 160 161 as described for the water samples. POC and PON were determined with an EuroVector 162 Elemental Analyzer, pre-treating the filters with diluted HCl [Knap et al., 1996]. The results were corrected for POC and PON blanks (1.7 \pm 0.1 and 0.35 \pm 0.06 µmol, respectively), 163 representing on average 5 and 8% of the POC and PON measurements, respectively. The filters 164 for ²¹⁰Po and ²¹⁰Pb determination were spiked with ²⁰⁹Po and stable Pb, digested using a mixture 165 of concentrated HNO₃, HCl and HF, evaporated to dryness and reconstructed with 1 M HCl. 166 Samples were processed and measured by alpha spectrometry as described for the water samples. 167 All data of particulate ²³⁴Th, ²¹⁰Po, ²¹⁰Pb, and organic carbon and nitrogen concentrations are 168 available at http://doi.pangaea.de/10.1594/PANGAEA.858790. 169

2.4 Pigments

1-L seawater samples were taken from Niskin bottles attached to the CTD rosette from 171 three to four depths in the upper 30 m at eight stations. The samples were immediately filtered on 172 GF/F filters, frozen in liquid nitrogen, and stored at -80 °C until further analyses by high 173 performance liquid chromatography (HPLC) at the home laboratory. The samples were measured 174 using a Waters 600 controller equipped with an auto sampler (717 plus), a photodiode array 175 detector (2996), a fluorescence detector (2475) and the EMPOWER software. 50 µL of internal 176 177 standard (canthaxanthin) and 1.5 mL acetone were added to each filter vial and then homogenized for 20 seconds in a Precellys® tissue homogenizer. After centrifugation the 178 supernatant liquids were filtered through 0.2 µm PTFE filters (Rotilabo) and placed in Eppendorf 179 cups. 100 µL-aliquots were transferred to the auto sampler (4 °C), premixed with 1 M 180 181 ammonium acetate solution in a 1:1 volume ratio just prior to analysis, and injected onto the 182 HPLC-system. Pigments were analyzed by reverse-phase HPLC using a VARIAN Microsorb-MV3 C8 column (4.6x100 mm) and HPLC-grade solvents (Merck). Solvent A consisted of 70% 183 184 methanol and 30% 1 M ammonium acetate, and solvent B contained 100% methanol. The gradient was modified after Barlow et al. [1997]. Eluting pigments were detected by absorbance 185 (440 nm) and fluorescence (Ex: 410 nm, Em: >600 nm). Pigments were identified by comparing 186 their retention times with those of pure standards. Additional confirmation for each pigment was 187 done by comparing spectra with on-line diode array absorbance spectra between 390 and 750 nm 188 stored in the library. Pigment concentrations were quantified based on peak areas of external 189 standards, which were spectrophotometrically calibrated using extinction coefficients published 190 by Bidigare [1991] and Jeffrey et al. [1997]. The taxonomic structure of the phytoplankton 191 groups (diatoms. dinoflagellates 1. dinoflagellates 2, haptophytes 3, haptophytes 4. 192 cryptophytes, prasinophytes_1, prasinophytes_2, pelagophytes and chlorophytes) was calculated 193 from marker pigment ratios using the CHEMTAX® program [Mackey et al., 1996]. Pigment 194 ratios were constrained as suggested by Higgins et al. [2011] based on molecular analyses of 18S 195 rDNA [Kilias et al., 2013] and microscopic examination of representative samples. 196 Phytoplankton size classes (micro-, nano-, and picoplankton) were estimated according to Uitz et 197 al. [2006] and Hirata et al. [2011], summarized by Taylor et al. [2011]. Microplankton 198 corresponded to phytoplankton with size between 20 and 200 µm, nanoplankton between 2 and 199

200 20 μ m and picoplankton <2 μ m. The phytoplankton classifications by group and size are 201 expressed as percentage of total chlorophyll *a* (Chl-a) biomass.

202

2.5 Primary production

In situ NPP was measured at eight stations using the ¹⁴C uptake method [Steemann 203 Nielsen, 1952], with minor modifications as described in Fernández-Méndez et al. [2015]. 204 Seawater, melted sea-ice cores and melt pond samples (one 200 mL sample per environment and 205 station) were spiked with 0.1 μ Ci mL⁻¹ of ¹⁴C labelled sodium bicarbonate (Moravek 206 Biochemicals, USA) and incubated for 12 hours at -1.3 °C under different scalar irradiances (0-207 420 µmol photons m⁻² s⁻¹). Depth-integrated in situ rates were calculated for each environment as 208 a function of the available photosynthetically active radiation (PAR) using the photosynthetic 209 parameters obtained in the photosynthesis vs. irradiance curves. Water column production was 210 211 integrated over the euphotic zone (1% of incoming irradiance) and sea-ice algae production over the ice thickness. 212

At the same stations we calculated the integrated amount of NPP that potentially occurred 213 one and two weeks before sampling using the Central Arctic Ocean Primary Productivity 214 (CAOPP) model [Fernández-Méndez et al., 2015]. This model calculates NPP from incident 215 216 light and sea-ice conditions based on different remote-sensing datasets on the basis of photosynthesis-irradiance curves measured during the cruise. NPP was calculated for each day 217 during the 14 days prior to sampling, summed up to integrate values for the one- and two-week 218 period before sampling, and divided by 7 and 14 days, respectively, to obtain average daily rates 219 for these two periods. 220

Annual new NPP was calculated from the nitrate drawdown in the mixed layer since 221 222 previous winter at nine stations, as described in *Fernández-Méndez et al.* [2015]. The annual total inorganic nitrogen uptake was then transformed to carbon units using the Redfield ratio 223 106C:16N [Smith et al., 1997; Codispoti et al., 2013], giving annual new NPP estimates for sea 224 225 ice and water column during the Arctic productive season. To calculate an average daily rate we assumed a productive season of 120 days [Gradinger et al., 1999]. Although most of the new 226 NPP occur before late summer, we note that these estimates may be underestimated mainly for 227 228 the first stations sampled in August. This method assumes that lateral input of nitrate from rivers 229 or shelves is negligible, which should be the case of the present study (>81°N) due to its

- 230 consumption in Arctic shelf waters [Le Fouest et al., 2013]. Further, this method do not take into
- consideration nitrification and upward flux of nitrate, which are assumed to have a relatively 231
- small contribution to the nitrate concentrations in the mixed layer in comparison with the 232
- biological uptake. 233
- **3 Results** 234
- 3.1 Study area 235

- Sea-ice conditions, phytoplankton communities and primary production rates in the study 236
- 237 area are described below and summarized in Table 1.
- Table 1: Location and date of the stations sampled during the ARK-XXVII/3 cruise together with information on 238 239 oceanographic and sea-ice conditions, Chl-a inventory at 30 m depth, phytoplankton classifications by size and
- group, and NPP estimates (see text for further details). The methods used to estimate POC export fluxes in each station are specified as: Th/U (234 Th/ 238 U), Po/Pb (210 Po/ 210 Pb) and ST (sediment trans. [Lalande et al. 2014]) 240 241

on are specified as: Th/U	$(^{234}\text{Th}/^{238}\text{I})$	U), Po/Pb) (²¹⁰ Po/ ²¹	Pb) and S	ST (sedin	ient traps	, [Laland	e et al., 2	014]).
Station	1	2	3	4	5	6	7	8	9
Polarstern station #	PS80/	PS80/	PS80/	PS80/	PS80/	PS80/	PS80/	PS80/	PS80/
	224	237	255	277	323	335	349	360	384
Longitude (°E)	31.19	75.99	110.11	129.83	131.12	123.47	60.97	57.07	17.59
Latitude (°N)	84.03	83.92	83.08	82.89	81.93	85.17	87.93	88.80	84.37
Date (2012)	9-11	14-16	20-22	25-26	4-5	7-9	18-19	22-23	28-29
Dute (2012)	Aug.	Aug.	Aug.	Aug.	Sept.	Sept.	Sept.	Sept.	Sept.
Methods	Th/U, Po/Pb, ST	Th/U, Po/Pb, ST	Th/U, Po/Pb, ST	Th/U, Po/Pb, ST	Th/U, ST	Th/U, Po/Pb, ST	Th/U, Po/Pb, ST	Th/U, Po/Pb, ST	Th/U, ST
Euphotic zone depth (m) ^a	24	29	30	29	33	29	15	7	
Mixed layer depth (m)	16	20	18	22	20	25	29	30	22
Sea-ice thickness $(m)^{a,b}$	1.0	1.3	0.9	0.9	0.8	0.7	1.6	1.8	1.2
Sea ice-concentration (%) ^{a,b}	80	80	70	80	60	50	100	100	100
Chl-a inventory (mg m ⁻²)	4.8	22.8	8.9	7.0	8.9	11.2	6.1	3.3	2.5
Phytoplankton size (% Chl-a biomass)									
Microplankton	36	29	38	54	36	14	34	nd	39
Nanoplankton	22	0	2	13	4	28	44	nd	32
Picoplankton	42	71	60	33	60	58	22	nd	29
Phytoplankton group (% Chl-a biomass)									
Diatoms	19	5	4	44	27	3	23	nd	22
Dinoflagellates_1	0	0	8	2	0	0	0	nd	0
Dinoflagellates_2	8	0	9	0	2	11	1	nd	12
Haptophytes_3	21	0	0	4	0	26	61	nd	19

Cryptophytes	0	0	1	18	10	0	0	nd	0
Prasinophytes_1	51	33	8	26	27	35	12	nd	22
Prasinophytes_2	0	61	61	0	28	0	0	nd	0
Pelagophytes	0	0	1	1	3	0	0	nd	21
Chlorophytes	0	0	0	0	0	21	1	nd	0
<i>NPP estimates</i> (mmol $C m^{-2} d^{-1}$)									
In-situ	3.3	2.7	1.3	0.5	5.0	2.3	0.2	0.1	nd
One week	2.3	2.3	2.2	3.5	1.8	1.9	0.6	0.5	nd
Two weeks	2.4	2.5	2.2	3.3	2.2	2.3	0.8	0.6	nd
Annual	3.3	4.8	2.1	2.9	5.6	3.2	11.9	9.9	7.9

nd = no available data. ^a Data from *Fernández-Méndez et al.* [2015]. ^b Data from *Katlein et al.* [2014].

3.1.1 Oceanographic and sea-ice conditions

Stations were located over the deep Arctic (>3000 m) in the Nansen (stations 1-3 and 9) 244 and Amundsen Basins (stations 4-8, Figure 1). The sea-ice conditions encountered during the 245 expedition are described in *Katlein et al.* [2014]. Stations located north of 87°N (stations 7 and 8) 246 had multi-year ice, 1.6-1.8 m thick, while the rest consisted of degraded first-year ice of 0.7-1.3 247 m. The sea-ice concentration varied from 50 to 80% at stations 1-6, but it was 100% at those 248 stations visited in mid-late September (stations 7-9, Table 1). The coverage of melt-ponds ranged 249 from 10 to 50% [Boetius et al., 2013]. The euphotic zone (1% of incoming irradiance) was on 250 average 25 m deep and was nutrient depleted by phytoplankton consumption: i) silicate-depleted 251 at stations 1-3; ii) nitrate-depleted at stations 4 and 5; and iii) silicate, nitrate and phosphate-252 depleted at stations 6-9 [Fernández-Méndez et al., 2015]. The mixed layer was on average 22 m 253 thick and was defined by the depth where density increased from its surface value to 20% of the 254 difference between 100 m and the surface [Shaw et al., 2009] using the CTD profiles obtained 255 during the cruise (doi:10.1594/PANGAEA.802904). The winter mixed layer depth was found at 256 around 55 m [Fernández-Méndez et al., 2015] above the lower halocline (salinity range: 33.5-257 34.5) [Rudels, 2009], which reached depths down to 150 m. The potential temperature maximum 258 indicative of the Atlantic Water core was found between the depth range 180-290 m. Underneath 259 260 the Atlantic layer, Arctic intermediate waters as well as deep and bottom waters were located.

²⁴³

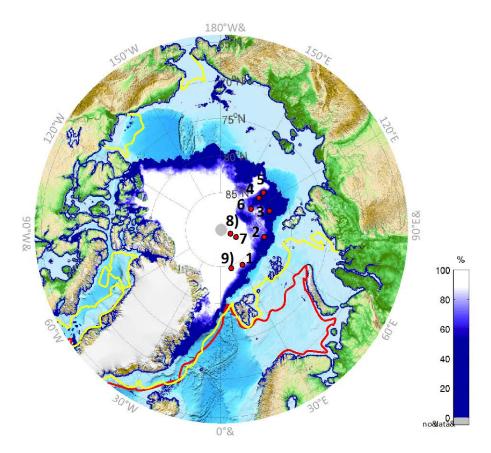


Figure 1: Location of sea-ice stations sampled during the IceArc cruise (ARK-XXVII/3, August-September 2012) (red dots). Average sea-ice concentration in September 2012. Contour lines represent the sea-ice extent in February (red) and July (yellow) 2012. Sea-ice concentration data were obtained from http://www.meereisportal.de (grant: REKLIM-2013-04) [*Spreen et al.*, 2008].

261

The Chl-a inventories in the upper 30 m of the water column were on average 8.4 ± 6.1 267 mg m⁻², with a maximum at station 2 (22.8 mg m⁻²) and a minimum at station 9 (2.5 mg m⁻²). 268 The phytoplankton community was picoplankton dominated at many stations (1, 2, 3, 5 and 6), 269 accounting for ~40-70% of the total Chl-a biomass. At those stations prasinophytes were the 270 most relevant group with a relative biomass up to 95%. Large cells dominated the community at 271 272 station 4 with a significant contribution from diatoms (44%), while nanoplankton prevailed at station 7 with a dominance of haptophytes (63%). Finally, station 9 had a similar biomass 273 274 distribution between size classes (Table 1).

The integrated in situ NPP rates in the euphotic zone, sea ice and melt ponds ranged from 0.1 mmol C m⁻² d⁻¹ at the northernmost station (8), to 5.0 mmol C m⁻² d⁻¹ at the southernmost station (5). In situ NPP was highest at the picoplankton-dominated stations (>1.3 mmol C m⁻² d⁻¹ ¹). The daily NPP estimates during one and two weeks prior to sampling were higher than the in situ estimates by a factor of 2-7 at stations 3, 4, 7, 8, while they were a factor of 3 lower at station 5. These estimates were comparable at stations 1, 2 and 6. The annual new primary production estimates compared well with the in situ, one- and two-week daily estimates from stations 1 to 6. However, north of 87°N (stations 7 and 8) the annual estimates were higher than the average of the other estimates by a factor >20 (~10-12 mmol C m⁻² d⁻¹, Table 1).

284 3.2 Total ²³⁴Th/²³⁸U and ²¹⁰Po/²¹⁰Pb

285 3.2.1 Seawater profiles

The profiles of the total activities of ²³⁴Th and ²³⁸U, and ²¹⁰Po and ²¹⁰Pb are illustrated in Figure 2.

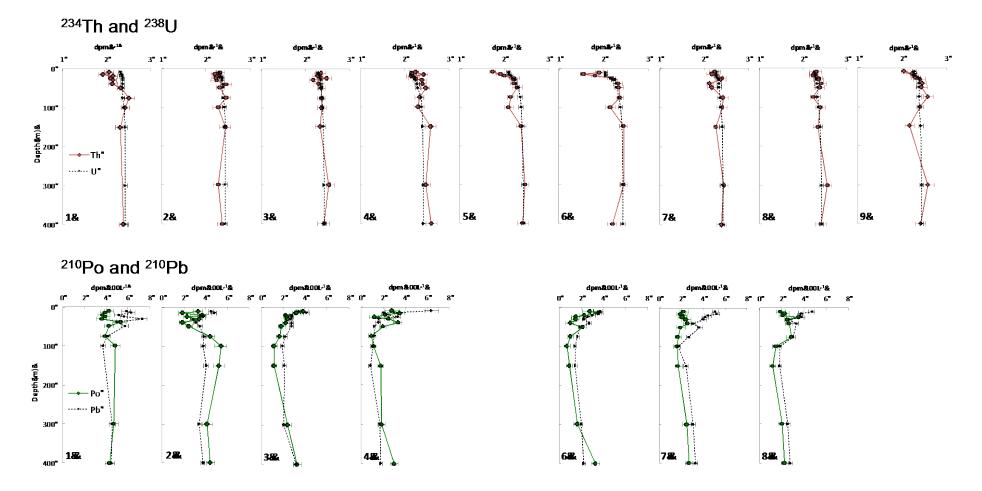


Figure 2: Vertical activity profiles for ²³⁴Th (red solid line) and ²³⁸U (dotted line) (top panels) and for ²¹⁰Po (green solid line) and ²¹⁰Pb (dotted line) (bottom panels), from 10 to 400 m depth. ²³⁸U was derived from salinity [*Owens et al.*, 2011].

The specific activities of each radionuclide ranged from 1.54 ± 0.06 to 2.59 ± 0.13 dpm 290 L^{-1} for 234 Th, 2.04 ± 0.05 to 2.44 ± 0.05 dpm L^{-1} for 238 U, 0.7 ± 0.3 to 5.4 ± 0.5 dpm 100 L^{-1} for 291 210 Po, and 0.84 \pm 0.09 to 7.3 \pm 0.4 dpm 100L⁻¹ for 210 Pb. Within the upper 25 m of the water 292 column, significant deficits of 234 Th (i.e. 234 Th/ 238 U <0.9, given uncertainties) were observed at 293 stations 1, 5 and 6, while significant deficits of 210 Po (i.e. 210 Po/ 210 Pb <0.8, given uncertainties) 294 were detected at all the stations. Below 25 m depth, deficits of ²³⁴Th were detected at one single 295 depth at stations 5 and 9 (100-150 m), but deficits of ²¹⁰Po were found at every station usually at 296 several depths (30-150 m). Excesses of 234 Th (i.e. 234 Th/ 238 U >1.1) were not observed at any 297 profile below 25 m, whereas excesses of ²¹⁰Po (i.e. ²¹⁰Po/²¹⁰Pb >1.2) were observed at four 298 stations (1, 2, 4 and 6). 299

Station 1 showed deficits of 234 Th and 210 Po within the upper 50 m: 11500 ± 2100 and 300 770 ± 120 dpm m⁻², respectively. Station 6 also showed deficits of both isotopes, down to 25 m 301 for 234 Th (160 ± 40 dpm m⁻²) and 150 m for 210 Po (930 ± 200 dpm m⁻²). At five stations (2, 3, 4, 302 7 and 8) ²³⁴Th was not significantly depleted in the upper water column. On the contrary, at those 303 stations the integrated ²¹⁰Po deficits in the upper water column (50-150 m) ranged from 130 \pm 304 150 to 1640 ± 220 dpm m⁻². The integrated excesses of ²¹⁰Po observed at stations 2 (30, 100-300 305 m) and 4 (15, 30-50, 150, 400 m) exceeded the integrated deficits observed in the surface water. 306 Finally, at stations 5 and 9 (only ²³⁴Th sampling), ²³⁴Th was in equilibrium with ²³⁸U throughout 307 the upper 400 m with only a few exceptions. 308

309

$$3.2.2^{234}$$
Th and ²¹⁰Po fluxes

The ²³⁴Th and ²¹⁰Po fluxes (F_D) are attributed to scavenging of ²³⁴Th and ²¹⁰Po onto sinking particles. The fluxes were calculated using a steady state (SS) model, neglecting advective and diffusive fluxes [*Buesseler et al.*, 1992]:

$$F_{\rm D} = \lambda_{\rm D} (A_{\rm P} - A_{\rm D})$$

where D stands for "daughter" (²³⁴Th or ²¹⁰Po) and P for "parent" (²³⁸U or ²¹⁰Pb, respectively). λ_D is the decay constant of ²³⁴Th (0.029 d⁻¹) or ²¹⁰Po (0.0050 d⁻¹), and (A_P – A_D) is the integrated daughter deficit with respect to its parent (dpm m⁻²). The fluxes calculated down to 25, 50, 100 and 150 m are listed in Table 2.

317

port	fluxes as	suming s				ons at	25, :	<u>50, 100</u>				
	Station	Depth	²³⁴ T	h flu	ixes	²¹⁰ Po fluxes						
	Station	(m)	(dpn	$(dpm m^{-2} d^{-1})$			$(dpm m^{-2} d^{-1})$					
		25	200	±	50	2.1	±	0.4				
	1	50	330	±	60	3.9	±	0.6				
	1	100	230	±	130	4.3	±	1.0				
		150	280	±	190	1.8	±	1.5				
		25	40	±	50	1.6	±	0.3				
	2	50	70	±	70	2.4	±	0.4				
	2	100	100	±	120	1.3	±	0.9				
		150	200	±	200	-2.2	±	1.5				
		25	-20	±	50	0.2	±	0.3				
	3	50	10	±	70	0.8	±	0.4				
	3	100	20	±	120	2.5	±	0.7				
		150	70	±	200	4.5	±	1.0				
		25	-70	±	40	2.2	±	0.6				
	4	50	-160	±	60	1.0	±	0.7				
	4	100	-160	±	120	0.8	±	0.8				
		150	-220	±	180	-0.4	±	1.0				
		25	170	±	40	nd						
	5	50	190	±	60	nd						
		100	440	±	110	nd						
		150	660	±	180	nd						
		25	160	±	40	0.9	±	0.3				
	C	50	130	±	60	2.1	±	0.4				
	6	100	180	±	120	3.2	±	0.7				
		150	310	±	190	4.7	±	1.0				
		25	20	±	50	3.4	±	0.4				
	7	50	90	±	70	5.0	±	0.5				
	7	100	90	±	140	7.2	±	0.8				
		150	160	±	200	8.2	±	1.1				
		25	0	±	60	2.5	±	0.4				
	o	50	-50	±	80	3.0	±	0.5				
	8	100	0	±	120	3.7	±	0.9				
		150	20	±	190	4.9	±	1.1				
	-	25	90	±	40	nd						
	0	50	60	±	70	nd						
	9	100	-110	±	140	nd						
		150	70	±	190	nd						
				-								

318 **Table 2:** ²³⁴Th and ²¹⁰Po export <u>fluxes assuming steady state conditions at 25, 50, 100</u> and 150 m.

319 nd = no available data

The ²³⁴Th fluxes were negligible or very low at 5 out of 9 stations (2, 3, 4, 7 and 8). At stations 1, 5 and 6, the ²³⁴Th fluxes averaged 175 ± 19 dpm m⁻² d⁻¹ at 25 m, 210 ± 100 dpm m⁻² d⁻¹ ¹ at 50 m, 280 ± 140 dpm m⁻² d⁻¹ at 100 m and 400 ± 200 dpm m⁻² d⁻¹ at 150 m. At station 9 the already low ²³⁴Th flux at 25 m (90 ± 40 dpm m⁻² d⁻¹) became negligible in deeper waters. The ²¹⁰Po fluxes were significant at all the stations, averaging 1.8 ± 1.1 dpm m⁻² d⁻¹ at 25 m, 2.6 ± 1.5 dpm m⁻² d⁻¹ at 50 m, 3 ± 2 dpm m⁻² d⁻¹ at 100 m and 3 ± 3 dpm m⁻² d⁻¹ at 150 m. The ²¹⁰Po fluxes did not decrease with depth at the majority of stations (1, 3, 6, 7 and 8), whereas at stations 2 and 4 the fluxes became negligible at 100-150 m.

328 3.3. Particulate fraction

Particulate ²³⁴Th, ²¹⁰Po, ²¹⁰Pb, and organic carbon and nitrogen concentrations in large particles are given in Table 3, as well as the ²¹⁰Po/²¹⁰Pb and molar C/N ratios.

The mean 234 Th activities in particles decreased with depth, ranging from ~1 dpm $100L^{-1}$ 331 at 25 m to ~0.3 dpm $100L^{-1}$ at 150 m. ²¹⁰Po activities were on average ~0.04 dpm $100L^{-1}$ at 25 m 332 and ~ $0.02 \text{ dpm } 100\text{L}^{-1}$ below that depth, while ²¹⁰Pb activities were ~ $0.06 \text{ dpm } 100\text{L}^{-1}$ at 25 and 333 50 m, and ~ 0.02 dpm $100L^{-1}$ at 100 and 150 m. The variation between stations was large, with 334 deviations from those means of >50% for 234 Th, >80% for 210 Po and >100% for 210 Pb. Only 335 about 0.3% of the total activity of ²³⁴Th in water, 1.1% of ²¹⁰Po and 1.7% of ²¹⁰Pb was associated 336 with large particles. The maximum particulate activities were found at stations 2 and 3 and the 337 minimum at stations 7 and 8 (negligible in some instances for ²¹⁰Po and ²¹⁰Pb). The ²¹⁰Po/²¹⁰Pb 338 ratios ranged from 0.2 to 6 (average: 1.2 ± 1.4 , n = 18), varying considerably between stations 339 and depths. 340

The POC and PON concentrations were highest at 25 m, averaging 0.23 ± 0.08 and $0.028 \pm 0.011 \mu$ mol L⁻¹ (n = 8), respectively. Below that depth the concentrations decreased by a factor of 3. The mean C/N ratio was similar at all the investigated depths, averaging 8.8 ± 1.9 (n = 34).

Table 4 displays the POC/²³⁴Th and POC/²¹⁰Po ratios (C/Th and C/Po) at 25, 50, 100 and 150 m. The average ratios at the different horizon depths ranged from 17 to 40 μ mol dpm⁻¹ for C/Th and from 300 to 1100 μ mol dpm⁻¹ for C/Po. The ratios did not change significantly with depth (Kruskal-Wallis test, p >0.05).

3 1	Table 3: Pa	rticulate ²	³⁴ Th, ²¹⁰ Po, ²	²¹⁰ Pb, and organic	carbon and nitrogen	concentrations, and ²	$^{10}\text{Po}/^{210}\text{Pb}$ and	molar C/N rati	os in particles :	>53 µm.
		Station	Depth (m)	Part. ²³⁴ Th	Part. ²¹⁰ Po	Part. ²¹⁰ Pb	²¹⁰ Po/ ²¹⁰ Pb	POC	PON	C/N
		Station	Depth (III)	(dpm 100L ⁻¹)	(dpm 100L ⁻¹)	(dpm 100L ⁻¹)	P0/ P0	$(\mu mol C L^{-1})$	$(\mu mol N L^{-1})$	(mol:mo
			15	0.45 ± 0.03	$0.026 ~\pm~ 0.003$	0.0042 ± 0.0012	6 ± 2	0.25	0.038	6.6
		1	50	nd	nd	nd	nd	nd	nd	nd
		1	90	$0.335 ~\pm~ 0.019$	$0.008 ~\pm~ 0.003$	0.0144 ± 0.0018	0.6 ± 0.2	0.074	0.0092	8.1
			190	$0.138 ~\pm~ 0.008$	0.011 ± 0.002	0.0057 ± 0.0011	1.8 ± 0.5	0.034	0.0054	6.3
			25	nd	nd	nd	nd	nd	nd	nd
		2	50	1.71 ± 0.12	$0.032 ~\pm~ 0.006$	0.031 ± 0.003	1.0 ± 0.2	0.23	0.034	6.7
		2	100	1.86 ± 0.12	$0.120 ~\pm~ 0.011$	$0.073 ~\pm~ 0.004$	1.6 ± 0.2	0.12	0.020	6.3
			150	0.89 ± 0.06	0.050 ± 0.007	$0.055 ~\pm~ 0.004$	$0.91 \hspace{0.1in} \pm \hspace{0.1in} 0.14$	0.042	0.0068	6.1
			25	1.63 ± 0.09	$0.066 ~\pm~ 0.010$	$0.217 ~\pm~ 0.010$	$0.30~\pm~0.05$	0.27	0.033	8.1
		3	50	1.60 ± 0.11	$0.054 \hspace{0.2cm} \pm \hspace{0.2cm} 0.014$	$0.221 ~\pm~ 0.011$	$0.24 ~\pm~ 0.07$	0.15	0.020	7.8
		5	100	$0.215 ~\pm~ 0.012$		0.039 ± 0.003	-	0.032	0.0043	7.4
			150	0.51 ± 0.02	$0.035 ~\pm~ 0.006$	0.040 ± 0.004	0.9 ± 0.2	0.087	0.010	8.5
			25	$0.55 ~\pm~ 0.03$	0.025 ± 0.003	0.044 ± 0.003	0.57 ± 0.08	0.28	0.029	9.8
		4	50	$0.47 ~\pm~ 0.03$	$0.015 ~\pm~ 0.003$	$0.031 ~\pm~ 0.002$	$0.47 ~\pm~ 0.12$	0.036	0.0056	6.4
			100	$0.276 ~\pm~ 0.010$	<0.003	0.0152 ± 0.0016	-	0.097	0.014	7.0
			150	0.43 ± 0.02	0.007 ± 0.003	0.023 ± 0.002	0.31 ± 0.15	0.058	0.0078	7.5
			25	1.50 ± 0.10	nd	nd	nd	0.38	0.047	8.0
		5	50	0.88 ± 0.06	nd	nd	nd	0.14	0.016	8.5
			100	0.414 ± 0.016	nd	nd	nd	0.097	0.011	9.2
			150	0.58 ± 0.03	nd	nd	nd	0.061	0.0075	8.2
			25	1.25 ± 0.08	0.077 ± 0.006	0.106 ± 0.005	0.73 ± 0.06		0.014	11.1
		6	50	0.250 ± 0.014	0.006 ± 0.003	0.026 ± 0.002	0.21 ± 0.12	0.025	0.0025	10.1
			100	0.094 ± 0.005	< 0.003	0.008 ± 0.002	-	0.015	0.0014	10.1
			150	0.096 ± 0.009	0.013 ± 0.003	0.0059 ± 0.0016		0.012	0.00090	13.6
			25	0.42 ± 0.02	0.0126 ± 0.0018	0.0076 ± 0.0014	1.7 ± 0.4	0.23	0.023	10.0
		7	50	0.062 ± 0.007		<0.003	-	0.029	0.0028	10.6
			100	0.061 ± 0.009	<0.003	0.0040 ± 0.0018	-	0.088	0.0074	11.9
			150	0.078 ± 0.006	0.009 ± 0.003	<0.003	-	0.036	0.0029	12.4
			25 50	0.51 ± 0.02	0.009 ± 0.003	0.010 ± 0.002	0.9 ± 0.3	0.16	0.020	8.1
		8	50	0.286 ± 0.016	0.008 ± 0.003 < 0.003	0.026 ± 0.003	0.31 ± 0.13	0.049	0.0051	9.6
			100	0.133 ± 0.014		0.012 ± 0.003	-	0.080	0.0070	11.5
			150	0.100 ± 0.008		0.0040 ± 0.0018	-	0.032	0.0040	8.2
			25 50	1.34 ± 0.10	nd	nd	nd	0.15	0.020	7.4
		9	50	0.64 ± 0.03	nd	nd	nd	0.088	0.0091	9.7
		-	100	0.229 ± 0.011	nd	nd	nd	0.069	0.0077	9.0
			150	0.18 ± 0.02	nd	nd	nd	0.059	0.0064	9.3

Table 3: Particulate ²³⁴Th, ²¹⁰Po, ²¹⁰Pb, and organic carbon and nitrogen concentrations, and ²¹⁰Po/²¹⁰Pb and molar C/N ratios in particles >53 μm.

- 349 nd = no available data
- 350 3.4 POC fluxes

The POC fluxes were calculated multiplying the ²³⁴Th and ²¹⁰Po fluxes derived from the SS model by the C/Th and C/Po ratios in large particles, respectively (Table 4). **Table 4:** Particulate POC/²³⁴Th and POC/²¹⁰Po ratios (C/Th and C/Po), and POC fluxes derived from ²³⁴Th and ²¹⁰Po.

anc	1 P0.													1
	Station	Depth	(C/Th	ı.		C/Po			POC fluxes (mmol C $m^{-2} d^{-1}$)				
_	Station	(m)					$(\mu mol C dpm^{-1})$				rived			rived
		15	56	±	4	970	\pm	100	7	\pm	2	1.2	±	0.4
	1	50	nd			nd			nd			nd		
	1	90	22.1	±	1.3	900	±	400	5	±	3	4	±	2
_		190	25.0	±	1.5	330	±	70	10	±	6	0.1	±	0.7
		25	nd			nd			nd			nd		
	2	50	13.2	±	0.9	700	±	120	0.9	±	0.9	1.7	±	0.4
	2	100	6.7	±	0.5	104	±	9	0.7	±	0.8	0.13	±	0.09
_		150	4.7	±	0.3	83	±	12	0.9	±	0.9	-0.19	±	0.13
		25	16.3	±	0.9	400	±	60	-0.4	\pm	0.8	0.06	±	0.14
	3	50	9.6	±	0.6	280	±	80	0.1	±	0.7	0.23	±	0.14
	5	100	14.8	±	0.8	-			0	±	2	-		
_		150	16.9	±	0.8	250	±	40	1	±	3	1.1	±	0.3
		25	51	±	3	1140	±	140	-4	\pm	2	2.5	±	0.7
	4	50	7.7	±	0.4	250	±	60	-1.2	\pm	0.5	0.3	±	0.2
	-	100	35.2	±	1.3	-			-6	±	4	-		
_		150	13.6	±	0.6	800	±	400	-3	±	2	-0.3	±	0.9
		25	25.1	±	1.7	nd			4.3	\pm	1.0	nd		
	5	50	15.6	±	1.0	nd			2.9	±	0.9	nd		
	5	100	23.5	±	0.9	nd			10	±	3	nd		
_		150	10.5	±	0.5	nd			7	±	2	nd		
		25	12.2	±	0.8	197	±	14	1.9	\pm	0.5	0.18	±	0.07
	6	50	10.1	±	0.5	500	±	200	1.3	±	0.6	1.0	±	0.5
	0	100	15.5	±	0.8	-			3	±	2	-		
_		150	12.7	±	1.2	90	±	20	4	±	2	0.44	±	0.13
		25	54	±	3	1800	±	300	1	±	3	6.3	±	1.1
	7	50	48	±	6	-			4	±	3	-		
	,	100	150	±	20	-			10	\pm	20	-		
_		150	47	±	4	390	±	110	7	±	9	3.2	±	1.0
		25	32.3	±	1.4	1900	±	600	0	±	2	4.8	±	1.6
	8	50	17.1	±	1.0	600	±	300	-0.9	\pm	1.3	1.8	±	1.3
	0	100	60	±	6	-			0	±	7	-		
_		150	32	±	3	-			1	<u>+</u>	6	-		
_		25	11.2	±	0.8	nd			1.0	±	0.5	nd		
	9	50	13.7	±	0.8	nd			0.9	\pm	0.9	nd		
	9	100	30.2	±	1.4	nd			-3	±	4	nd		
		150	32	±	4	nd			2	±	6	nd		
-														

355 nd = no available data.

The ²³⁴Th-derived POC fluxes ranged from negligible to 10 mmol C m⁻² d⁻¹ and averaged 1.3 to 4 mmol C m⁻² d⁻¹ at 25, 50, 100 and 150 m. The ²¹⁰Po-derived POC fluxes ranged from negligible to 6.3 mmol C m⁻² d⁻¹ and were on average from 0.8 to 3 mmol C m⁻² d⁻¹ at the same depths. The POC fluxes estimated using the two proxies were not significantly different considering all depths together, or each depth individually (Wilcoxon test, p > 0.05).

362 4 Discussion

In this study we have used two pairs of radionuclides, 234 Th/ 238 U and 210 Po/ 210 Pb, as tools to estimate POC fluxes in the Eurasian Basin of the Arctic Ocean in summer 2012. Deficits of 234 Th and 210 Po are attributed to particle export, while the excesses of these radionuclides evidence their release from sinking particles by means of remineralization or particle disaggregation into the suspended pool. Their simultaneous application allows integrating a temporal scale over a span of weeks (234 Th mean life = 35 days) to months (210 Po mean life = 200 days).

370 $4.1^{234} \text{Th}^{238} \text{U}$

371

4.1.1 ²³⁴Th export fluxes

²³⁴Th export fluxes were calculated using a SS model because the stations were not reoccupied during the expedition. Yet in a review study *Savoye et al.* [2006] did not find significant differences between the SS and non-steady state (NSS) models at low flux rates (<800 dpm m⁻² d⁻¹), which is the case of the present work.

Significant ²³⁴Th fluxes within the upper 150 m of the water column were 376 obtained at stations 1, 5 and 6, and at stations 7 and 9 only at one single depth (Table 2). 377 The 234 Th fluxes ranged from negligible to 660 dpm m⁻² d⁻¹, averaging 120 ± 140 dpm 378 $m^{-2} d^{-1}$ (n = 36). Our results are one order of magnitude lower than the ²³⁴Th flux 379 average reported by Le Moigne et al. [2013b] for the world ocean (1200 \pm 900 dpm m⁻² 380 d^{-1} ; 75-210 m; n = 421). Previous research conducted in the central Arctic, mainly 381 during summer, has also revealed low export fluxes escaping from the ocean surface 382 (Figure 3). Cai et al. [2010] reported an average of 90 ± 300 dpm m⁻² d⁻¹ (n = 26) in the 383 most extensive study of ²³⁴Th over the central basins to date, and *Moran et al.* [1997] 384 and *Gustafsson and Andersson* [2012] reported similar flux averages of 190 ± 140 dpm 385 $m^{-2} d^{-1}$ (n = 7) and 130 ± 100 dpm $m^{-2} d^{-1}$ (n = 3), respectively. Le Moigne et al. [2015] 386 reported 234 Th fluxes of 140 ± 210 dpm m⁻² d⁻¹ in the ice covered Fram Strait, which is 387 also in line with our results. Nevertheless, other studies have reported high ²³⁴Th export 388 fluxes (>2000 dpm m⁻² d⁻¹) at specific locations in the Canada Basin [Ma et al., 2005], 389 although they are more typical of the shelf environment [e.g. Coppola et al., 2002; 390 Lepore et al., 2007] (Figure 3). Overall, the ²³⁴Th flux data presented here and the 391

- 392 limited data available to date illustrate the central Arctic basins as deserts in terms of
- 393 particle export during summer.

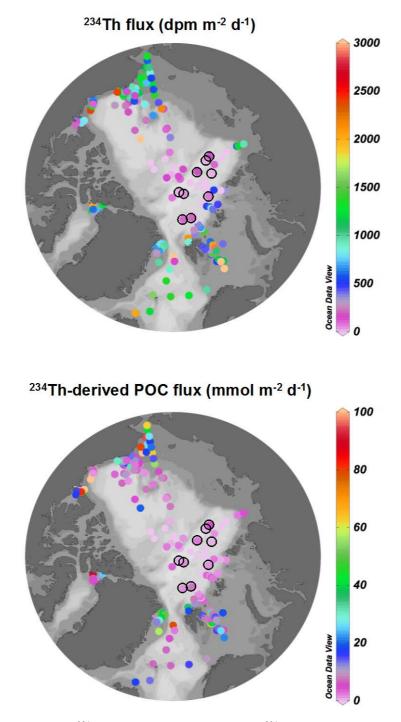


Figure 3: Compilation of ²³⁴Th flux data (upper panel) and ²³⁴Th-derived POC flux data (lower panel)
from the Arctic Ocean (236 stations) [*Cochran et al.*, 1995b; *Moran et al.*, 1997, 2005; *Moran and Smith*,
2000; *Amiel et al.*, 2002; *Coppola et al.*, 2002; *Baskaran et al.*, 2003; *Chen et al.*, 2003; *Ma et al.*, 2005; *Trimble and Baskaran*, 2005; *Lepore et al.*, 2007; *Lalande et al.*, 2007, 2008; *Amiel and Cochran*, 2008; *Yu et al.*, 2010, 2012; *Cai et al.*, 2010; *Gustafsson and Andersson*, 2012; *Le Moigne et al.*, 2015; this
study]. Black circles indicate the results obtained in this study. The depth horizon taken to calculate the
POC export fluxes ranges from 25 to 200 m.

4.1.2 ²³⁴Th-derived POC export fluxes

The mean 234 Th-derived POC export fluxes measured in the upper 150 m were 3 ± 3 403 mmol C m⁻² d⁻¹ (n = 34), with a maximum of 10 mmol C m⁻² d⁻¹ (Table 4). At the bottom of the 404 euphotic zone (~25 m) the fluxes ranged from negligible to 7 mmol C m⁻² d⁻¹ (average: 2 ± 2 405 mmol C m⁻² d⁻¹, n = 8). These results are in very good agreement with the POC fluxes measured 406 with cylindrical sediment traps (HydroBios, Kiel, Germany) deployed under the ice during 407 periods of 24-53 hours from station 1 to 9 [Lalande et al., 2014]. The sediment trap results 408 ranged from 0.4 to 9 mmol C m⁻² d⁻¹ (average: 3 ± 3 mmol C m⁻² d⁻¹, n = 9). The in situ NPP 409 rates showed positive correlations with ²³⁴Th fluxes at 25 m (p <0.05; Spearman correlation 410 coefficient, $\rho = 0.83$; n = 8), ²³⁴Th-derived POC fluxes at 25 m (p <0.05; $\rho = 0.78$; n = 7) and 411 sediment trap-derived POC fluxes at 25 m (p < 0.05; $\rho = 0.83$; n = 8), which indicates enhanced 412 413 particle fluxes with increasing NPP. Our results also compare well with previous literature values from sediment traps deployed at 150-175 m north of the Laptev Sea continental margin in 414 August-September during the years 95/96 and 05/06 (~0.5-2.5 mmol C m⁻² d⁻¹) [Fahl and 415 Nöthig, 2007; Lalande et al., 2009] and ²³⁴Th-derived POC fluxes in the central Arctic (Figure 416 3). Cai et al. [2010] documented very low POC export fluxes (average: $0.2 \pm 1.0 \text{ mmol C m}^{-2} \text{ d}^{-1}$. 417 n = 26) across the deep Arctic, suggesting that they were a consequence of low biological 418 productivity. Our low POC export flux estimates are in good agreement with the low NPP 419 observed in the present study within the ²³⁴Th time window (in situ, one- and two-week 420 estimates; $\leq 5 \mod C \mod^{-2} d^{-1}$; Table 1). 421

422

4.1.3 Relationships with phytoplankton community

We did not find any significant relationship between the ²³⁴Th data (particulate ²³⁴Th 423 activity, ²³⁴Th fluxes and ²³⁴Th-derived POC fluxes) and the phytoplankton size structure at the 424 sampling time, although two correlations were obtained with regards to the phytoplankton 425 composition. The relative biomass of prasinophytes_1 was positively correlated with ²³⁴Th fluxes 426 $(p < 0.05; \rho = 0.75; n = 8)$ and ²³⁴Th-derived POC fluxes $(p < 0.05; \rho = 0.77; n = 7)$ at 25 m. This 427 suggests that prasinophytes 1 would have contributed significantly to vertical export fluxes 428 during the late summer in 2012 when picoplankton, and particularly prasinophytes, were the 429 predominant group in terms of biomass (Prasinophytes 1 and 2, Table 1). Prasinophytes are 430 green algae that can be usually found in the eukaryotic picoplankon fraction. A molecular study 431

by *Metfies et al.* [2016] corroborates the biomass dominance of picoplankton in the upper water 432 column during our expedition and identifies the prasinophyte *Micromonas* spp. as its major 433 constituent. Our finding is in line with recent observations that reveal that small cells are 434 important contributors to POC export fluxes in diverse oceanic regimes [e.g. Richardson and 435 Jackson, 2007; Lomas and Moran, 2011; Durkin et al., 2015; Mackinson et al., 2015; Puigcorbé 436 et al., 2015]. Prasinophytes, including Micromonas spp., are common in the central Arctic 437 [Booth and Horner, 1997; Sherr et al., 2003; Zhang et al., 2015], and are considered to be 438 among the most abundant photosynthetic cells in pan-Arctic waters [Lovejoy et al., 2007]. 439 Genetic analyses in trap samples revealed that prasinophytes contributed to downward fluxes in 440 the Sargasso Sea [Amacher et al., 2013], but to our knowledge, this has not been observed before 441 in Arctic waters. It is relevant to note that neither molecular nor pigment techniques inform about 442 whether they sink as single cells or as part of other export pathways. 443

444 The pathways by which picoplankton cells can be removed from the ocean surface are fundamentally: i) zooplankton grazing and subsequent incorporation into fecal pellets [Waite et 445 446 al., 2000; Wilson and Steinberg, 2010]; ii) adhesion into mucous nets formed by gelatinous zooplankton, such as pteropods, and later settling [Noji et al., 1997]; iii) inclusion into marine 447 snow via particle aggregation, which is enhanced by transparent exopolymer particles (TEP) 448 [Passow, 2002]. Passive sinking of fecal pellets could be a significant pathway for particle export 449 in the central Arctic where zooplankton exert a strong grazing pressure on algae, preventing their 450 biomass accumulation and sedimentation [Olli et al., 2007]. Indeed, the copepod food demand 451 during our cruise was estimated to be similar to the in situ NPP rates [David et al., 2015], leaving 452 a small fraction of algae available for direct export. Yet Lalande et al. [2014] estimated that only 453 up to 7.5% of the POC collected by traps at 25 m consisted of fecal pellets. Trap samples also 454 consisted of marine snow, debris, appendicularian houses, animal body parts and very sticky 455 material, even though their relative importance in POC content was not quantified (C. Lalande, 456 pers. comm.). Copepods clearly dominated the zooplankton community with regards to 457 abundance, whereas pteropods, ctenophores and appendicularians, which are prone to produce 458 mucous, represented less than 3-5% of the total zooplankton abundance either beneath the ice 459 [David et al., 2015] or within the upper 50 m [Ehrlich, 2015]. However, ctenophores and 460 appendicularians dominated the under-ice zooplankton biomass at some stations, which could 461 have contributed notably to the export of mucous (C. David, pers. comm.). Moreover, sea-ice 462

algal aggregates of the centric diatom *Melosira arctica* and pennate diatoms were observed at all 463 the stations [Fernández-Méndez et al., 2014]. They reached abundances up to 16 ind/m² and 464 extraordinary sizes (diameter mean: 2.1-4.1 cm), although they showed a highly patchy 465 distribution [Katlein et al., 2014]. The aggregates were associated with mucous matrices that 466 increased their stickiness and, at the same time, their predisposition to aggregation [Fernández-467 Méndez et al., 2014]. Indeed, Melosira arctica was intercepted using sediment traps deployed at 468 25 m at some stations [Lalande et al., 2014], confirming that it was part of the sinking pool. 469 Taken all together, sea-ice algal aggregates and zooplankton-derived material might have acted 470 as carriers of picoplankton cells from the ocean surface to depth (Figure 4b). 471

472

 $4.2^{210} Po/^{210} Pb$

473

4.2.1 ²¹⁰Po and ²¹⁰Pb activities

²¹⁰Po activities were lower than those of ²¹⁰Pb at every station in the upper 50-150 m, 474 indicating export driven by sinking particles, while excess ²¹⁰Po was observed at several depths 475 throughout the upper 400 m at stations 2 and 4, suggesting remineralization or particle 476 disaggregation (Figure 2). At stations 2 and 4 the integrated excess surpassed the integrated 477 deficit at 150 m and below, which can be explained by: i) a previous large export event that 478 occurred at the study sites, and/or ii) advection of waters that were enriched in ²¹⁰Po as 479 consequence of a previous export event [Stewart et al., 2007]. Thus, the assumption of SS and/or 480 neglecting the advective term would have added uncertainty to our flux estimates of ²¹⁰Po. We 481 note that the ²¹⁰Po flux estimates are subject to be affected by NSS conditions or advection 482 transport processes to a larger extent than the ²³⁴Th flux estimates due to the longer half-life of 483 ²¹⁰Po. 484

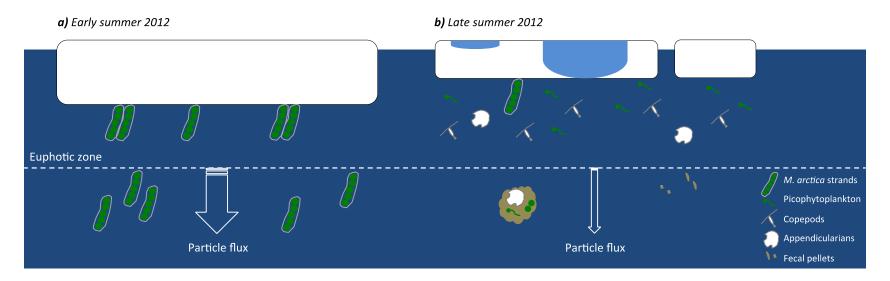
Very few studies have investigated the distribution of ²¹⁰Po and/or ²¹⁰Pb in the Arctic water column [*Moore and Smith*, 1986; *Cochran et al.*, 1995a; *Smith and Ellis*, 1995; *Roberts et al.*, 1997; *Smith et al.*, 2003; *Lepore et al.*, 2009; *Chen et al.*, 2012]. The ²¹⁰Pb and ²¹⁰Po activities presented here are comparable to the wide activity range reported by those studies, including shelf and basin areas.

In the Arctic, sea ice intercepts and accumulates atmospheric fluxes of chemical species, such as ²¹⁰Pb, during its transit through the ocean [*Masqué et al.*, 2007; *Cámara-Mor et al.*, 2011] and, therefore, sea ice melting may increase ²¹⁰Pb activities in surface waters where that 493 occurs [Roberts et al., 1997; Masqué et al., 2007; Chen et al., 2012]. One might wonder whether sea ice melting may significantly impact the ²¹⁰Po and ²¹⁰Pb activities in seawater and, thus, 494 affect the use of the ²¹⁰Po proxy. Data on ²¹⁰Pb and ²¹⁰Po activities in entire sea-ice cores 495 collected during the same expedition (results not shown) show that the ²¹⁰Po/²¹⁰Pb ratios were 496 <0.5. indicating ²¹⁰Pb enrichment in sea ice, and consistent with the dominance of first-year ice 497 [Masqué et al., 2007]. Given the inventories of both isotopes in sea-ice cores, even with 498 complete melting of sea ice, the ²¹⁰Po/²¹⁰Pb ratio in the upper 25 m of the water column would 499 have not changed or would have decreased as much as 10%. Since this change is relatively small, 500 we are confident that the principal cause of the ²¹⁰Po deficit in the upper water column was its 501 preferential removal via particle scavenging with respect to ²¹⁰Pb. 502

503

$4.2.2^{210}$ Po export fluxes

The ²¹⁰Po export fluxes in the upper 150 m ranged from negligible to 8.2 dpm m⁻² d⁻¹, 504 averaging 3 ± 2 dpm m⁻² d⁻¹ (n = 28, Table 2). The fluxes obtained in this study are very low in 505 comparison to other studies conducted in other regions of the world ocean [Shimmield et al., 506 1995; Kim and Church, 2001; Friedrich and Rutgers van der Loeff, 2002; Murray et al., 2005; 507 Stewart et al., 2007a; Buesseler et al., 2008; Verdeny et al., 2008; Le Moigne et al., 2013a], 508 which reported fluxes from 5 to >100 dpm m⁻² d⁻¹. However, the ²¹⁰Po fluxes were significant at 509 every station and at most of the investigated depths, in contrast to ²³⁴Th fluxes, which were only 510 measurable at more than one depth at three stations (Table 2). Given the time scales of both 511 tracers, ²¹⁰Po would track particle export for the entire productive season, whereas ²³⁴Th 512 distribution misses events that occurred >1 month before sampling. Thus, the more common 513 ²¹⁰Po depletion than that of ²³⁴Th in the upper water column suggests that the magnitude of 514 particle export fluxes was more important before July/August 2012 than in the weeks prior to 515 and during the sampling (Figure 4). 516



518 Figure 4: Scheme of the magnitude and composition of the particle fluxes in the central Arctic during the early (a) and late summer (b) in 2012 based on results

from the present study and others [*Boetius et al.*, 2013; *Lalande et al.*, 2014; *David et al.*, 2015; *Fernández-Méndez et al.*, 2015] (see sections 4.1 and 4.2 for further details). Symbols are not drawn to scale.

Boetius et al. [2013] revealed the presence of vast deposits of sea-ice algal aggregates on 521 the seafloor at the majority of stations, which would have been exported from the ocean surface 522 earlier in the season, particularly before June at stations 4, 5 and 6 as suggested by the large body 523 size and fecundity of the deep-sea holothurians that fed on the algae. The aggregates were mainly 524 composed of *Melosira arctica* [Boetius et al., 2013] that can form long strands hanging from the 525 ice bottom, sometimes up to 6 m long [Melnikov and Bondarchuk, 1987], allowing a rapid 526 sinking throughout the water column once detached. Boetius et al. [2013] estimated that algae 527 covered up to 10% of the seafloor by means of high-resolution pictures, accounting for a median 528 of 750 mmol C m⁻² (\pm 50%). This POC inventory of algae was obtained by applying a cell 529 volume to carbon ratio (0.15 pg C μ m³) and a fixed thickness of the algal cover (0.01 m). This 530 supports the ²¹⁰Po evidence that the peak of export in the study area occurred in early summer 531 and sheds light on the composition of a major part of the sinking pool (Figure 4a). It was 532 estimated that diatoms were responsible for at least 45% of the total primary production in 2012 533 [Boetius et al., 2013], indicating that the phytoplankton community varied over the productive 534 season, since diatoms did not contribute much to the Chl-a biomass during our cruise (~20%, 535 Table 1), when surface waters were silicate-depleted in most of the study area. Previous studies 536 with sediment traps also revealed that highest fluxes in the central Arctic occur mainly in June-537 August [Fahl and Nötig 2007, Lalande et al. 2009]. 538

539

4.2.3 Relationships with sea-ice conditions

There were significant relationships between the sea-ice conditions and the ²¹⁰Po-derived 540 fluxes. Sea-ice concentration was positively correlated with both ²¹⁰Po fluxes (p < 0.01; $\rho = 0.92$; 541 n = 7) and $^{210}\text{Po-derived}$ POC fluxes (p <0.05; ρ = 0.91; n = 6) at 25 m. Indeed, the stations 542 located north of 87°N and covered by multi-year ice (stations 7 and 8) showed the strongest 543 depletion of ²¹⁰Po within the upper 400 m (Figure 2), and the highest annual NPP rates (Table 1) 544 and seafloor algal coverage [Boetius et al., 2013]. This suggests that primary production and 545 particle export were more important under heavy sea-ice conditions than under partially ice 546 covered stations and first-year ice, also suggesting that ²¹⁰Po tracked, to some extent, the massive 547 algal export that occurred earlier in 2012. On the contrary, at stations with heavy sea-ice 548 conditions we found the minimum in situ NPP rates (Table 1) and ²³⁴Th in equilibrium with ²³⁸U 549 throughout the upper water column (Figure 2), indicating low or negligible primary production 550

and particle export fluxes during the late summer.

The results presented here, combined with those from *Boetius et al.* [2013], show that the 552 central Arctic underwent significant changes during the productive season in terms of primary 553 production, phytoplankton composition and export fluxes during the record low of sea ice in 554 2012. This has implications for the use of ²¹⁰Po as a tracer: the depth distribution of total ²¹⁰Po 555 activity likely changed with time (NSS conditions) and the sinking material collected during the 556 survey probably did not cause the observed ²¹⁰Po depletion in the upper water column. Actually, 557 ²¹⁰Po activities in large particles collected at the time of sampling were inversely correlated with 558 ²¹⁰Po export fluxes at 25 m (p <0.05; $\rho = -0.89$; n = 6). The SS model would tend to smooth out 559 episodic export events that took place earlier in the season, and hence underestimate the mean 560 ²¹⁰Po fluxes and ²¹⁰Po-derived POC fluxes on a seasonal scale. On the other hand, we measured 561 C/Po ratios in particles that fall in the upper range of previous values (see review by Verdeny et 562 563 al. [2009]). Stewart et al. [2007] showed that C/Po ratios varied according to the sinking material composition as follows: degraded material > fresh phytoplankton > fecal pellets. We also found 564 in some instances particulate ²¹⁰Po/²¹⁰Pb ratios below one (Table 3), which is inconsistent with 565 the ²¹⁰Po deficiency observed in surface waters. Particle types that may potentially explain low 566 ²¹⁰Po/²¹⁰Pb ratios could be: particles remineralized by chemical and biological processes [*Stewart* 567 et al., 2007b], fecal material [Stewart et al., 2005; Rodriguez y Baena et al., 2007], picoplankton 568 aggregates [Stewart et al., 2010], and substrates rich in transparent exopolymer particles 569 [*Quigley et al.*, 2002]. Overall, if the sinking pool responsible for ²¹⁰Po scavenging had different 570 C/Po ratios with respect to that collected at the sampling time, the ²¹⁰Po-derived POC fluxes 571 obtained in this study would not be fully representative of the fluxes that occurred in the 572 productive season in 2012. 573

5744.3 Export efficiency

575 We have estimated the export efficiency by dividing the POC export fluxes derived from 576 ²³⁴Th and ²¹⁰Po at 25 m (i.e. ~bottom of the euphotic zone) by different estimates of NPP that 577 encompass daily, weekly and annual time scales (Table 5).

578	Table 5: Export efficiency according to the ²³⁴ Th and ²¹⁰ Po proxies estimated using different estimates of daily NPP (in situ, one and two weeks before sampling
579	and annual new primary production; see text for further details).

Station	Export efficiency (%)											
Station	In situ			Or	One week			o we	eks	Annual		
Th proxy												
1^{a}	>100			>100			>100			>100		
2 ^b	30	±	40	40	±	40	30	±	40	20	±	20
3	0			0			0			0		
4	0			0			0			0		
5	90	±	20	>100			>100			77	±	18
6	80	±	20	100	±	30	80	±	20	60	±	17
7	>100			>100			>100			10	±	20
8	0			0			0			0		
9	nd			nd			nd			13	±	6
Po proxy												
1^{a}	37	±	12	53	±	17	50	±	16	37	±	12
2 ^b	64	±	16	74	±	19	67	±	17	36	±	9
3	5	±	11	3	±	6	3	±	6	3	±	7
4	>100			70	±	20	80	±	20	90	\pm	30
5	nd			nd			nd			nd		
6	8	±	3	9	\pm	4	8	±	3	6	±	2
7	>100			>100			>100			53	±	9
8	>100			>100			>100			48	±	16
9	nd			nd			nd			nd		

nd = no available data. The values in italics have relative uncertainties $\geq 100\%$.^a POC fluxes used to estimate the export efficiency were measured at 15 m instead of 25 m.^b POC fluxes used to estimate the export efficiency were measured at 50 m instead of 25 m. 580

581

Considering the in situ NPP rates, the export efficiency varied widely over the study site, 582 from 0 to >100%, averaging 50 \pm 50% (n = 8) and 60 \pm 40% (n = 7) for the ²³⁴Th and ²¹⁰Po 583 proxies, respectively. The export efficiency calculated using the fluxes measured with sediment 584 585 traps [Lalande et al., 2014] was >100% at 6 out of 8 stations. Export efficiencies over 100% suggest that primary production that occurred earlier in the season contributed to the export 586 fluxes measured (i.e. temporal decoupling between production and export). In order to cover 587 longer time scales of NPP, we have also used estimates that integrate one and two weeks before 588 sampling and the entire productive season (see section 3.1.2). The increase in daily NPP 589 observed between the in-situ and the weekly estimates at stations 3, 4, 7 and 8, only changed 590 significantly the export efficiency at station 4 (210 Po proxy), obtaining estimates of ~70% (Table 591 5). Export efficiencies over 100% were still observed in several instances, indicating that the lag 592 593 between production and export was longer than two weeks. On the contrary, the export efficiencies decreased by about 40% applying the annual NPP estimates (²³⁴Th: $30 \pm 40\%$, n = 9; 594 ²¹⁰Po: 40 \pm 30%, n = 7) and were mostly below 100%, except for ²³⁴Th at station 1. In contrast to 595 ²³⁴Th. ²¹⁰Po fluxes and ²¹⁰Po-derived POC fluxes at 25 m showed a positive correlation with the 596 integrated deficits of nitrate found in the upper water column [Fernández-Méndez et al., 2015] (p 597 <0.05; $\rho = 0.83$; n = 6), which are used to estimate the annual new NPP rates [e.g. Codispoti et 598 al., 2013]. This confirms that the ²¹⁰Po proxy covered the productive season better than ²³⁴Th, 599 and suggests that consumption of nitrate resulted in the increase in export production. Thus, the 600 ²¹⁰Po-derived POC fluxes and annual NPP estimates can be useful to assess the seasonal strength 601 of the biological pump, allowing to overcome the temporal decoupling between production and 602 export, which is especially long in Arctic waters conforming to a global biogeochemical model 603 [*Henson et al.*, 2015]. 604

The export efficiency according to the annual NPP and the ²¹⁰Po-derived POC fluxes is 605 illustrated in Figure 5. Only two locations showed export efficiencies <10% (stations 3 and 6, 606 Table 5), which are those typically found in the world ocean [Buesseler, 1998]. Export 607 efficiencies >30% (average: $50 \pm 20\%$, n = 5) were found at the other stations, which is in good 608 agreement with those reported by Gustafsson and Andersson [2012] in the Eurasian Basin 609 (average: $34 \pm 8\%$, n = 3) and Chen et al. [2003] in the Canada Basin (26%, n = 1) applying 610 ²³⁴Th-derived POC fluxes and in situ NPP rates. Our estimates are also similar to the ²³⁴Th-611 derived export efficiencies of ~30-40% reported for Chukchi shelf, slope and basin stations in 612

summer [*Moran et al.*, 2005; *Lepore et al.*, 2007]. Although only a limited set of observations of export efficiency is available in the central Arctic, overall they point to high export efficiencies as also indicated by *Henson et al.* [2015]. The assessment of the export efficiency in the central Arctic deserves more attention to better understand its role as an export regime in a climate change framework. Observations of strong aggregation and rapid algal falls in the central Arctic [*Boetius et al.*, 2013; *Katlein et al.*, 2014] suggest a export system that works differently than in most of the world ocean.

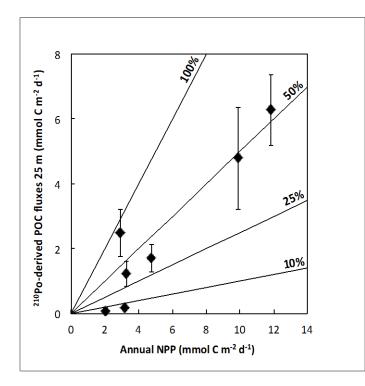


Figure 5: ²¹⁰Po-derived POC export fluxes at 25 m vs. annual new primary production reported in *Fernández- Méndez et al.* [2015]. Solid lines indicate the export efficiency.

623 **5 Conclusions**

620

We have used concurrently the ²³⁴Th/²³⁸U and ²¹⁰Po/²¹⁰Pb proxies to estimate POC fluxes in the central Arctic during the record sea-ice minimum in 2012. The main findings of the present work are:

627 1) ²³⁴Th reveals that POC fluxes at the bottom of the euphotic zone were very low $(2 \pm 2 \mod C \mod^{-2} d^{-1})$ in August/September, which is in good agreement with results obtained 629 using sediment traps $(3 \pm 3 \mod C \mod^{-2} d^{-1})$ deployed at the same locations [*Lalande et al.*, 2014]. The positive relationships found between prasinophytes_1 and ²³⁴Th and ²³⁴Th-derived POC fluxes suggest that picoplankton contributed significantly to
 downward fluxes in late summer.

- 633 2) In contrast to ²³⁴Th, the upper water column was depleted in ²¹⁰Po over the entire study
 634 area, indicating that particle export fluxes were higher before July/August than in the
 635 weeks prior to and during the survey.
- 3) The positive relationships obtained between sea-ice concentration and ²¹⁰Po and ²¹⁰Poderived POC fluxes show that particle sinking was greater under heavy sea-ice conditions
 than under partially ice covered areas. Further, the strongest ²¹⁰Po deficits in the water
 column coincided with the highest seafloor coverage of algae reported by *Boetius et al.*[2013], suggesting that ²¹⁰Po tracked, to some extent, the massive algal export that
 occurred earlier in the season.
- 4) Although the POC fluxes were low, a large fraction of primary production (>30%) was
 exported at the base of the euphotic zone in most of the study area, according to ²¹⁰Poderived POC fluxes and annual NPP estimates. Seasonal estimates of primary production
 and export would be very helpful in characterizing the role of the Arctic biological pump
 in the context of climate change.
- We encourage future studies applying radionuclide proxies to consider NSS conditions and follow the trend of C/Th and C/Po ratios with time to better constrain the POC fluxes in the Arctic. Further, the simultaneous use of sediment traps would allow the determination of the particle flux composition, which has been pointed out as a crucial factor shaping the biological pump efficiency [e.g. *Mackinson et al.*, 2015; *Puigcorbé et al.*, 2015; *Roca-Martí et al.*, 2016].

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- http://doi.pangaea.de/10.1594/PANGAEA.858790.

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