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New insights on the ^7Be cycle in the ocean

Grenier M.^{1,*}, van Beek P.¹, Lerner P.^{2,3}, Sanial V.⁴, Souhaut M.¹, Lagarde M.¹,
Marchal O.⁵, Reyss J.L.⁶

¹ LEGOS, University of Toulouse, CNRS, CNES, IRD, UPS, Toulouse, 31400, France

² NASA-GISS, New York City, NY, 10025, USA

³ Department of Applied Physics and Applied Mathematics, Columbia University, New York City, NY, 10027, USA

⁴ Université de Toulon, Aix Marseille Univ., CNRS, IRD, MIO, Toulon, 83041, France

⁵ Woods Hole Oceanographic Institution, Woods Hole, MA, 02543, USA

⁶ Laboratoire des Sciences du Climat et de l'Environnement, Gif-sur-Yvette, 91198, France

* Corresponding author: Mélanie Grenier (melanie.grenier@legos.obs-mip.fr)

Abstract

The cosmogenic radionuclide ^7Be has been applied as a tracer of dynamical processes in the upper ocean and of atmospheric deposition of trace elements at the sea surface. These applications usually assume that ^7Be is entirely in the dissolved form, and that scavenging and downward export of ^7Be by settling particles can be neglected. In this work, we explore these assumptions and more generally assess the significance of the ^7Be activity in the particulate fraction, through the generation of vertical profiles of particulate ^7Be in the open ocean. From detailed measurements obtained from low-background gamma spectrometers placed in underground facilities, we report vertical profiles of ^7Be activity in suspended particles ($^7\text{Be}_p$) collected in various oceanic regions: the Mediterranean Sea (DYFAMED station), the Indian Sector of the Southern Ocean (station A3-2 from the KEOPS2 cruise), the Sargasso Sea (OFP station), and the subpolar North Atlantic Ocean (GEOVIDE cruise).

We find that, in each oceanic region, $^7\text{Be}_p$ activities are generally higher in the mixed layer than in the thermocline. They vary in the mixed layer from 3.0 dpm/m³ at DYFAMED to 33.1 dpm/m³ at GEOVIDE cross-over station 51/60, i.e., within a range consistent with previous $^7\text{Be}_p$ measurements for the open ocean. For the GEOVIDE cruise, the $^7\text{Be}_p$ activities measured on different filter types at different depths are corrected for filter offsets derived from multiple $^7\text{Be}_p$ measurements at a near-coastal station in the western Mediterranean Sea. We then combine measurements of total ^7Be activity (Shelley et al., 2017) with our measurements of $^7\text{Be}_p$ activity to estimate the solid/solution partitioning of ^7Be . On average, the particulate fraction would represent 6% of total ^7Be activity at 5-m water depth ($n = 6$), 22% at 20 m ($n = 2$) and 9% at 70

34 m ($n = 3$). At GEOVIDE stations, ${}^7\text{Be}_p$ inventories range from 5% to 19% of the total ${}^7\text{Be}$
35 inventories. In the Labrador Sea, the measured ${}^7\text{Be}_p$ inventories are lower than the dry ${}^7\text{Be}$
36 deposition fluxes estimated from aerosol samples collected during GEOVIDE, suggesting that
37 a significant portion of ${}^7\text{Be}_p$ may be removed by sinking particles. The distribution coefficient
38 K_d for ${}^7\text{Be}$ increases with depth, with $\log_{10}K_d$ averaging 5.2 ± 0.1 at 5 m to 6.1 ± 0.1 between
39 70 and 150 m, suggesting that scavenging of dissolved ${}^7\text{Be}$ by particles is more pronounced in
40 the thermocline than in the mixed layer when differences in particle concentrations are taken
41 into account. Overall, our study suggests that, at least in some oceanic regions, the removal of
42 ${}^7\text{Be}$ by marine particles may be significant and that it may need to be considered in applications
43 of ${}^7\text{Be}$ as a tracer of oceanic processes and atmospheric deposition.

44 **Keywords:** Beryllium-7; suspended particles; vertical profiles; open ocean; solid/solution
45 partitioning; GEOTRACES

46 **1. Introduction**

47 Beryllium-7 (${}^7\text{Be}$) is produced in the atmosphere by cosmic ray spallation mostly of oxygen
48 and nitrogen nuclei (Lal and Peters, 1967). ${}^7\text{Be}$ then enters the marine and terrestrial
49 environments via wet and dry depositions (Feely et al., 1989). The relatively short half-life of
50 ${}^7\text{Be}$ (53.3 days; Browne et al., 1978) makes it a useful tracer of processes in the coastal ocean
51 and the open upper ocean on seasonal and regional time-scales.

52 In estuaries and coastal environments, ${}^7\text{Be}$ can be quickly scavenged by particles (Aaboe
53 et al., 1981; Dibb and Rice, 1989; Olsen et al., 1986). Several studies have reported a significant
54 affinity of ${}^7\text{Be}$ for particles in these environments, where the distribution coefficients K_d for
55 ${}^7\text{Be}$ can be of the order of $10^5 \text{ cm}^3/\text{g}$, with K_d being the ratio of particulate ${}^7\text{Be}$ activity to
56 dissolved ${}^7\text{Be}$ activity, normalized to particle concentration (Baskaran and Santschi, 1993; Dibb
57 and Rice, 1989). Consistent with this result, ${}^7\text{Be}$ has been applied in coastal and estuarine
58 systems to study short-term variations of sediment accumulation rate and sediment transport
59 (e.g., Dibb and Rice, 1989; Wu et al., 2018).

60 In the open ocean, ${}^7\text{Be}$ is generally well mixed in the surface mixed layer, and decreases
61 about exponentially with depth in the upper thermocline (Aaboe et al., 1981; Kadko et al., 2015;
62 Kadko and Olson, 1996; Silker, 1972a; Young and Silker, 1980). This pattern has led to the use
63 of ${}^7\text{Be}$ for quantifying mixing in the thermocline and surface water subduction (e.g., Kadko and
64 Olson, 1996), as well as upwelling rates (Haskell et al., 2015; Kadko, 2017; Kadko and Johns,
65 2011). Recently, Kadko et al. (2015) used the inventory of ${}^7\text{Be}$ measured in the upper water

66 column to estimate the atmospheric deposition fluxes of other trace elements in remote oceanic
67 regions (Kadko et al., 2019; Shelley et al., 2017).

68 Many of these applications of ^7Be as a tracer of oceanic and atmospheric processes rely on
69 the assumption that ^7Be behaves conservatively in seawater, i.e., that radioactive decay is the
70 only sink for ^7Be . Previous studies on ^7Be cycling in the open ocean usually considered that
71 ^7Be occurs predominantly in the dissolved form and neglected particulate scavenging
72 (adsorption of dissolved ^7Be onto particles) and downward export by settling particles. These
73 assumptions are based on evidence that ^7Be is relatively soluble in low particle environments
74 such as in the oligotrophic regions of the open ocean (Aaboe et al., 1981; Kadko and Prospero,
75 2011). However, particulate ^7Be ($^7\text{Be}_p$) measurements are scarce in the open ocean. ^7Be activity
76 in suspended particles has been rarely determined, and the few particulate ^7Be measurements
77 that are available are found mostly in the mixed layer (Andrews et al., 2008; Kadko and Johns,
78 2011; Kremenchutskii et al., 2021; Silker, 1972b, 1972a; Silker et al., 1968). For example,
79 Andrews et al. (2008) reported ^7Be activities below the detection limit in a particulate sample
80 collected by pumping 1000 L of surface water in the Sargasso Sea. From a study conducted in
81 the upper 100 m of the Sargasso Sea off Bermuda, Silker (1972a) concluded that < 10% of the
82 ^7Be was present in the particulate form. However, to our knowledge, no $^7\text{Be}_p$ measurements
83 exist below a water depth of 100 m.

84 Regarding the atmospheric ^7Be source, it is often considered that ^7Be enters the surface
85 ocean primarily via precipitation (wet deposition), although our understanding of ^7Be
86 deposition in terrestrial and marine environments remains limited. For instance, Gaffney et al.
87 (1994) analysed rain samples from Illinois and New Mexico (USA), and concluded that ^7Be
88 was primarily in the dissolved form, the particulate ^7Be representing only 1.2 to 12.1% of total
89 (dissolved + particulate) ^7Be . On the other hand, Papastefanou and Ioannidou (1996) analyzed
90 aerosols sampled near Thessaloniki (Greece), and reported that particulate ^7Be (nominal filter
91 pore size = 1.1 μm) could represent 20-50% of the total ^7Be activity in aerosols. The wet and
92 dry depositions of ^7Be to the surface ocean are even less well constrained.

93 Similarly, the significance and fate of particulate ^7Be deposited at the sea surface have been
94 rarely investigated. Dissolution of aerosols in the ocean mixed layer may release into solution
95 ^7Be present at the surface and/or within the aerosol particles. Alternatively, adsorption of
96 dissolved ^7Be onto settling particles may remove a significant fraction of ^7Be from solution.
97 Aggregation of small particles to form larger particles may further increase the vertical flux of
98 particulate ^7Be to deep waters by increasing particle settling speeds (e.g., Burd and Jackson,

99 2009). The removal of ^7Be from the surface ocean by attachment to settling particles may
100 impact the vertical distribution of dissolved ^7Be and may thus introduce biases in ^7Be -based
101 estimates of dynamical parameters such as vertical eddy diffusivity (e.g., Haskell, Kadko, et
102 al., 2015; Kadko and Olson, 1996) and upwelling rate (e.g., Kadko and Johns, 2011).

103 Similarly, the method developed by Kadko et al. (2015) to estimate trace element (TE)
104 atmospheric fluxes could suffer from a bias, should particle scavenging and export, in addition
105 to radioactive decay, be a significant sink of ^7Be . In order to assess whether scavenging and
106 export of ^7Be by settling particles can be neglected in the open ocean, the significance of
107 particulate ^7Be and the exchange of ^7Be between the dissolved and particulate phases need to
108 be better constrained.

109 Here we report measurements of ^7Be activity determined in suspended particles collected
110 between 2002 and 2014 in the upper water column in different oceanic regions. Through paired
111 estimates of ^7Be activity in the particulate and total phases, our goals are (i) to provide
112 constraints on the solid-solution partitioning of ^7Be in different oceanic environments, (ii) to
113 develop a better understanding of the processes affecting the cycling of this cosmogenic isotope
114 in the water column, and (iii) to assess the applicability of ^7Be as a tracer of transport processes
115 in the ocean and of the deposition of TEs at the sea surface.

116

117 **2. Methods**

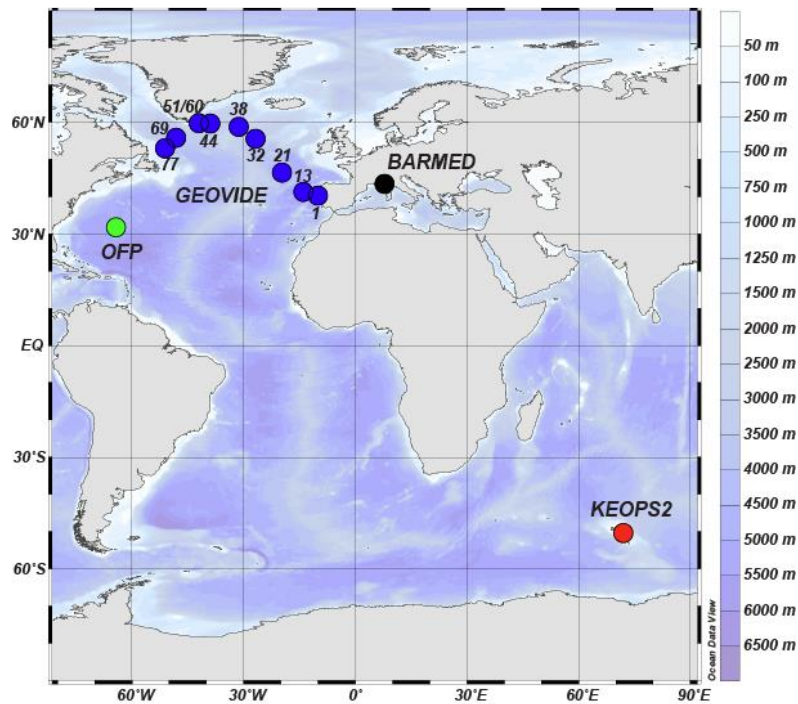
118 **2.1 Sampling**

119 ^7Be activities were determined in suspended particle samples collected at different depths
120 in four different oceanic regions (Tables 1 and 2; Figure 1): 1) at the OFP (Oceanic Flux
121 Program) station off Bermuda in the Sargasso Sea in May 2002, aboard the R/V *Weatherbird*
122 *II* (PIs: Maureen Conte and Roger François; van Beek et al., 2007); 2) at the DYFAMED
123 (Dynamics of Atmospheric Fluxes in the MEDiterranean sea) station in the western
124 Mediterranean Sea in March and May 2003 during the BARMED project, aboard the R/V
125 *Téthys II* (PI: Catherine Jeandel; van Beek et al., 2009); 3) at station A3-2 on the Kerguelen
126 Plateau in the Southern Ocean in November 2011 during the KEOPS2 project, aboard the R/V
127 *Marion Dufresne* (PI: Stéphane Blain); and 4) at nine stations of the GEOVIDE section
128 completed in the North Atlantic in May-June 2014, aboard the R/V *Pourquoi Pas?*
129 (GEOTRACES cruise GA01; PIs: Géraldine Sarthou and Pascale Lherminier). The suspended

130 particles were collected using different types of filters with 0.8-1 μm nominal porosity and 142-
131 mm diameter mounted on McLane large volume pumps. After collection, the filters were stored
132 at room temperature in petri dishes until their return to the laboratory. Specifically, 0.8- μm pore
133 size Versapor filters were used for OFP and BARMED cruises, and 0.8- μm pore size Supor
134 filters were used for KEOPS2. For the GEOVIDE cruise, we used either 1 μm -pore size QMA
135 filters or 0.8- μm pore size Supor filters. During GEOVIDE, surface samples (ca. 5 m below the
136 sea surface) were also collected using the ship seawater intake and were filtered through pocket
137 filters (polypropylene filters Pentek BP-410-1; hereafter referred as socks) with a pore size of
138 1 μm . This method allows the filtration of very large volumes of seawater, although it is not a
139 conventional technique for collecting particulate samples for TE analyses. Filters such as
140 Versapor, QMA, and Supor are more commonly used to collect suspended particles in marine
141 environments.

142 The use of different filter types may generate systematic differences in the measured $^7\text{Be}_p$
143 activities, as was observed for $^{234}\text{Th}_p$ measured on different quartz filters (glass fiber filters, or
144 GF/F, and microquartz filters; Benitez-Nelson et al., 2001; Maiti et al., 2012). In particular,
145 $^7\text{Be}_p$ measurements at different depths are combined in this study to generate vertical profiles
146 of $^7\text{Be}_p$ at GEOVIDE stations, although at these stations ^7Be activities at different depths have
147 been measured on different filter types. In Section 4.2, we assess the impact of the different
148 filter types used for GEOVIDE on the determination of $^7\text{Be}_p$ activity, and we derive filter-
149 specific corrections for the GEOVIDE $^7\text{Be}_p$ activities.

150 During GEOVIDE, total ^7Be activities (i.e., sum of dissolved and particulate ^7Be activities)
151 were also determined on unfiltered samples gathered at different depths at the same stations as
152 those considered in this study (Shelley et al., 2017). However, samples dedicated to particulate
153 and total ^7Be analyses came from different casts, and some of these casts exhibited significant
154 differences in vertical density profiles (sections 4 and 5). As a result, some caution should be
155 exercised when combining the measurements of total and particulate ^7Be at GEOVIDE stations.



156

157 **Figure 1:** Stations where samples for ^7Be measurements were collected and analysed in this study. These
 158 samples were collected in the framework of different programs: OFP in the Sargasso Sea (green dot),
 159 BARMED in the Mediterranean Sea (black dot), KEOPS2 in the Southern Ocean (red dot), and
 160 GEOVIDE/GA01 in the North Atlantic (blue dots).

161

162 **2.2 ^7Be analyses**

163 Upon return to the laboratory, the particulate (filter) samples were folded and sealed in
 164 counting tubes. The analyses of $^7\text{Be}_p$ were conducted within two weeks to four months of the
 165 arrival of the samples in the laboratory, using low background gamma-ray spectrometers at two
 166 different facilities: the laboratory LAFARA (Laboratoire de mesure des Faibles Radioactivités;
 167 Université Toulouse III Paul Sabatier; van Beek et al., 2013) in the French Pyrénées, and the
 168 laboratory of Modane in the French Alps (Reyss et al., 1995). Both laboratories host high-purity
 169 germanium (HPGe) gamma-ray spectrometers manufactured with selected materials. Both are
 170 underground, so that the detectors are protected from the influence of cosmic rays and provide
 171 very low background levels (Reyss et al., 1995; van Beek et al., 2013). The filter samples were
 172 placed within the germanium crystals (well-type detectors), which allowed us to further
 173 increase the sensitivity of the analyses. By combining the low background levels achieved at
 174 LAFARA and Modane with the use of sensitive well-type detectors, very low levels of
 175 radioactivity could be quantified. The ^7Be activities were determined using the 477.6 keV
 176 gamma line. Due to the lack of any reference material for ^7Be , we determined the efficiency at
 177 477.6 keV from the efficiency curve obtained from different reference materials (RGU-1,

178 RGTH-1 and #375) provided by the International Atomic Energy Agency (Martínez-Ruiz et
179 al., 2007).

180 The uncertainties of the $^7\text{Be}_p$ activities measured at LAFARA and Modane and reported in
181 this study are 1 sigma uncertainties (counting statistics). The relative uncertainties range from
182 3% to 33% for samples collected in the upper 300 m (14% on average) and can be higher for
183 samples collected below 300 m. Note that the precision on the ^7Be activity primarily depends
184 on the number of counts determined by the gamma spectrometers, which itself depends on
185 different parameters, including i) counting time, ii) sample volume (larger volumes may lead
186 to higher count rates), iii) the time elapsed between sampling and analysis (a short time will
187 limit loss of ^7Be by radioactive decay), and iv) the sensitivity of the gamma spectrometers (the
188 large, well-type detectors that are placed underground and that are used for this study tend to
189 reduce the background). The detection limits for ^7Be are 0.36-0.48 dpm for the well-type
190 detectors at LAFARA. The detection limits were not determined for the gamma spectrometers
191 at Modane, but they can reasonably be assumed similar or even slightly lower.

192 LAFARA regularly participates to interlaboratory comparison exercises organized by the
193 French institution IRSN (Institut de Radioprotection et Sûreté Nucléaire) to evaluate the ability
194 of this laboratory to quantify natural and artificial radionuclides in various substrates. The
195 accuracy of radionuclide measurements at LAFARA is thus regularly tested. Because LAFARA
196 successfully participated to these exercises and follows the recommendations of the
197 international standard ISO/CEI-17025, this laboratory has been certified by the French Nuclear
198 Safety Authority (Autorité de Sûreté Nucléaire, ASN). Although ^7Be is not part of the
199 interlaboratory comparison organized by IRSN, overall, the successful analysis of gamma
200 emitters with a wide range of energies (30-2500 keV) validates the efficiency curves of the
201 gamma spectrometers (calibration of the detectors), which are key for the quantification of
202 accurate radionuclide activities.

203 LAFARA also participated to an interlaboratory comparison exercise organized by the
204 GEOTRACES Standards and Intercalibration Committee to evaluate the ability to quantify ^7Be
205 activity in water samples. This exercise involved six laboratories worldwide and aimed at
206 testing accuracy, precision, and reproducibility by preparing and analysing three replicates (W.
207 Geibert, unpublished data). Overall, the different labs involved reported ^7Be activities that are
208 in good agreement with each other. The mean ^7Be activity of the three replicates analysed by
209 LAFARA was 162.6 ± 4.8 dpm/kg (3% precision with the 1 sigma uncertainty). The mean of

210 162.6 dpm/kg was within the one standard deviation of the mean ^7Be activity measured by the
211 six labs (149.4 ± 14.4 dpm/kg). Note that, at the different stations reported here, we did not
212 analyse replicates because the large seawater volumes needed to quantify ^7Be usually prevent
213 from collecting replicates.

214 **2.3 Suspended particle matter (SPM) concentration**

215 Concentrations of suspended particle matter ([SPM]) were estimated following the method
216 described in Lam et al. (2015) from the concentrations of particulate lithogenic matter, organic
217 matter, opal, calcium carbonate, iron (Fe) hydroxides, and manganese (Mn) oxides. Particulate
218 organic matter concentration was determined from concentrations of particulate organic carbon
219 and biogenic silica (Sarhou et al., 2018). Lithogenic matter concentration was determined from
220 concentrations of particulate aluminum, calcium carbonate from particulate calcium, Fe
221 hydroxides from particulate Fe, and Mn oxides from particulate Mn (Gourain et al., 2019).
222 These analyses were not performed on the filters dedicated to ^7Be analysis but on (i) filters
223 dedicated to lead-210 and polonium-210 analyses (^{210}Pb - ^{210}Po) mounted on *in situ* pumps (ISP;
224 Y. Tang et al., 2018) and on (ii) filter samples collected from Niskin and Go-Flo bottles, as
225 described in Lagarde et al. (submitted). We generally used [SPM] estimates determined from
226 bottle samples rather than those determined from ISP samples (Tang et al., 2018); the rosette
227 samples have greater vertical resolution than the ISP samples, especially in the upper 40 m of
228 the water column, which provides a better agreement with the depths of $^7\text{Be}_p$ samples. The
229 [SPM] estimated from Niskin/Go-Flo and ISP samples are compared in section 5.4.

230

231 **3. Oceanographic context**

232 We chose to measure particulate ^7Be on samples collected at OFP (Sargasso Sea) and
233 DYFAMED (Mediterranean Sea), since oceanographic conditions at these stations are
234 relatively well documented, particularly through time series programs. The OFP sediment trap
235 time series mooring (Conte et al., 2001) is located in the northern Sargasso Sea (4,200 m water
236 depth) in a transitional region between relatively eutrophic waters to the north and oligotrophic
237 subtropical waters to the south. The area is also the site of the Bermuda-Atlantic Times Series
238 (BATS, Steinberg et al., 2001) and the Bermuda Testbed Mooring (BTM, Dickey et al., 2001).
239 At the time of sample collection (May 2002), the mixed layer at OFP was ~30 m deep (van
240 Beek et al., 2007).

241 The DYFAMED sediment trap time-series mooring was initiated in 1988 and is located in
242 the north-western Mediterranean Sea (43°25' N; 7°52' E). It is situated in 2,350 m of water,
243 approximately 45 km south of Cape Ferrat, France. The DYFAMED station is generally
244 considered to be representative of open ocean conditions (Marty et al., 2002), although it may
245 be episodically impacted by continental inputs (Sternberg et al., 2008; van Beek et al., 2009).
246 It receives significant atmospheric input from the Saharan Desert (Sarhou and Jeandel, 2001).
247 Between the two visits of the DYFAMED station considered here (BARMED 2 in March 2003
248 and BARMED 4 in May 2003), the mixed layer shoaled from ~25 m in March to ~15 m in May
249 (van Beek et al., 2009).

250 Station A3-2 is located in the Indian sector of the Southern Ocean on the Kerguelen Plateau,
251 south-east of the Kerguelen Islands and south of the Polar Front (Blain et al., 2007; Sanial et
252 al., 2015). Following a first occupation during the KEOPS1 project in 2005 (PI: Stéphane
253 Blain), this station was visited again in year 2011, in the framework of the KEOPS2 project (PI:
254 Stéphane Blain). It is located in the HNLC (High Nutrient, Low Chlorophyll) waters of the
255 Southern Ocean in water depth of 520 m. The Kerguelen Plateau was shown to deliver
256 significant amounts of iron that promote an annual phytoplankton bloom in the region (Blain et
257 al., 2007; van Beek et al., 2008). The mixed layer at station A3-2 was ~150 m deep when the
258 samples were collected (Jouandet et al., 2014).

259 Finally, we report data from nine stations visited during the GEOVIDE cruise
260 (GEOTRACES GA01) in the subpolar North Atlantic. The stations occupied during GEOVIDE
261 present large differences in biogeochemical and hydrological properties (García-Ibáñez et al.,
262 2018; Lemaitre et al., 2018b; Sarhou et al., 2018; Zunino et al., 2017). Stations 1 and 13 are
263 located in the Iberian basin (on and west of the Iberian margin, respectively) and were occupied
264 during the decline of a phytoplankton bloom. Several stations visited during GA01 are located
265 in the vicinity of the North Atlantic Current, which flows in the West European basin (station
266 21) and in the Iceland basin (station 32 in the southern part of the basin, station 38 in the
267 northern part), where nutrient availability and/or light limit primary production. West of these
268 stations, along GA01, stations 44 and 51/60 in the Irminger Sea and stations 69 and 77 in the
269 Labrador Sea were also sampled for ⁷Be analysis. For this study, stations 51 and 60 are grouped
270 as station 51/60 because these two stations are a cross-over station visited two days apart.
271 Samples dedicated to particulate ⁷Be analysis were collected at station 51 (on June 16) while
272 those dedicated to total ⁷Be analysis were collected two days later at station 60 (Shelley et al.,
273 2017). The mixed layer depth (MLD), as estimated from density and temperature profiles

274 (Tonnard et al., 2018), ranged from ~12 m at stations 60 and 77 to ~48 m at station 51. The
275 mixed layer at station 51/60 thus shoaled considerably between the two occupations, which was
276 perhaps due to variability of the East Greenland Current and/or the Irminger Current (Daniault
277 et al., 2011). Circulation patterns and water masses along GA01 are detailed in García-Ibáñez
278 et al. (2018) and Zunino et al. (2017).

279

280 **4. Results**

281 **4.1. $^7\text{Be}_p$ in the Sargasso Sea, Mediterranean Sea, and Kerguelen Plateau**

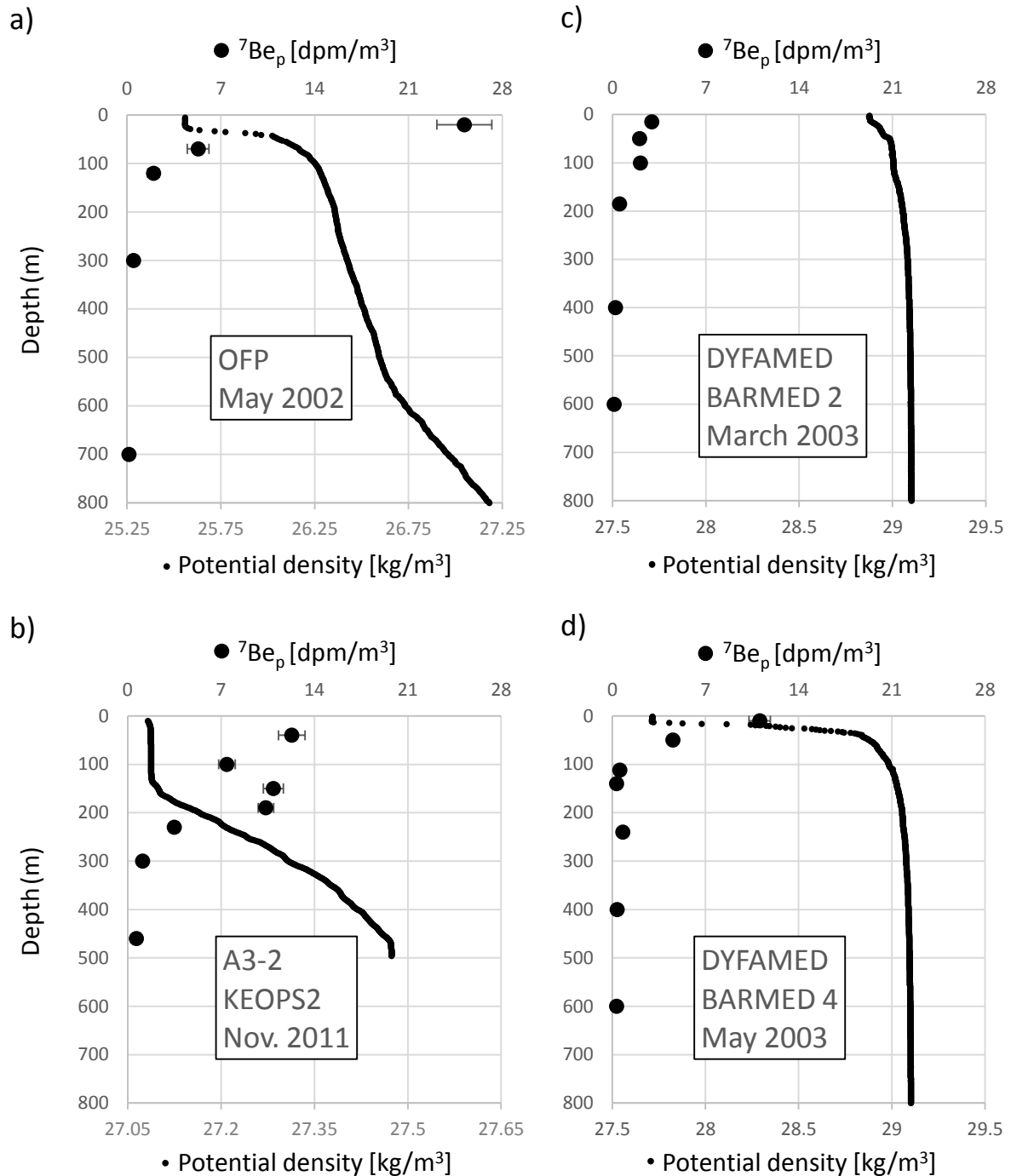
282 The $^7\text{Be}_p$ activities determined on samples collected in the Sargasso Sea, Mediterranean
283 Sea, and Kerguelen Plateau are given in Table 1 and shown in Figure 2. Overall, the $^7\text{Be}_p$
284 activities determined on mixed layer samples vary by one order of magnitude across these
285 oceanic regions, from 3.0 dpm/m³ in the Mediterranean Sea at 15 m (DYFAMED, BARMED
286 2), to 25.2 dpm/m³ in the Sargasso Sea at 20 m (OFF). In the Southern Ocean (station A3-2),
287 $^7\text{Be}_p$ activity is found to be ~12 dpm/m³ for the shallowest sample (40 m) and is comparable at
288 the two other depths sampled in the mixed layer (average $^7\text{Be}_p$ activity of ~10 dpm/m³).
289 Significant temporal variations in surface water $^7\text{Be}_p$ activity are observed at station
290 DYFAMED, with a much higher value in May 2003 (BARMED 4; 11.07 dpm/m³) than in
291 March 2003 (BARMED 2; 2.95 dpm/m³; Figure 2). These variations may reflect (i) variations
292 in MLD, which was shallower in May (~15 m) compared to March (~25 m), resulting in a
293 greater dilution of ^7Be activities in March, and (ii) the greater stratification of the upper water
294 column in May (van Beek et al., 2009), resulting in abated turbulent mixing with thermocline
295 waters in May. Other factors may also be involved. Similar seasonal variations in total ^7Be were
296 observed in other oceanic regions, such as in the Sargasso Sea, and were mainly ascribed to
297 MLD variations (Kadko, 2009): the deeper the mixed layer, the lower the ^7Be activity as a result
298 of dilution.

299 Noticeably, at the four stations in the Sargasso Sea, Mediterranean Sea, and Kerguelen
300 Plateau, $^7\text{Be}_p$ activities are lower in the upper thermocline than in the mixed layer (Figure 2).
301 This pattern is consistent with the atmospheric origin and the relatively short half-life of ^7Be .
302 The vertical profiles generally show a monotonic decrease of $^7\text{Be}_p$ activity with depth below
303 the mixed layer (Figure 2).

304

Station	Depth (m)	$^7\text{Be}_p$ (dpm/m ³)		
OFP Time-series (Sargasso Sea, May 2002)				
OFP	20	25.16	±	2.05
OFP	70	5.30	±	0.80
OFP	120	1.96	±	0.41
OFP	300	0.47	±	0.13
OFP	700	0.12	±	0.08
OFP	1420	0.04	±	0.09
OFP	4250	0.05	±	0.09
BARMED 2 (DYFAMED, Mediterranean Sea, March 2003)				
DYFAMED	15	2.95	±	0.09
DYFAMED	50	2.03	±	0.05
DYFAMED	100	2.09	±	0.06
DYFAMED	185	0.54	±	0.02
DYFAMED	400	0.23	±	0.05
DYFAMED	600	0.13	±	0.05
DYFAMED	1000	0.06	±	0.06
BARMED 4 (DYFAMED, Mediterranean Sea, May 2003)				
DYFAMED	10	11.07	±	0.80
DYFAMED	50	4.55	±	0.38
DYFAMED	112	0.56	±	0.08
DYFAMED	140	0.32	±	0.08
DYFAMED	240	0.78	±	0.15
DYFAMED	400	0.37	±	0.06
DYFAMED	600	0.32	±	0.07
DYFAMED	2200	0.15	±	0.04
KEOPS2 (Indian sector of the Southern Ocean, November 2011)				
A3-2	40	12.31	±	1.00
A3-2	100	7.44	±	0.62
A3-2	150	10.92	±	0.76
A3-2	190	10.36	±	0.59
A3-2	230	3.49	±	0.35
A3-2	300	1.12	±	0.21
A3-2	400	BDL		
A3-2	460	0.66	±	0.19

305 **Table 1:** Particulate ^7Be activities (in dpm/m³) determined at stations OFP (Sargasso Sea), DYFAMED
306 (Mediterranean Sea), and A3-2 (Kerguelen Plateau). Versapor filters (0.8 μm) were used at stations OFP
307 and DYFAMED, and Supor filters (0.8 μm) at station A3-2. The reported errors are standard deviations
308 from the counting statistics. BDL: Below Detection Limit.



309

310 **Figure 2:** Profiles of particulate $^7\text{Be}_p$ activity ($^7\text{Be}_p$, in dpm/m^3 ; black dots) determined in suspended
 311 particles collected using *in situ* pumps at stations OFP (Sargasso Sea), DYFAMED (Mediterranean Sea),
 312 and A3-2 (Kerguelen Plateau). Error bars represent ± 1 standard deviation from the counting statistics.
 313 Versapor filters of $0.8 \mu\text{m}$ pore size were used at all depths for OFP and BARMED cruises, whereas
 314 Supor filters of $0.8 \mu\text{m}$ pore size were used at all depths for KEOPS2 (Table 1). Potential density profiles
 315 from CTD cast data are also shown (small black dots).

316

317 4.2. Impact of the use of different filter types on $^7\text{Be}_p$ measurement

318 In contrast to stations OFP, DYFAMED, and A3-2, $^7\text{Be}_p$ measurements at different depths
319 were obtained for samples collected from different filter types at GEOVIDE stations. These
320 filter types include 1- μm pore size sock filters for surface waters (5 m) and 1- μm pore size
321 QMA and 0.8- μm pore size Supor filters for deeper waters.

322 The use of different filter types may generate systematic differences in the determination
323 of the $^7\text{Be}_p$ activities and thus produce artifacts in the $^7\text{Be}_p$ vertical profiles. To our knowledge,
324 the impact of different filter types on the measurement of particulate Be concentration in
325 seawater has never been examined. Most relevant to this issue is perhaps the study of Maiti et
326 al. (2012), who compared particulate ^{234}Th activities of samples collected on QMA and Supor
327 filter punches. The inter-filter variability was found to be 8.1% for QMA filter punches and
328 16.8% for Supor ones. For both filter types (QMA and Supor), inter-filter variability (i.e., the
329 variability associated with subsamples taken from the same filter) in $^{234}\text{Th}_p$ was partly due to
330 (i) an uneven distribution of particles on the filter and (ii) a potential bias associated with
331 subsampling the particulate material on the filters (Maiti et al., 2012). In the present study, we
332 did not analyze punches but the entire filter, so inter-filter variability associated with factor (i)
333 above should not be a source of variability in our $^7\text{Be}_p$ dataset.

334 It was also shown that QMA filters may adsorb dissolved ^{234}Th , leading to ^{234}Th activities
335 which are 10 to 20% higher on QMA filters (filtered volume: 450-600 L) than on Supor filters
336 (filtered volume: 200-400 L; Maiti et al., 2012). Other studies, however, showed that the
337 sorption effect tends to be larger for smaller sample volumes (Benitez-Nelson et al., 2001;
338 Buesseler et al., 1998); for filtered volumes smaller than 150 L, ^{234}Th activities were found to
339 be at least twice as high on QMA filters than on Nuclepore filters (Benitez-Nelson et al., 2001).
340 In the present study, the volumes of seawater that was pumped through the QMA filters varied
341 from 200 L to 850 L, while the volumes of seawater that passed through the 1- μm socks via the
342 ship seawater intake varied from 1,200 L to 10,800 L (see Table 2). To our knowledge, this
343 study is the first to report ^7Be activities of particulate material collected by 1- μm pore size
344 socks, so that previous values are not available for comparison.

345 In order to estimate the impact of different filter types on the determination of $^7\text{Be}_p$,
346 preliminary tests have been conducted on particulate samples collected near the Mediterranean
347 coast. Surface seawater samples were collected in June 2022 aboard the R/V *Nereis II* at station
348 POLA (42° 28' 300" N, 03° 15' 500" E; water depth: 95 m), located five miles offshore of
349 Banyuls-sur-Mer. The samples were filtered in duplicate through the different types of filters

350 used during GEOVIDE: 0.8- μm pore size Supor filters, 1- μm pore size QMA filters, and 1- μm
351 pore size socks.

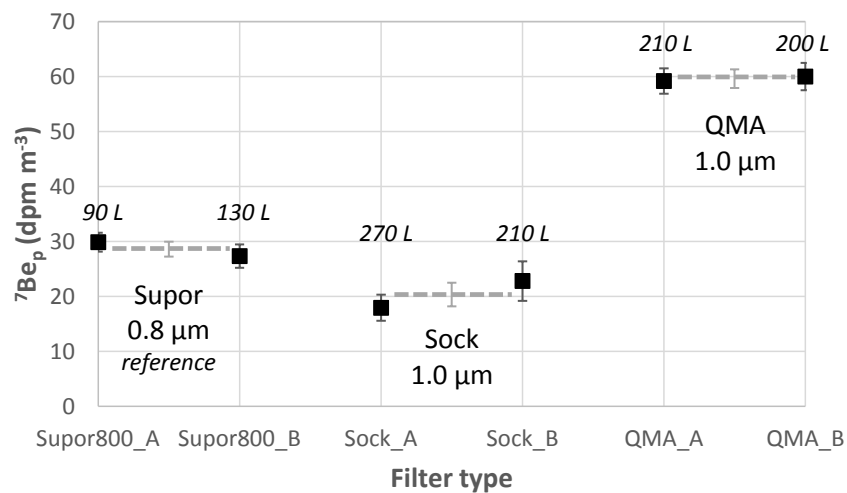
352 We found that (i) the mean activity of $^7\text{Be}_p$ on 1- μm QMA filters ($59.6 \pm 1.7 \text{ dpm/m}^3$) was
353 2.1 times higher than the mean activity on 0.8- μm Supor filters ($28.6 \pm 1.4 \text{ dpm/m}^3$), and (ii)
354 the mean activity of $^7\text{Be}_p$ on 1- μm sock filters ($20.4 \pm 2.2 \text{ dpm/m}^3$) was 1.4 times lower than
355 the mean activity on 0.8- μm Supor filters (Figure 3). The activity differences far exceeded the
356 errors associated with the counting statistics. Supor filters are considered here as a reference
357 since they are the filters of choice for GEOTRACES (Maiti et al., 2012). In our tests, the
358 seawater volume that passed through each filter was $\sim 200 \text{ L}$, except for 0.8- μm Supor filters,
359 which got clogged for smaller volumes (Figure 3). Although our tests are preliminary, they do
360 suggest that dissolved ^7Be absorbs onto QMA filters, thus increasing the measured $^7\text{Be}_p$ activity,
361 similarly to ^{234}Th (Maiti et al., 2012).

362 As noted above, we also found that a significant fraction of the particulate ^7Be is not
363 recovered during the filtration through sock filters compared to Supor filters. This lower
364 recovery for the sock filters is consistent with our results at GEOVIDE station 77, where the
365 $^7\text{Be}_p$ activity determined at 5 m on 1- μm sock (6.54 dpm/m^3) is lower than the $^7\text{Be}_p$ activity
366 determined at 10 m on 0.8- μm Supor filter (12.76 dpm/m^3 ; Table 2), although natural variability
367 could also contribute to the difference.

368 In summary, our preliminary tests suggest that $^7\text{Be}_p$ activities on QMA and sock filters may
369 show systematic difference with the $^7\text{Be}_p$ activities on the (reference) Supor filters. Here the
370 results from these tests are used to correct the $^7\text{Be}_p$ activities on QMA and sock filters at
371 GEOVIDE stations. The $^7\text{Be}_p$ activities on QMA filters are divided by 2.1, whereas the $^7\text{Be}_p$
372 activities on sock filters are multiplied by 1.4 (Table 2). The errors in the corrected $^7\text{Be}_p$
373 activities on QMA and sock filters are calculated by propagating the errors in the uncorrected
374 activities and the errors in the correction factors, neglecting error covariances (Bevington and
375 Robinson, 1992).

376 It is clear that the filter corrections applied to the $^7\text{Be}_p$ activities measured on QMA and
377 sock filters are not without limitations. The correction factor applied to $^7\text{Be}_p$ activities on QMA
378 filters for $> 400 \text{ L}$ of filtered seawater (see Table 2) may be overestimated if the sorption effect
379 is smaller for larger sample volumes, as was observed for ^{234}Th (Maiti et al., 2012). Other
380 effects, such as the concentration and the chemical composition of the particulate material may
381 also contribute to the differences in $^7\text{Be}_p$ activity measured on different filter types. It is

382 therefore unclear whether filter corrections determined from one oceanographic environment
 383 (e.g., station POLA) could be applied to other stations (e.g., GEOVIDE stations). Nevertheless,
 384 we feel that the correction factors reported above for QMA and sock filters are currently the
 385 best approach to estimate the vertical distribution of ${}^7\text{Be}_p$ activity at GEOVIDE stations, where
 386 different filters were used at different depths.



387
 388 **Figure 3:** Preliminary tests of the impact of different filter types on the determination of ${}^7\text{Be}_p$ activity
 389 in seawater samples. Surface samples from station POLA were filtered through the different filters used
 390 at GEOVIDE stations (Supor 0.8 μm , Sock 1.0 μm , and QMA 1.0 μm). Duplicates were done for each
 391 filter (A and B; see filtered volume above each symbol) and gave reproducible ${}^7\text{Be}_p$ for each filter type
 392 (within ± 1 standard deviation of each individual ${}^7\text{Be}_p$ measurement, derived from counting statistics).
 393 The grey dashed lines and associated error bars show, respectively, the mean ${}^7\text{Be}_p$ for each filter type
 394 and its uncertainty (error propagation).

395

Station	Depth (m)	Filter	Filtered volume (L)	Uncorrected ${}^7\text{Be}_p$ (dpm/m ³)	Corrected ${}^7\text{Be}_p$ (dpm/m ³)	${}^7\text{Be}_{\text{tot}}$ (dpm/m ³)*	Particulate fraction (%)
GEOVIDE (North Atlantic Ocean, May-June 2014)							
1	5	1 μm sock	10166	4.27 \pm 0.93	5.99 \pm 1.48	350 \pm 30	2
13	5	1 μm sock	10815	BDL	BDL	150 \pm 30	
21	5	1 μm sock	4768	BDL	BDL	144 \pm 18	
32	5	1 μm sock	2899	BDL	BDL	210 \pm 20	
38	5	1 μm sock	6534	3.02 \pm 0.84	4.24 \pm 1.28	152 \pm 19	3
44	5	1 μm sock	2677	8.34 \pm 1.80	11.72 \pm 2.87	130 \pm 20	9
44	40	0.8 μm Supor	297	1.16 \pm 0.35	1.16 \pm 0.35		
44	80	0.8 μm Supor	428	BDL	BDL		
44	150	0.8 μm Supor	740	BDL	BDL		
44	300	0.8 μm Supor	425	BDL	BDL		
51/60	5	1 μm sock	1191	12.32 \pm 1.35	17.32 \pm 2.77	195 \pm 18	9
51/60	20	1 μm QMA	196	68.85 \pm 3.21	33.05 \pm 3.94	104 \pm 14	32
51/60	70	1 μm QMA	586	18.64 \pm 1.11	8.95 \pm 1.12	53 \pm 7	17

51/60	150	1 μm QMA	467	7.27 \pm 0.94	3.49 \pm 0.59	40 \pm 7	9
69	5	1 μm sock	3277	11.60 \pm 0.83	16.29 \pm 2.22	180 \pm 20	9
69	20	1 μm QMA	262	49.60 \pm 1.72	23.82 \pm 2.74	182 \pm 8	13
69	30	0.8 μm Supor	162	2.52 \pm 0.84	2.52 \pm 0.84		
69	60	0.8 μm Supor	280	BDL	BDL		
69	70	1 μm QMA	481	5.87 \pm 0.54	2.82 \pm 0.40	43 \pm 7	7
69	100	0.8 μm Supor	453	BDL	BDL	BDL	
77	5	1 μm sock	2349	4.65 \pm 0.54	6.54 \pm 1.08	212 \pm 17	3
77	10	0.8 μm Supor	105	12.76 \pm 1.28	12.76 \pm 1.28		
77	20	1 μm QMA				112 \pm 8	
77	45	1 μm QMA	842	4.42 \pm 0.28	2.12 \pm 0.27		
77	70	1 μm QMA	560	4.92 \pm 0.39	2.36 \pm 0.32	83 \pm 6	3
77	149	1 μm QMA	761	2.29 \pm 0.26	1.10 \pm 0.17		
77	200	0.8 μm Supor	511	BDL	BDL		

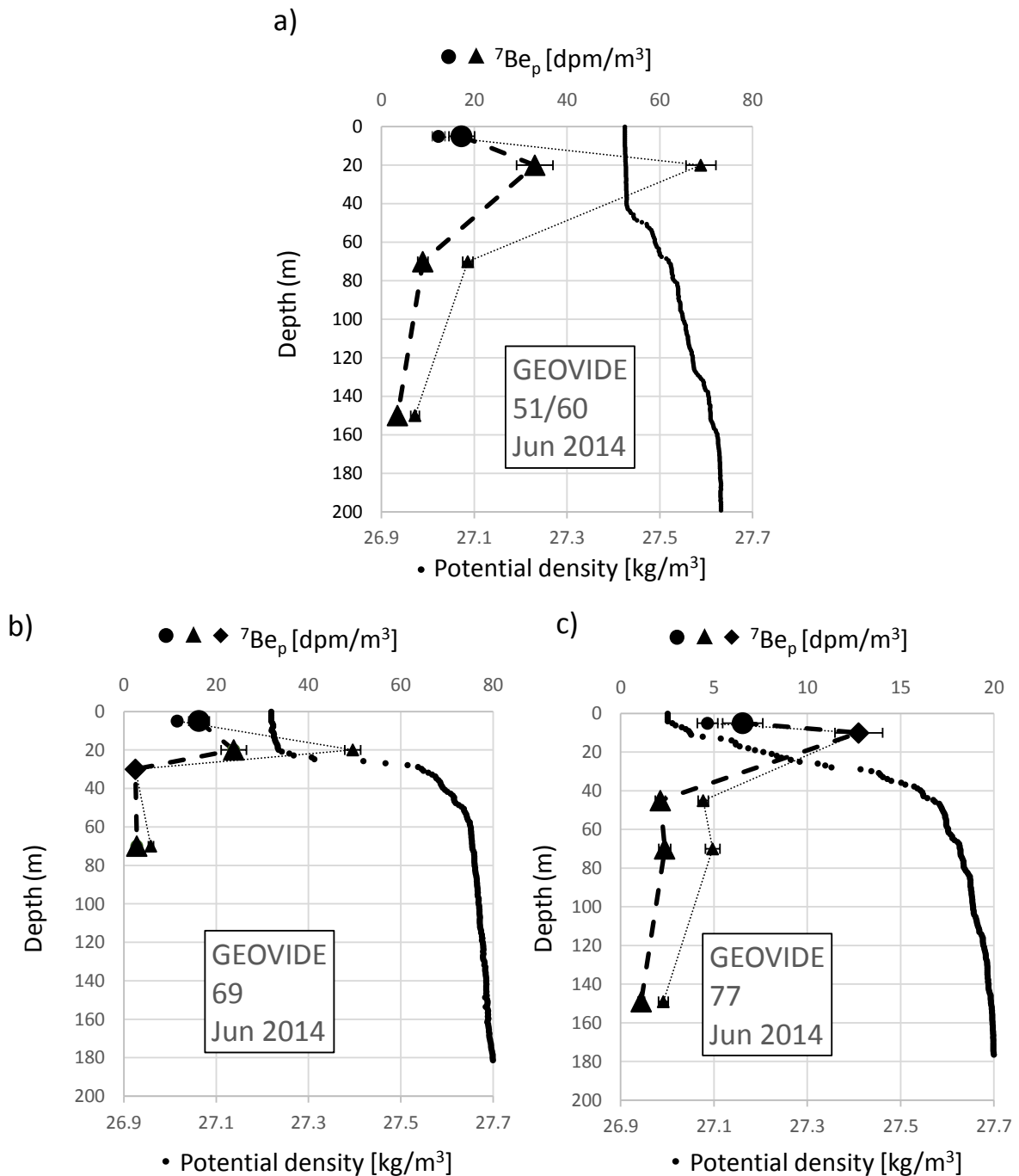
396 *Shelley et al. (2017)

397 **Table 2:** Particulate and total activities of ^7Be ($^7\text{Be}_p$, $^7\text{Be}_{\text{tot}}$, in dpm/m³) at GEOVIDE stations. Filter
398 types and filtered volume are also specified. Particulate activities (uncorrected and corrected following
399 filter types used) are reported (see Section 4.2). Total activities are from Shelley et al. (2017). The
400 particulate fraction (%) refers to the ratio between the corrected particulate ^7Be and total ^7Be activities.
401 BDL: Below Detection Limit.

402

403 **4.3. $^7\text{Be}_p$ in the subpolar North Atlantic Ocean (GEOVIDE)**

404 As observed for the other oceanic areas of the study, the $^7\text{Be}_p$ activity (corrected from the
405 filter biases as defined in Section 4.2; Table 2 and Figure 4) is higher in the mixed layer than
406 below at the GEOVIDE stations. At 5-m depth, the $^7\text{Be}_p$ activity varies from below the detection
407 limit in the Iberian, West European and south Iceland basins (stations 13, 21, and 32,
408 respectively) to ~ 17 dpm/m³ near the southern tip of Greenland in the western Irminger Sea and
409 in the central Labrador Sea (respectively, station 51/60 and station 69). The $^7\text{Be}_p$ activity
410 reaches 33 dpm/m³ and 24 dpm/m³ at 20 m at stations 51/60 and 69, respectively, in the lower
411 part of the mixed layer (Table 2 and Figure 4). Note that the filter correction reduces, but does
412 not completely suppress, the apparent $^7\text{Be}_p$ activity maximum observed in the lower part of the
413 mixed layer (Figure 4). Below the mixed layer, the $^7\text{Be}_p$ activity generally drops to a maximum
414 of ~ 3 dpm/m³, similarly to what is observed in the other oceanic areas of the study.



415

416 **Figure 4:** Profiles of particulate ^7Be activity ($^7\text{Be}_p$, in dpm/m^3) at GEOVIDE stations (uncorrected $^7\text{Be}_p$:
 417 small symbols and thin dotted line; corrected $^7\text{Be}_p$: large symbols and thick dashed line). Samples
 418 filtered through 1- μm pore size socks are identified by circles, 1- μm QMA filters by triangles, and 0.8-
 419 μm Supor filters by diamonds. Error bars for uncorrected activities represent ± 1 standard deviation
 420 (counting statistics). Error bars for corrected activities are calculated by propagating the errors in the
 421 uncorrected activities and the errors in the filter corrections. Note that the x-axis range for $^7\text{Be}_p$ is smaller
 422 for station 77. The potential density profile from CTD cast data is also shown for each station (in kg/m^3 ;
 423 small dots).

424

425

426 5. Discussion

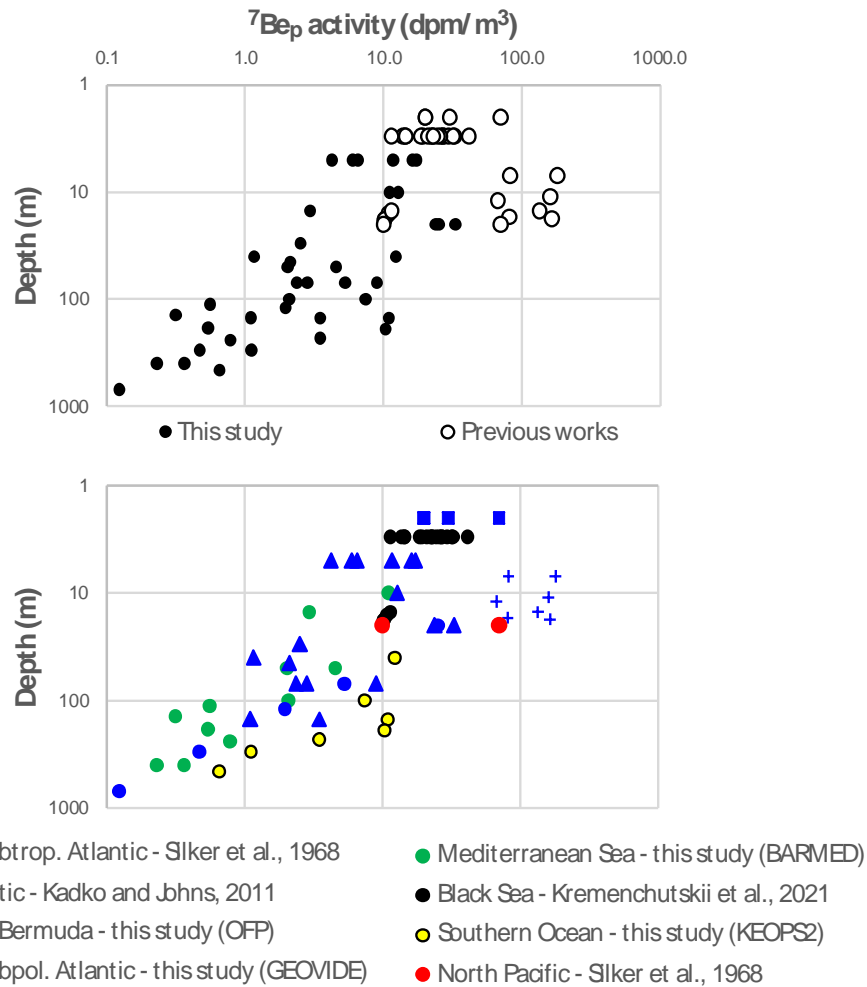
427 5.1. Comparison to previous $^7\text{Be}_p$ measurements

428 In this section, we compare the particulate ^7Be activities reported here with previous data
429 from the literature (Figure 5 and Table 3). Silker et al. (1968) reported $^7\text{Be}_p$ activities ranging
430 from below the detection limit (< 10 dpm/1000 L) to 70 dpm/1000 L in surface waters (2-20
431 m) of the Atlantic and Pacific oceans. Silker (1972a) published vertical profiles of ^7Be in the
432 upper 100 m of the Atlantic Ocean and concluded that less than 10% of the measured
433 radioactivity (i.e., < 90 dpm/m³) was found in the insoluble fraction. Similar findings were
434 reported in surface waters of the North Pacific (Silker, 1972b; < 65 dpm/m³). Andrews et al.
435 (2008) pumped ~1000 L of surface water from the Sargasso Sea through a 1- μm Hytrec filter
436 and found that the $^7\text{Be}_p$ activity was below the detection limit (< 35 dpm/m³). Kadko and Johns
437 (2011) reported $^7\text{Be}_p$ activities in the mixed layer (between 7 and 18 m depth) in the equatorial
438 Atlantic, with values ranging from below the detection limit (< 60 dpm/m³) to 180 dpm/1000
439 L. Finally, Kremenchutskii et al. (2021) determined the $^7\text{Be}_p$ activity of samples collected in
440 the Black Sea in the upper 65 m of the water column. The $^7\text{Be}_p$ values from that study ranged
441 from 11 to 41 dpm/m³ in surface (3 m) waters, from 10 to 11 dpm/m³ between 15 and 18 m,
442 and were below the detection limit (< 10 dpm/m³) between 20 and 65 m.

443 Overall, the $^7\text{Be}_p$ activities reported in this study are of the same orders of magnitude as
444 $^7\text{Be}_p$ activities reported in most previous studies (< 70 dpm/m³; Kremenchutskii et al., 2021;
445 Silker et al., 1968), although all are noticeably lower than the value of 180 dpm/m³ observed in
446 the equatorial Atlantic (Kadko and Johns, 2011; Figure 5). The $^7\text{Be}_p$ decrease with depth that is
447 generally observed in this study is also qualitatively consistent with previous measurements in
448 the Black Sea (Kremenchutskii et al., 2021). It is worth noting that, in the study of Kadko and
449 Johns (2011), particle samples were collected by passing 200 L of seawater through GF/F filters
450 made of borosilicate glass, a material which was shown to retain dissolved ^{234}Th , as for QMA
451 quartz filters (Benitez-Nelson et al., 2001). Our preliminary tests show that $^7\text{Be}_p$ activities
452 measured on QMA filters significantly exceed $^7\text{Be}_p$ activities measured on reference Supor
453 filters (section 4.2), which suggests that the $^7\text{Be}_p$ activities of equatorial Atlantic samples
454 reported in Kadko and Johns (2011) may be overestimated.

455

456



457

458 **Figure 5:** Compilation of open ocean ${}^7\text{Be}_p$ data (in dpm/m^3 , log scale) as a function of depth (in m, log
459 scale). Top panel shows previous data (open circles) and those from this study (black dots). Bottom
460 panel shows different oceanic areas: the Atlantic Ocean (blue), the Mediterranean Sea (green), the Black
461 Sea (black), the Southern Ocean (yellow), and the North Pacific (red).

462

Location	Sampled depths (m)	Filtration method	Material	Porosity (μm)	Diameter (mm)	Volume filtered (L)	${}^7\text{Be}_p$ range (dpm/m^3)	${}^7\text{Be}_p/{}^7\text{Be}_{\text{tot}}$ range (%)	Reference
North Atlantic	2	Millipore filtration unit	Plastic	0.3	305	1000-5000	20-70	6-15	Silker et al. (1968)
North Pacific	20	Millipore filtration unit	Plastic	0.3	305	1000-5000	10-70	4-24	Silker et al. (1968)
NW Atlantic	0-97	Millipore filtration unit	Glass	0.3	305?	not specified	<5-10%	NA	Silker (1972)
Sargasso Sea	3	HYTRESX filter	Polypropylene	1	NA	1000	BDL	NA	Andrews et al. (2008)
Eq. Atlantic	7-18	GF/F filter	Borosilicate glass	not specified	142	200	70-180	18-38	Kadko and Johns (2011)
Black Sea	3-18	Aquafilter FCPS1	Polypropylene	1	NA	2000-10000	10-40	5-13	Kremenchutskii et al. (2021)
Med. Sea	10-2200	Versapor	Acrylic copolymer	0.8	142	200-2400	0.1-11.1	NA	This study

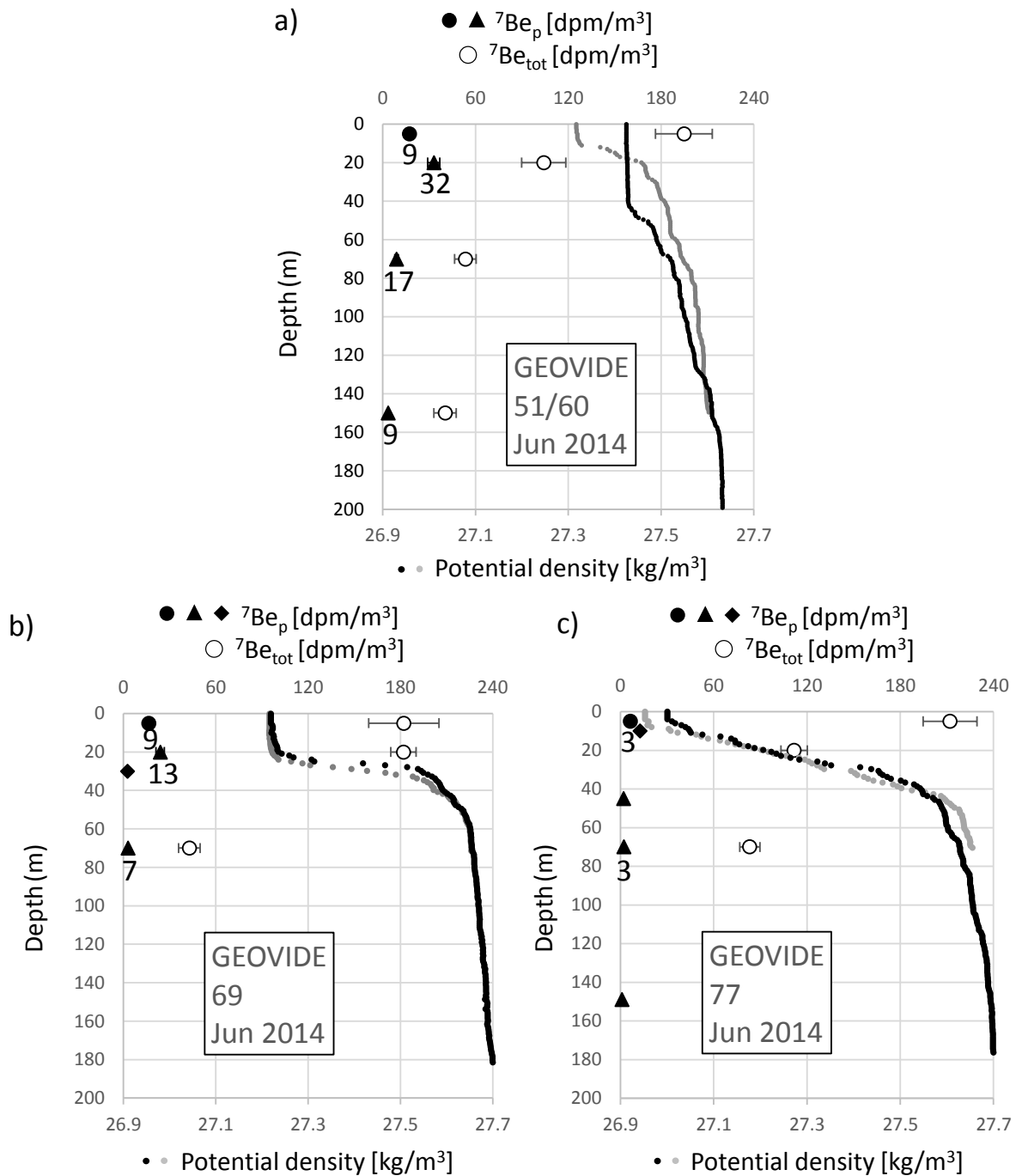
Sargasso Sea	20-4250	Versapor	Acrylic copolymer	0.8	142	100-2300	0.1-25.2	NA	This study
Southern Ocean	40-460	Supor	Hydrophilic polyethersulfone	0.8	142	100-300	0.7-12.3	NA	This study
North Atlantic	5	Pentek BP-410-1	Polypropylene	1	NA	1000-10000	4-17	2-9	This study
	>5-150	Supor	Hydrophilic polyethersulfone	0.8	142	100-300	1-13	NA	
	>5-150	QMA	Quartz	1	142	200-800	1-33	3-32	

463 *NA: not applicable

464 **Table 3:** Characteristics of $^7\text{Be}_p$ sampling methods and range of $^7\text{Be}_p$ values obtained in previous studies
465 and in this study.

466 5.2. Are the oceanic $^7\text{Be}_p$ activities significant?

467 By combining the corrected $^7\text{Be}_p$ activities at GEOVIDE stations and the total ^7Be activities
468 at the same stations, at the same depths, but from different casts (Shelley et al., 2017), the
469 fraction of total ^7Be that is bound to suspended particles can be tentatively estimated. From this
470 approach, we find that the particulate fraction would account for 2-9% of the total ^7Be activity
471 at a water depth of 5 m ($n = 6$), 13-32% at 20 m ($n = 2$), and 3-17% at 70 m ($n = 3$; Table 2;
472 Figures 6 and 7). Note that at station 77, the highest $^7\text{Be}_p$ activity is observed at 10 m (Supor
473 filter), at a depth where $^7\text{Be}_{\text{tot}}$ was not determined (Table 2). Considering the $^7\text{Be}_{\text{tot}}$ activity
474 determined either at 5 m or 20 m (respectively, above and below the depth of the particulate
475 sample), we find that the particulate fraction would amount to 6 or 11%, respectively, in
476 agreement with the values reported above. These fractions are similar to those estimated for
477 surface waters in (i) the North Atlantic at 2 m (6-15%; Silker et al., 1968, using 0.3- μm pore
478 size filters; Table 3), (ii) the Black Sea between 3 and 18 m (5-13%; Kremenchutskii et al.,
479 2021, using 1- μm pore size filters), (iii) the North Pacific at 20 m (4-24%; Silker et al., 1968),
480 and (iv) the equatorial Atlantic between 7 and 18 m (18-38%; Kadko and Johns, 2011; Figure
481 7).

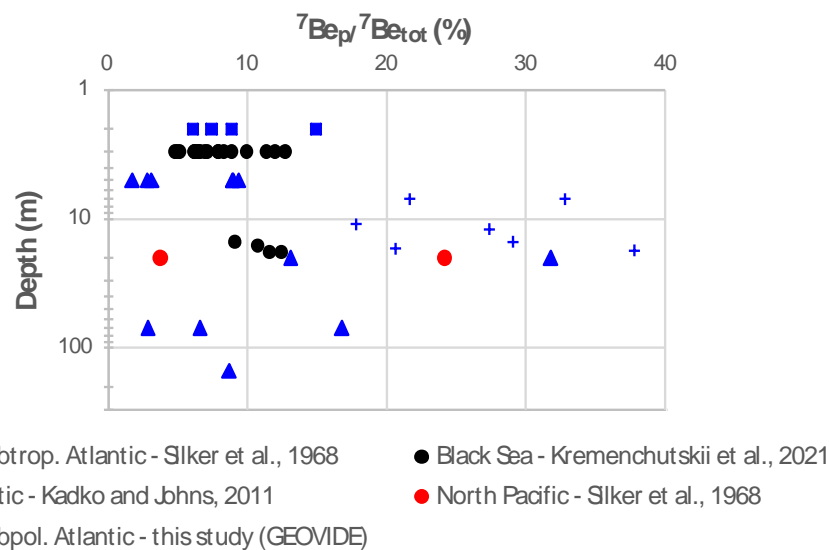


482

483 **Figure 6:** Profiles of corrected particulate ${}^7\text{Be}$ activity (${}^7\text{Be}_p$, in dpm/m³; solid symbols) and total ${}^7\text{Be}$
 484 activity (${}^7\text{Be}_{\text{tot}}$, in dpm/m³; open circles; Shelley et al., 2017) at GEOVIDE stations. For ${}^7\text{Be}_p$, samples
 485 filtered through 1- μm pore size socks are identified by circles, 1- μm QMA filters by triangles, and 0.8-
 486 μm Supor filters by diamonds. Numbers below data points are the ratio of particulate ${}^7\text{Be}$ to total ${}^7\text{Be}$
 487 (in %). Error bars for the corrected ${}^7\text{Be}_p$ are calculated by propagating the errors in the uncorrected ${}^7\text{Be}_p$
 488 and the errors in the filter corrections. Potential density profiles from CTD cast data are also shown for
 489 each station (in kg/m³; small grey dots for the cast associated with ${}^7\text{Be}_{\text{tot}}$ sample collection, small black
 490 dots for ${}^7\text{Be}_p$).

491

492



494

495 **Figure 7:** Compilation of estimates of the $^7\text{Be}_p/^7\text{Be}_{\text{tot}}$ activity ratio in the open ocean (in %, linear scale)
 496 as a function of depth (in m, log scale). Data from the Atlantic Ocean are in blue, in black for the Black
 497 Sea, and in red for the North Pacific Ocean.

498

499 To further assess the significance of the ^7Be particulate fraction in comparison to the
 500 dissolved fraction, we calculate the inventory (vertical integral) of $^7\text{Be}_p$ activity at stations
 501 where inventories of total ^7Be activities were reported in the literature (i.e., at the OFP and
 502 GEOVIDE stations). $^7\text{Be}_p$ inventories are calculated by integrating the $^7\text{Be}_p$ activities at the
 503 different sampling depths using the trapezoidal formula. For this calculation, we assume that
 504 (i) $^7\text{Be}_p$ activity at the surface ($z = 0$ m) is equal to the activity determined at the shallowest
 505 sampling depth and (ii) $^7\text{Be}_p$ activity is 0 dpm/m³ at 1500 m at OFP and is 0 dpm/m³ at the
 506 maximum sampling depth at GEOVIDE stations (see Table 4; Shelley et al., 2017). At all
 507 stations, the error in the $^7\text{Be}_p$ inventory is estimated by propagating the errors in the individual
 508 $^7\text{Be}_p$ measurements (Bevington and Robinson 1992).

509 At the OFP station, we find that the $^7\text{Be}_p$ inventory amounts to 1830 ± 340 dpm/m², which
 510 represents only about 5% of the total ^7Be inventory of $\sim 40,000$ dpm/m² at this station (Aaboe
 511 et al., 1981; Kadko et al., 2015; Kadko and Prospero, 2011; Silker, 1972b).

512 The $^7\text{Be}_p$ inventories at the GEOVIDE stations 51/60, 69 and 77 vary significantly, ranging
 513 from 530 ± 70 dpm/m² at station 77 to 2060 ± 270 dpm/m² at station 51/60, while $^7\text{Be}_{\text{tot}}$
 514 inventories are comparable at these three stations ($\sim 10,000$ dpm/m²; Shelley et al., 2017). The
 515 errors of 70 and 270 dpm/m² reflect the uncertainties in the $^7\text{Be}_p$ measurements and in the filter

516 biases (Bevington and Robinson, 1992). The ratio of $^7\text{Be}_p$ inventory to total ^7Be inventory varies
 517 therefore widely across the GEOVIDE stations, from 5% at station 77 to up to 19% at station
 518 51/60 (Table 4). The upper end of this range (19%) should be considered with caution,
 519 considering that it results from $^7\text{Be}_{\text{tot}}$ and $^7\text{Be}_p$ activities estimated from samples from different
 520 casts under different hydrographic conditions (Station 51/60). A number of factors that could
 521 explain the wide range are discussed in the following subsections.

Station	^7Be inventory depth range* [m]	$^7\text{Be}_{\text{tot}}$ inventory* [dpm/m ²]	$^7\text{Be}_p$ inventory [dpm/m ²]	Relative $^7\text{Be}_p$ inventory [%]	$^7\text{Be}_{\text{tot}}$ flux* [dpm/m ² /d]	$^7\text{Be}_p$ flux [dpm/m ² /d]
51/60	0-175	11000 ± 300	2060 ± 270	19 ± 2	143 ± 4	27 ± 4
69	0-87	9700 ± 300	640 ± 90	7 ± 1	126 ± 4	8 ± 1
77	0-150	10500 ± 300	530 ± 70	5 ± 1	136 ± 4	7 ± 1

522 *Shelley et al. (2017)

523 **Table 4:** Inventories of $^7\text{Be}_{\text{tot}}$ (in dpm/m²) and corrected $^7\text{Be}_p$ (in dpm/m² and in % of $^7\text{Be}_{\text{tot}}$) at
 524 GEOVIDE stations 51/60, 69, and 77, and equivalent surface fluxes from atmospheric deposition (in
 525 dpm/m²/d). $^7\text{Be}_{\text{tot}}$ water column inventories and surface fluxes are from Shelley et al. (2017).

526 **5.3. Which processes shape the vertical profiles of $^7\text{Be}_p$?**

527 The higher $^7\text{Be}_p$ activities in the ocean mixed layer than below that are found in this study
 528 are consistent with the combined effects of atmospheric deposition and radioactive decay. The
 529 decrease in $^7\text{Be}_p$ activity with depth below the mixed layer (Figures 2 and 6) is expected to
 530 result, at least partly, from radioactive decay, as is the case for $^7\text{Be}_{\text{tot}}$ (Silker, 1972b, 1972a).
 531 One can wonder, however, if other processes may also influence the vertical distribution of $^7\text{Be}_p$
 532 in the upper ocean. Since Be is a particle-reactive element, exchanges of ^7Be between the
 533 dissolved and particulate phases may occur and impact the vertical distribution of $^7\text{Be}_p$ as well
 534 as the $^7\text{Be}_p$ inventory in the water column. In order to assess whether processes other than
 535 surface deposition impact the $^7\text{Be}_p$ activities measured during GEOVIDE, we compare the $^7\text{Be}_p$
 536 deposition fluxes estimated from aerosol samples collected during GEOVIDE (Shelley et al.,
 537 2017) with $^7\text{Be}_p$ deposition flux estimated from our $^7\text{Be}_p$ inventories.

538 Shelley et al. (2017) estimated surface $^7\text{Be}_{\text{tot}}$ deposition fluxes from $^7\text{Be}_{\text{tot}}$ inventories
 539 determined in the water column, following a method which assumes that the only loss of ^7Be
 540 from the water column is radioactive decay (Kadko et al. 2015). A similar method is applied
 541 here for $^7\text{Be}_p$ deposition. It assumes that (i) $^7\text{Be}_p$ is supplied to the ocean only via atmospheric
 542 deposition (dry deposition + particulate fraction of wet deposition), (ii) no particulate ^7Be is
 543 lost in the water column by particle degradation (dissolution and/or remineralization) or by
 544 desorption, and (iii) no particulate ^7Be is gained in the water column by adsorption of dissolved

545 ^7Be onto particles (scavenging). Collectively, assumptions (i)-(iii) imply that, at steady state,
546 the flux of $^7\text{Be}_p$ at the sea surface from atmospheric deposition is balanced by the $^7\text{Be}_p$ decay
547 rate integrated over the water column.

548 The above method gives atmospheric deposition fluxes of $^7\text{Be}_p$ of 27 dpm/m²/d at station
549 51/60, 8 dpm/m²/d at station 69, and 7 dpm/m²/d at station 77 (Table 4). These fluxes are
550 compared below to the dry deposition fluxes of $^7\text{Be}_p$ that have been derived at these stations
551 from the analysis of aerosols sampled on the ship during GEOVIDE (Shelley et al., 2017).

552 Consider first the comparison for stations 69 and 77. Our estimates of $^7\text{Be}_p$ deposition are
553 two to three times lower than those derived from aerosols for station 69 (25 dpm/m²/d) and
554 station 77 (16 dpm/m²/d; Table 2 of Shelley et al., 2017). Several factors could explain the
555 discrepancies between the $^7\text{Be}_p$ deposition flux estimated from aerosols and from $^7\text{Be}_p$
556 inventories in the water column. They are similar to those discussed by Shelley et al. (2017) in
557 their effort to explain the discrepancies between estimates of $^7\text{Be}_{\text{tot}}$ deposition based on (i)
558 precipitation and aerosol samples collected on the ship and (ii) $^7\text{Be}_{\text{tot}}$ inventories in the water
559 column.

560 For example, the dry ^7Be fluxes estimated during GEOVIDE were obtained from the
561 analysis of aerosols sampled during short periods of time (~2 days; Shelley et al., 2017).
562 Estimates of dry ^7Be deposition based on measurements collected over a period of few days
563 may not be adequate to interpret measurements of ^7Be activity in the water column, which
564 should integrate the collective effects of surface inputs extending over several months. The
565 different time scales characterizing aerosol sampling and the evolution of ^7Be in the water
566 column could explain why the estimates of $^7\text{Be}_p$ deposition derived from $^7\text{Be}_p$ water column
567 inventories are different than those derived from ship data (Shelley et al., 2017), although why
568 the former are lower than the latter remains unclear (see below).

569 At least three other factors could be responsible for the relatively low $^7\text{Be}_p$ deposition flux
570 estimated from $^7\text{Be}_p$ inventory at stations 69 and 77. A first factor is a loss of ^7Be from the
571 particulate phase to the dissolved phase owing to the dissolution of aerosols in the upper water
572 column. A second factor is the downward export of particulate ^7Be to depth due to some
573 combination of particle aggregation and gravitational settling. Evidence of high POC export at
574 stations 69 and 77 is provided by the observation of strong ^{234}Th disequilibria with its
575 radioactive parent (Lemaitre et al., 2018a). Shelley et al. (2017) reported that ^7Be deposition
576 fluxes derived from water column $^7\text{Be}_{\text{tot}}$ inventories are generally lower than ^7Be deposition

577 fluxes from precipitation at GEOVIDE stations (their Table 2), and they also suggested that the
578 discrepancy could be explained by scavenging of ^7Be onto sinking particles. A third factor is
579 the overestimation of the dry deposition velocity assumed by Shelley et al. (2017). In this earlier
580 study, a deposition velocity of 0.3 cm/s was assumed, but the authors acknowledged that the
581 relative error in this parameter could be up to 300%.

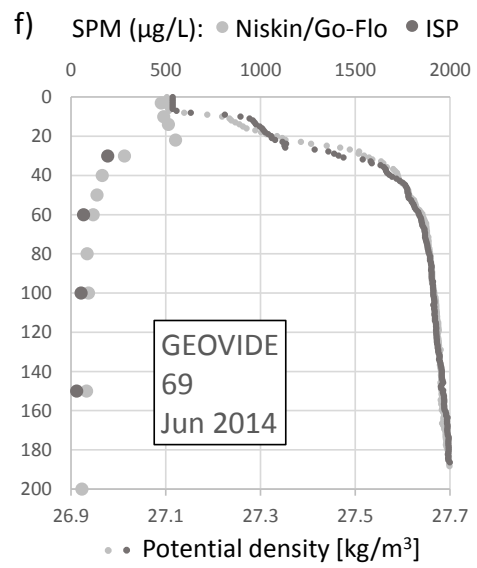
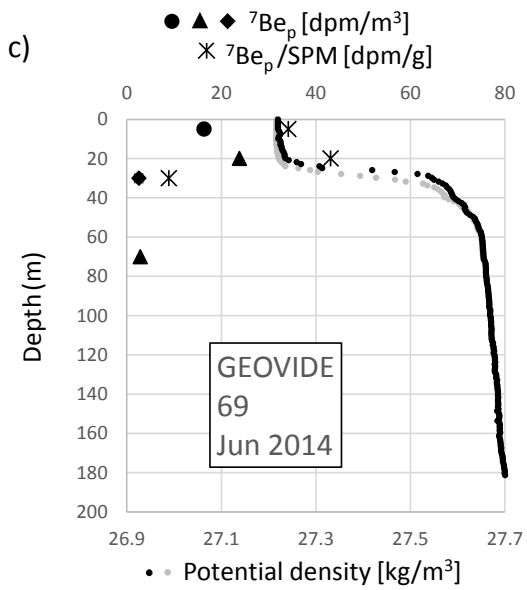
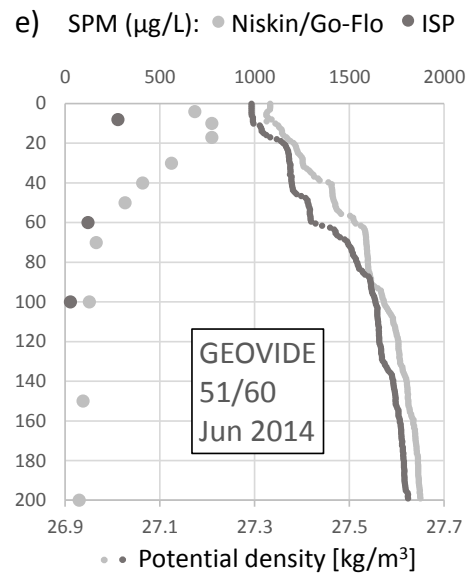
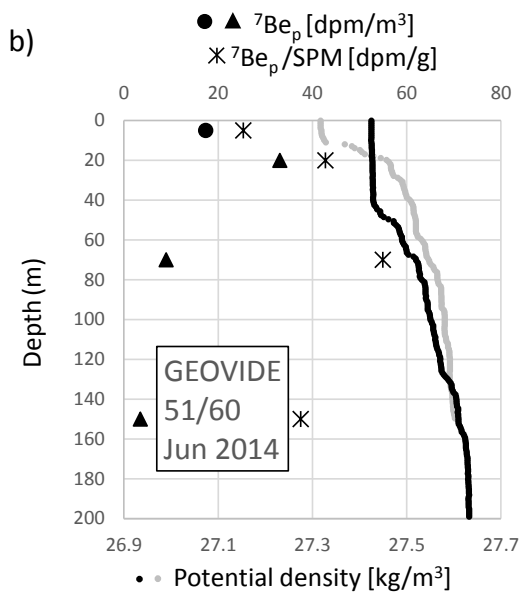
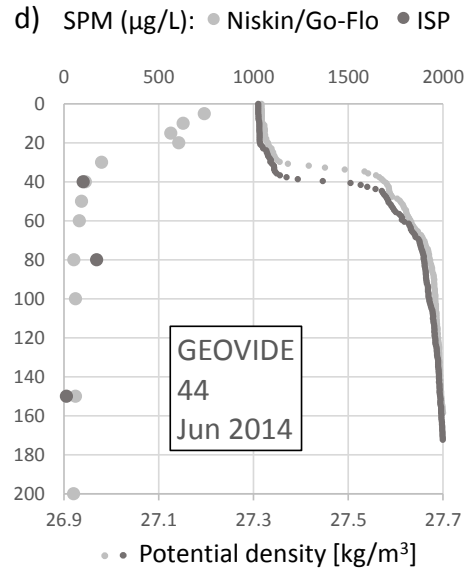
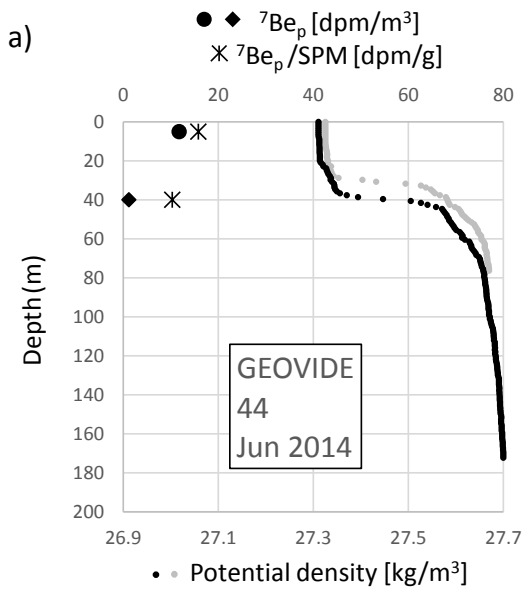
582 We now compare the estimate of $^7\text{Be}_p$ surface flux based on $^7\text{Be}_p$ water column inventory
583 with that based on aerosol data at station 51/60. The ^7Be activity in the aerosol sample collected
584 near station 51/60 was below the detection limit (Shelley et al., 2017), while it is at this station
585 that we estimate the highest atmospheric deposition flux from the $^7\text{Be}_p$ inventory (27 dpm/m²/d)
586 among stations 51/60, 69 and 77. The higher $^7\text{Be}_p$ surface flux deduced from the $^7\text{Be}_p$ inventory
587 compared to that deduced from aerosol data near station 51/60 could also be due to a number
588 of factors. These include, again, the different time scales captured by the atmospheric and
589 oceanic samples, but also the adsorption of dissolved ^7Be onto particles, and a significant source
590 of particulate ^7Be from wet deposition, which is unaccounted for in the calculation of dry $^7\text{Be}_p$
591 deposition from ship data. On this note, precipitation events did occur near station 51/60 during
592 the first half of May, 2014 (see Shelley et al., 2017; their Figure S5).

593 **5.4. Exchange of ^7Be between the dissolved and particulate phases**

594 It is instructive to consider the $^7\text{Be}_p$ activities per mass of particles in order to assess how
595 the $^7\text{Be}_p$ activities at GEOVIDE stations relate to particle concentrations. The $^7\text{Be}_p$ activities per
596 volume of water are divided by the suspended particle matter concentrations ([SPM]; Figure 8)
597 to derive specific $^7\text{Be}_p$ activities (in dpm per gram of particles) at these stations. Interestingly,
598 at station 51/60, the specific $^7\text{Be}_p$ activities below the mixed layer (70 and 150 m) are similar
599 to that in the mixed layer (20 m), whereas the $^7\text{Be}_p$ activities (per volume of water) are
600 significantly lower below the mixed layer than at 20 m (SPM concentrations were not
601 determined at station 77). Thus, the amount of ^7Be bound to particles per mass of particles is
602 approximately the same in the mixed layer and in the upper thermocline.

603

604



606 **Figure 8:** (left panels) Profiles of ${}^7\text{Be}_p$ (in dpm/m³; black markers: circles for 1- μm pore size sock
607 samples, triangles for 1- μm QMA filter samples, diamonds for 0.8- μm Supor filters, with filter
608 corrections applied) and ${}^7\text{Be}_p/\text{SPM}$ (in dpm/g; asterisks) at GEOVIDE stations (a) 44, (b) 51/60, and (c)
609 69. Potential density profiles from CTD cast data are also shown (in kg/m³; small grey dots for the cast
610 associated with ${}^7\text{Be}_{\text{tot}}$ sample collection, small black dots for ${}^7\text{Be}_p$). (right panels) Profiles of suspended
611 particulate matter concentration (SPM, in $\mu\text{g/L}$) at GEOVIDE stations (d) 44, (e) 51/60, and (f) 69,
612 determined from particles collected using Niskin or Go-Flo bottles (Lagarde et al., submitted; light grey
613 dots) and *in situ* pumps (ISP; Tang et al., 2018; dark grey dots), together with their associated potential
614 density profiles (in kg/m³; small dots).

615

616 We combine the measurements of particulate ${}^7\text{Be}$, total ${}^7\text{Be}$, and SPM which are available
617 at GEOVIDE stations to derive tentative estimates of the distribution coefficient for ${}^7\text{Be}$:

$$618 \quad K_d = \frac{[{}^7\text{Be}_p]}{[{}^7\text{Be}_d] \times [\text{SPM}]} \quad (1)$$

619 Here $[{}^7\text{Be}_d]$ is the activity of ${}^7\text{Be}$ in the dissolved phase, deduced from the difference between
620 total activity $[{}^7\text{Be}_{\text{tot}}]$ (Shelley et al., 2017) and corrected particulate activity $[{}^7\text{Be}_p]$ (see Table
621 2). The distribution coefficient K_d quantifies the solid/solution partitioning of the nuclide in the
622 water column: a higher value of K_d indicates that a greater proportion of ${}^7\text{Be}$ is bound to
623 particles for the same amount of particles. SPM concentrations are expressed in g/cm^3 in order
624 to facilitate comparison to K_d values from previous studies (e.g., Baskaran et al., 1997; Chuang
625 et al., 2013). Thus, the K_d values reported in this paper are in cm^3/g . Estimates of $\log K_d$ (base
626 10) are reported in Table 5, together with SPM concentrations. Note that our K_d values should
627 be regarded as tentative estimates given that the concentrations of ${}^7\text{Be}_p$, ${}^7\text{Be}_{\text{tot}}$ and SPM
628 determined at GEOVIDE stations are generally for distinct casts and that, at some stations, these
629 casts exhibited different density profiles (Figure 8).

Station	Depth (m)	SPM (10^{-9} g/cm^3)	$\log K_d$
1	5	111	5.19
38	5	261	5.04
44	5	741	5.14
51/60	5	684	5.15
51/60	20	773	5.78
51/60	70	163	6.09
51/60	150	93	6.01
69	5	477	5.30
69	20	553	5.51

630 **Table 5:** Suspended particulate matter concentration (SPM; in 10^{-9} g/cm^3) and tentative estimates of the
631 distribution coefficient K_d for ${}^7\text{Be}$ at GEOVIDE stations (expressed as \log_{10} of K_d).

632 The $\log K_d$ values for GEOVIDE samples vary from 5.04 (surface sample; station 38) to
633 6.09 (70 m; station 60). They overlap with (i) values from 4.92 to 6.16 determined from ^{10}Be
634 measurements from the Middle Atlantic Bight, the equatorial Pacific, and the Pacific sector of
635 the Southern Ocean (Chase et al., 2002) and (ii) values from 5.04 to 5.30 determined from
636 recent ^7Be measurements in the Black Sea (Kremenchutskii et al., 2021). On the other hand,
637 they are higher, or slightly higher, than values between 3 and 5 obtained from samples collected
638 at the OFP station (Chuang et al., 2013), in Tampa Bay (Florida, USA; Baskaran and
639 Swarzenski, 2007), and in estuaries of the Sabine-Neché (Texas, USA) and the Loire (France;
640 Baskaran et al., 1997; Ciffroy et al., 2003).

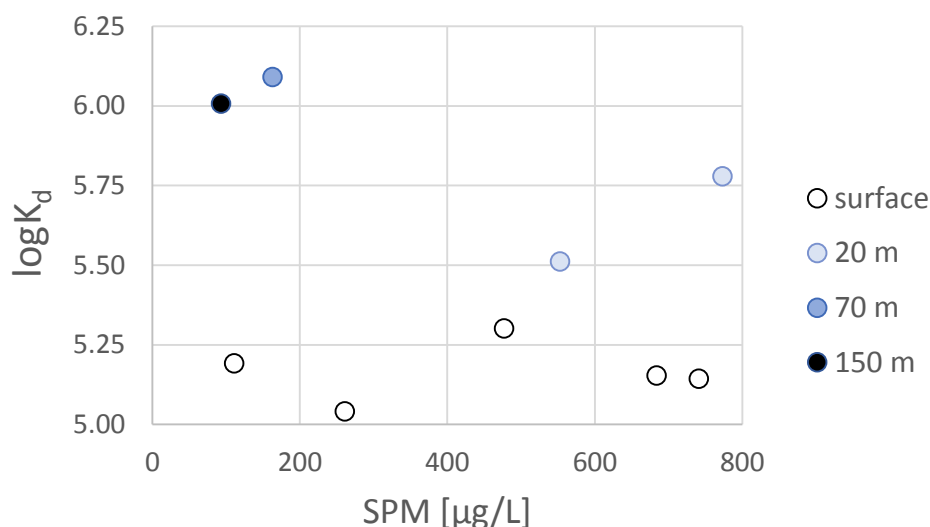
641 The distribution coefficient of a trace element in seawater can decrease with particle
642 concentration, the so-called particle concentration effect (e.g., Honeyman and Santschi, 1988).
643 The relatively large K_d values for ^7Be estimated at GEOVIDE stations may result from lower
644 SPM concentrations ($< 1 \text{ mg/L}$, as commonly found in open ocean waters; Guo et al., 1997),
645 since previous K_d estimates for ^7Be mostly pertain to near-shore or estuarine waters. Lower K_d
646 values are commonly observed in freshwater and estuarine environments, which are generally
647 characterized by higher [SPM] (tens to thousands of mg/L ; e.g. Baskaran and Santschi, 1993;
648 Woźniak et al., 2010) than in open ocean waters, a pattern often attributed to the particle
649 concentration effect (Benoit and Rozan, 1999; D. Tang et al., 2002). However, as observed in
650 estuarine environments (e.g. Baskaran et al., 1997), the K_d values estimated at GEOVIDE
651 stations do not show a significant correlation with [SPM] (Figure 9; $R^2 = 0.09$, $p\text{-value} = 0.43$).
652 On the other hand, the $\log K_d$ values at GEOVIDE stations show a clear difference between
653 surface and deep waters: they amount to 5.2 ± 0.1 (average ± 1 standard deviation) for surface
654 samples (5 m) and 6.1 ± 0.1 for the deepest samples (70-150 m). This difference in $\log K_d$
655 between surface and deep waters suggests that a higher fraction of dissolved ^7Be is found in the
656 particulate phase in deep waters than in surface waters, when differences in particle
657 concentration are taken into account. Note, however, that for station 51/60, the particulate
658 fraction at each depth may be biased because $^7\text{Be}_p$ and $^7\text{Be}_{\text{tot}}$ were measured at different times
659 under different hydrographic conditions (Figure 6a).

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664
 665 **Figure 9:** Distribution coefficient for ⁷Be (logK_d), as a function of SPM concentrations at GEOVIDE
 666 stations. The distribution coefficient could be determined only for stations and depths where [SPM],
 667 particulate ⁷Be activity, and total ⁷Be activity were all determined (see Table 5). ⁷Be_d activities were
 668 determined by difference (total ⁷Be minus corrected particulate ⁷Be).

669
 670 **6. Conclusions**

671 The purpose of this study is to better assess the significance of particulate ⁷Be (⁷Be_p) in the
 672 oceanic cycle of ⁷Be and for the use of ⁷Be as a tracer of atmospheric inputs and dynamical
 673 processes in the upper water column. Using low-background gamma-ray spectrometers
 674 operating in underground facilities, we produce relatively precise measurements of ⁷Be activity
 675 in suspended particles collected in four different oceanic regions: the subpolar North Atlantic,
 676 the Sargasso Sea, the western Mediterranean Sea, and the Indian sector of the Southern Ocean.
 677 At three stations in the subpolar North Atlantic (GEOVIDE stations), we consider our results
 678 in the light of published measurements of total ⁷Be activity, estimates of ⁷Be_p deposition, and
 679 estimates of particulate matter concentration, yielding insights into the solid-solution
 680 partitioning of ⁷Be and into other aspects of the oceanic cycle of ⁷Be.

681 We find that, in each oceanic region, the ⁷Be_p activity generally decreases with depth, with
 682 maxima in the mixed layer and minima in the thermocline. The trend is particularly robust at
 683 stations where the particulate samples dedicated to ⁷Be have been collected by filtration through
 684 the same filter type at all depths (stations OFP, DYFAMED, and A3-2). Preliminary tests based
 685 on particulate samples collected at a near-coastal site in the western Mediterranean Sea, show
 686 that ⁷Be_p measured on QMA filters and polypropylene filters are, respectively, greater and
 687 smaller than ⁷Be_p measured on the Supor filters used in GEOTRACES. Results from these tests

688 are used to apply filter-specific corrections to the $^7\text{Be}_p$ measurements at GEOVIDE stations,
689 where measurements of total ^7Be activity are also available but where QMA and polypropylene
690 filters were used to collect the particulate material at different depths.

691 We find that, at GEOVIDE stations, in agreement with previous studies, the corrected $^7\text{Be}_p$
692 activity accounts for less than 10% of the total ^7Be activity in surface waters (water depth of 5
693 m; 6% on average, $n = 6$) and for a larger proportion of total ^7Be in the lower part of the mixed
694 layer (20 m; 22% on average, $n = 2$). Below the mixed layer, in the thermocline, the corrected
695 $^7\text{Be}_p$ fraction generally also accounts for less than 10% (70 m; 9% on average, $n = 3$). The
696 corrected $^7\text{Be}_p$ is estimated to represent 5-19% of the total ^7Be inventory in the water column,
697 suggesting that the amount of ^7Be bound to particles is small but not insignificant.

698 Evidence gathered at GEOVIDE stations suggests that ^7Be bound to marine particles may
699 not originate exclusively from atmospheric deposition. The $^7\text{Be}_p$ deposition fluxes derived from
700 water column inventories are noticeably lower, or higher (depending on location), than those
701 derived from the $^7\text{Be}_p$ activity of aerosols and an assumed dry deposition velocity. Differences
702 could be explained by (i) the different time scales captured by the atmospheric and oceanic
703 samples, (2) the uncertainty in the dry deposition velocity, (3) an unaccounted significant source
704 of particulate ^7Be from wet deposition, (4) adsorption of dissolved ^7Be onto particulate matter,
705 and/or (5) a release of particulate ^7Be into solution. Shelley et al. (2017) also pointed out, from
706 a comparison between different deposition estimates, that water column scavenging of ^7Be
707 could have been significant at some GEOVIDE stations.

708 Evidence at GEOVIDE stations also suggests that the sorptive properties of particles with
709 respect to ^7Be may be different in the mixed layer and in the thermocline. Distribution
710 coefficients of ^7Be at these stations are estimated to be systematically higher in the thermocline
711 than in the mixed layer. Although the coefficient estimates suffer from relatively large
712 uncertainties, due in particular to differences in sampling, they do suggest that a larger fraction
713 of ^7Be occurs in the particulate phase in the thermocline, when differences in particle
714 concentration between surface and deep waters are considered.

715 The results reported in this paper support the notion that ^7Be may undergo an exchange
716 between the dissolved and particulate phases in the ocean. However, the implications of these
717 findings for the use of ^7Be as a tracer of oceanic processes and surface deposition are unclear.
718 The assumptions underlying the use of ^7Be as a tracer of oceanic and surface processes depend
719 not only on the affinity of ^7Be for marine particles, but also on the magnitude of particulate ^7Be

720 export to the deep ocean compared to radioactive decay. Future research should thus focus on
721 quantifying the downward export of $^7\text{Be}_p$ to deep waters, and on assessing its temporal and
722 spatial variability. The variability of the $^7\text{Be}_p$ and $^7\text{Be}_d$ pools should also be assessed in order to
723 investigate how inventories vary with time and space. A better understanding of the temporal
724 and spatial variability of (wet and dry) ^7Be deposition to the sea surface is also needed.
725 Moreover, our results raise the necessity to further assess the influence of different filter types
726 on the measurement of $^7\text{Be}_p$ activity in oceanic samples. Finally, future sampling programs
727 should aim to collect seawater and particulate samples at the same locations, at the same depths,
728 and at the same time. Using the present dataset, a companion paper further explores the
729 significance of particle scavenging and export for the cycling of ^7Be in the ocean (Lerner et al.,
730 in preparation).

731

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754

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