1 High abundances of microplastic pollution in deep-sea sediments:

- **Evidence from Antarctica and the Southern Ocean**
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17 Author contributions

- 18 EMC, JDS, and KK designed the study. JDS and KL collected the sediment samples. EMC,
- 19 KK, JD and SME processed the samples and conducted the analysis. All authors contributed
- 20 to the writing.

Plastic pollution in Antarctica and the Southern Ocean has been recorded in scientific literature since the 1980s; however, the presence of microplastic particles (< 5 mm) is less understood. Here, we aimed to determine whether microplastic accumulation would vary among Antarctic and Southern Ocean regions through studying 30 deep-sea sediment cores. Additionally, we aimed to highlight whether microplastic accumulation was related to sample depth or the sediment characteristics within each core. Sediment cores were digested and separated using a high-density sodium polytungstate solution (SPT) and microplastic particles were identified using micro-Fourier-transform infrared spectroscopy (µFTIR). Microplastic pollution was found in 93% of the sediment cores (28/30). The mean (\pm SE) microplastics per gram of sediment was 1.30 ± 0.51 , 1.09 ± 0.22 , and 1.04 ± 0.39 MP/g, for the Antarctic Peninsula, South Sandwich Islands, and South Georgia, respectively. Microplastic fragment accumulation correlated significantly with the percentage of clay within cores, suggesting that microplastics have similar dispersion behaviour to low density sediments. Although no difference in microplastic abundance was found among regions, the values were much higher in comparison to less remote ecosystems, suggesting that the Antarctic and Southern Ocean deep-sea accumulates higher numbers of microplastic pollution than previously expected. Keywords: Antarctica; Southern Ocean; µFTIR; fragments; sediment grain size; synthetic polymers

Abstract

1. Introduction

57	Despite being one of the most remote regions of the planet, with oceanic and atmospheric
58	circulation enclosing the continent from the rest of the Southern Ocean (Bargagli, 2008),
59	Antarctica has a well-documented history of anthropogenic pollution (Szopińska et al. 2016).
60	Scientific articles surrounding the presence of plastic debris in Antarctica can be found dating
61	back to the 1980s, with reports highlighting the entanglement of Antarctic fur seals
62	(Arctocephalus gazella) in discarded plastic waste (Bonner & McCann, 1982), and the
63	ingestion of plastic particles by breeding petrels (van Francker & Bell, 1988). Between the
64	years 2000 and 2001, plastic debris in excess of 6000 items washed up on the shore of sub-
65	Antarctic islands over a six month period (Eriksson et al. 2013), and more recently it was
66	estimated that an average of 1794 items/km ² of plastic debris are floating at sea around the
67	Antarctic Peninsula (Lacerda et al. 2019). Additionally, the long term monitoring of
68	anthropogenic debris reaching the shores of remote islands in the Scotia Sea region of the
69	Southern Ocean has helped to track the accumulation of plastic over time, with over 350 kg
70	recorded from 1989-2019 (Waluda et al. 2020).
71	Microplastic pollution (< 5 mm) is widely regarded by scientists and citizens alike as being a
72	potential threat to marine biodiversity and ecosystem functions (Henderson & Green, 2020).
73	Within Antarctica, the ingestion of microplastics has been described in species from a range
74	of trophic levels, from benthic invertebrates (Sfriso et al. 2020) to top predators such as the
, . 75	gentoo penguin (<i>Pygoscelis papua</i> ; Bessa et al. 2019). The ingestion of microplastic particles
76	has shown to have negative effects on a number of marine species from different
77	environments in laboratory-based studies (Cunningham & Sigwart, 2019). Additionally,
, , 78	microplastic pollution is known to act as a vector for the transportation of persistent organic
79	pollutants (POPs; Rodrigues et al. 2019) which may have adverse effects through trophic
30	transfer and subsequent bioaccumulation in top predators (Durante et al. 2016). Although
31	microplastic pollution has been described extensively in a range of habitats from intertidal
32	rocky shores (Ehlers & Ellrich, 2020) to deep sea trenches (Zhang et al. 2020) globally, the
33	literature describing the presence of microplastic pollution from Antarctica and the Southern
84	Ocean is limited. Previous studies have identified microplastic pollution in surface waters of
85	the Antarctic Peninsula (Lacerda et al. 2019; Suaria et al. 2020) and shallow coastal
36	sediments of the Ross Sea (Munari et al. 2017), with one study sampling a small number of
37	deep-sea sediments from regions north of the Polar Frontal Zone (PFZ; Van Cauwenberghe et
88	al. 2013).

In terms of deep-sea microplastic research, the Antarctic and Southern Ocean regions below the PFZ remain unstudied. As the Arctic deep-sea has shown to represent a major sink for microplastic pollution (Tekman et al. 2020), and as the Antarctic and Southern Ocean deepsea below the PFZ represent a more isolated system, the presence and accumulation of microplastic pollution in these regions requires immediate investigation. To our knowledge, no studies have assessed the abundance of microplastic pollution in sediments from multiple Antarctic regions and depths, including the deep-sea. In this study, we conducted the most comprehensive study of microplastic pollution from deep-sea habitats of the Antarctic and Southern Ocean to date. Here, we sampled deep-sea marine sediments from 30 individual sites within three Southern Ocean regions; the Antarctic Peninsula, South Georgia, and the South Sandwich Islands. The samples were collected at a number of depths ranging from 136m to 3633m. We aimed to determine whether microplastic accumulation would vary among sites and individual Antarctic and Southern Ocean regions. Additionally, we aimed to highlight whether microplastic accumulation was related to the core sampling depth or the sediment characteristics among cores, thus providing insights on the processes that may be responsible for their presence in these remote regions.

2. Methodology

2.1 Sample Collection

All sediment samples were collected using OKTOPUS multicores (MUC) on the following research expeditions between 2017 and 2019: JR17003a, (RRS *James Clark Ross*; Antarctic Peninsula), PS119 (RV *Polarstern*; South Sandwich Islands and South Georgia), and M134 (RV *Meteor*; South Georgia). One individual core from each of the 30 MUC sampling sites was utilised for the microplastic analysis. The number of cores analysed per region were as follows: Antarctic Peninsula (6), South Sandwich Islands (11), and South Georgia (13) (Fig. 1; Table 1). Only the 0-2 cm depth from each core was used in the analysis. The sediment was then mixed evenly prior to analysis as the accumulation of microplastics over time was not considered for this study. The 30 cores were placed into a pre-washed zip-lock bag and frozen at -20°C prior to analysis. As different core tubing diameters were used on each of the research expeditions (JR17003a; ø 10 cm, PS119; ø 6.7 cm, M134; ø 10 cm/ ø 6 cm), the weight of all cores and microplastic counts were subsequently scaled up and standardised to represent the largest core diameter utilised during the expeditions (ø 10 cm).

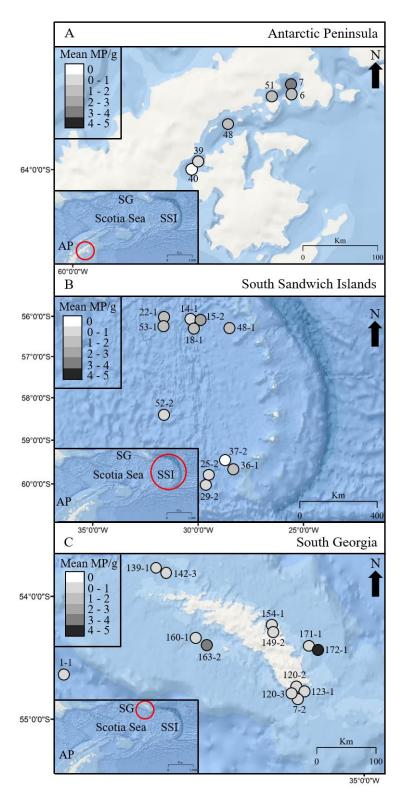


Figure 1: The overall mean microplastics per gram of sediment (MP/g) for each sampled sediment core within the Antarctic Peninsula, the South Sandwich Islands and South Georgia. All sampled cores are labelled with the MUC ID. Exact latitude, longitude, and depth values can be seen in Table 1.

Core	MUC ID	Lat	Long	Depth (m)
AP1	51	-63.6155	-57.4991	499
AP2	48	-63.7613	-57.9674	981.34
AP3	40	-63.9766	-58.4295	1246.33
AP4	39	-63.9765	-58.4294	1246.06
AP5	7	-63.5689	-57.2992	1031.61
AP6	6	-63.5756	-57.2986	1039.98
SSI1	14-1	-56.1284	-30.0696	2768
SSI2	15-2	-56.1099	-30.1521	3040
SSI3	18-1	-56.1489	-29.9758	3254
SSI4	22-1	-56.1390	-31.4783	3342
SSI5	25-2	-59.8790	-29.4710	2901
SSI6	29-2	-60.0418	-29.6972	2637
SSI7	36-1	-59.6953	-28.3275	1619
SSI8	37-2	-59.4863	-28.7772	2642
SSI9	48-1	-56.3534	-28.4714	2900
SSI10	52-2	-58.4714	-31.4767	3277
SSI11	53-1	-56.1284	-31.4740	3321
SG1	120-2	-54.8145	-36.01	201
SG2	120-3	-54.8146	-36.0101	201
SG3	123-1	-54.8545	-35.9111	318
SG4	139-1	-53.7702	-38.1402	367
SG5	142-3	-53.8142	-37.9943	211
SG6	149-2	-54.3518	-36.374	136
SG7	154-1	-54.2878	-36.3785	142
SG8	160-1	-54.3859	-37.5128	358
SG9	163-2	-54.4362	-37.3516	256
SG10	171-1	-54.4576	-35.8445	226
SG11	172-1	-54.4616	-35.8524	223
SG12	1-1	-54.7138	-39.5439	3633
SG13	7-2	-54.9025	-35.9443	320

132 2.2 Sample preparation133 Each of the 30 sediment

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Each of the 30 sediment cores were transferred to a pre-cleaned metal tray, labelled, placed in clean paper bags, and dried in an incubator at 40°C until all moisture was removed. The cores 134 were then gently ground using a pre-cleaned pestle and mortar and subsequently sieved 135 through a 2 mm sieve. Any particles larger than 2 mm were removed from the analysis by 136 sieving. Once sieved, the dry weight of the core was recorded and each core was then 137 separated into eight representative subsamples using a Quantachrome Rotary Micro Riffler. 138 The subsamples were then transferred into plastic 30 ml universal containers and labelled. All 139 universal containers were washed twice with deionized water and dried at 40°C before use. 140 141 2.3 Digestion Three subsamples from each of the 30 cores (n = 90), and 9 procedural blanks (i.e. purified 142 and pre-sieved sand of equal weight to the subsamples; Martin et al. 2017) were added to 250 143 ml pyrex glass beakers and covered in aluminium foil to avoid any airborne contamination. 144 The subsamples and procedural blanks were then digested in 50 ml of 30% hydrogen 145 peroxide at room temperature and left to stand overnight to remove any organic material. 146 Digestion with 30% hydrogen peroxide has already been applied to successfully remove 147 organic material from animal tissues and sediment samples (Mathalon & Hill 2014, 148 149 Kolandhasamy et al. 2018) while at the same time avoiding the dissolution of the microplastics themselves (Li et al. 2016). The following day, the beakers were heated on a 150 151 hotplate at 60°C until the reaction was completed and subsequently left to cool overnight. To 152 remove any calcium carbonate from the sediment, 2 mol hydrochloric acid (HCL) was added to each beaker and left to digest overnight at room temperature. Following this, the beakers 153 154 were then heated again on the hotplate at 60°C to ensure the reaction was completed. The subsamples and procedural blanks were then left to cool until the sediment had completely 155 156 settled on the bottom of the beaker. The liquid was then removed using a 10 ml pipette and 157 filtered deionized water was added to wash the sediment. 158 2.4 Density separation Each subsample and procedural blank was transferred into a pre-washed 50 ml falcon tube 159 and labelled. They were then topped up to 45 ml with filtered deionized water and 160 centrifuged (HERMLE Z 446) at 3000 rpm for three minutes for further washing. The water 161 was then replaced and the sample agitated using a vortex mixer for 30 seconds before 162

repeating the washing process. Once washed, the water was removed using a 50 ml pipette

and 30 ml of sodium polytungstate (SPT; 1.6 g/cm³) was added before further agitation 164 (Zhang et al. 2018). The subsamples and procedural blanks were then centrifuged at 3000 165 rpm for 20 minutes to float any potential microplastics from the sediment. Finally, the 166 supernatant was decanted and subsequently vacuum filtered using a three piece Hartley 167 pattern filter funnel and 25 mm VWR glass microfiber filter paper. Each filter paper was 168 immediately covered and dried at 40°C and the remaining SPT solution was removed and 169 170 filtered for recycling. 171 2.5 Visual identification and microscopy An initial visual analysis of potential microplastic pollution was carried out using a 172 173 stereomicroscope. Microplastics were identified following the visual identification protocol from Nor & Obbard (2014), i.e. particles that are bright/unnatural and homogenously 174 175 coloured, particles with no visible cellular or organic structures, and fibres that are equally thick and do not taper at the ends. All microplastic pollution was categorised into three 176 categories; fibres, fragments, and films (Ehlers et al. 2019). 177 2.6 Polymer identification 178 Once identified visually, the potential microplastics were measured (Sup Table 2) and 179 180 photographed using a digital microscope (VHX-2000, Keyence, Osaka, Japan) before being transferred to aluminium oxide membrane filters (Whatman Anodisc filter; pore size 0.2 µm; 181 diameter 47 mm) for subsequent spectroscopical analysis. This filter material is infrared 182 inactive in the wavenumber range in which characteristic plastic polymer peaks can be found; 183 184 therefore, it is recommended for transmission measurements in µFTIR spectroscopy (Löder et al. 2015). For the measurements, a subsample of the identified particles (20%; 29/147), 185 186 well representing the range of MP found in the samples, were analysed manually using a Fourier-transform infrared microscope (µFTIR, Hyperion 2000, Bruker, Ettlingen, Germany; 187 Ehlers et al. 2019). The µFTIR was equipped with a mercury-cadmium telluride detector and 188 the measurements were performed in transmission mode with the blank filter material used 189 for background measurements. For some thicker larger particles for which the transmission 190 mode was not suitable the attenuated total reflectance (µATR) mode with a germanium 191 crystal was used as suggested by several authors (Bergmann et al. 2017; Löder et al. 2015; 192 Vianello et al. 2013; Zhang et al. 2019). The measurements were performed in a wavenumber 193 range of 4000 to 600 cm⁻¹ with 32 co-added scans and a spectral resolution of 4 cm⁻¹. Finally, 194 195 each obtained spectrum was compared with the Bruker spectral library using the software

196 OPUS 7.5 to determine polymer types. For transmission measurements, only the part of the particle's spectrum between the wavenumbers 3800 cm⁻¹ and 1250 cm⁻¹ was analysed as 197 aluminium oxide membrane filters are infrared inactive in that region (Löder et al. 2015). 198 2.7 Contamination protocol and quality control 199 During the sediment core collection (2.1), all cores were stored in pre-washed zip-lock bags, 200 201 however, no pieces of the zip-lock bag from the sample collection were found in the samples. Prewashed glassware was also utilised throughout the sample preparation and aluminium foil 202 203 was used to cover samples at all times. In the case that plastic containers were used, they 204 were prewashed twice using filtered deionised water and inspected prior to use for 205 contamination under the stereomicroscope. All work benches and laboratory equipment were washed using deionised water and inspected for airborne contamination before and between 206 207 each stage of the analysis. Further to this, 100% cotton laboratory coats and nitrile gloves were worn at all times. Additionally, natural fibre clothing was worn under laboratory coats 208 throughout the analysis. Alongside the sediment subsamples, procedural blanks containing 209 210 purified sand of equal weight to the subsamples were used to quantify any contamination throughout the digestion and filtration stages. Additionally, damp filter paper placed in petri 211 dishes was left exposed to airborne contamination throughout the analysis to control for 212 further contamination from the laboratory. Both procedural and laboratory blanks were 213 quantified for microplastic pollution and accounted for during the analysis (Martin et al. 214 2017). 215 216 2.8 Sediment grain size A few drops of calgon was added to a 1 cm³ sediment subsample from each sediment core (n 217 218 = 30) and left overnight to form a paste. The sediment grain size was then calculated for each core using a Laser Diffraction Particle Size Analyser (LS13 320). A post hoc analysis was 219 220 then carried out using the Excel Macro GRADISTAT (Blott & Pye, 2001) to calculate the mean grain size and the percentage makeup of clay, silt, and sand for each sediment core. 221 2.9 Statistical analysis 222 All data was assessed for normality of residual distributions (Shapiro-Wilk test, P > 0.05) and 223 homoscedasticity of variances (Fligner-Killeen, P > 0.05). The mean microplastic per gram 224 225 data (MP/g) followed a non-normal distribution and exhibited heteroscedasticity; therefore, a Welch's ANOVA was employed to determine the difference among the mean MP/g per 226

227	region. The mean sediment grain size showed a normal distribution (Shapiro-Wilk test, $P >$
228	0.05) and therefore a one way ANOVA test was used to calculate the variance in means.
229	Additionally, the mean sediment grain size and the percentage sand/silt/clay of each sample
230	was correlated with site depth, counts of microplastic type (fibres/fragments), and
231	microplastic counts per site using a Spearman's correlation. All statistical analyses were
232	carried out using the software program R v3.4.4 (R Core Development Team 2018).
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234	3. Results
235	A total of 147 microplastic particles were identified and at least one microplastic particle was
236	found in 93% of the 30 sediment cores from the three Antarctic regions (28/30). Only one
237	core from the Antarctic Peninsula (MUC: 40) and one from the South Sandwich Islands
238	(MUC: 37-2) showed no microplastic pollution overall (Fig.1). The mean (±SE)
239	microplastics per gram of sediment for each region was 1.30 ± 0.51 MP/g, 1.09 ± 0.22 MP/g,
240	and 1.04 \pm 0.39 MP/g, for the Antarctic Peninsula, South Sandwich Islands, and South
241	Georgia, respectively. The Welchs ANOVA test showed no significant difference among the
242	mean MP/g values for each of the three regions (F = 2.21, df = 2, $P > 0.05$) (Fig.1/ Fig.2a).
243	Fragments were the most common particle found and contributed to $56\% \ (82/147)$ of the total
244	microplastics overall. Fibres and films made up the remainder of the particles found
245	representing 39% (57/147), and 5% (8/147), respectively. A total of seven different polymer
246	types were identified from the $\mu FTIR$ analysis; Polyesters (PEst, such as Alkyd),
247	Polypropylene (PP), Polystyrene (PS), Polyurethane (PU), Polyvinyl chloride (PVC) Rubber
248	(TPE), and Acrylic polymers (AP; Fig.2c). The majority of the microplastics found within
249	this study were polyester which were identified as blue fragments (Fig.3g; Sup Table 2), and
250	a range of coloured fibres. These blue polyester fragments were found in 35% of the total
251	sediment subsamples (32/90); and collectively, polyester accounted for 59% (17/29) of the

identified microplastics subsample within the three regions (Fig.2c).

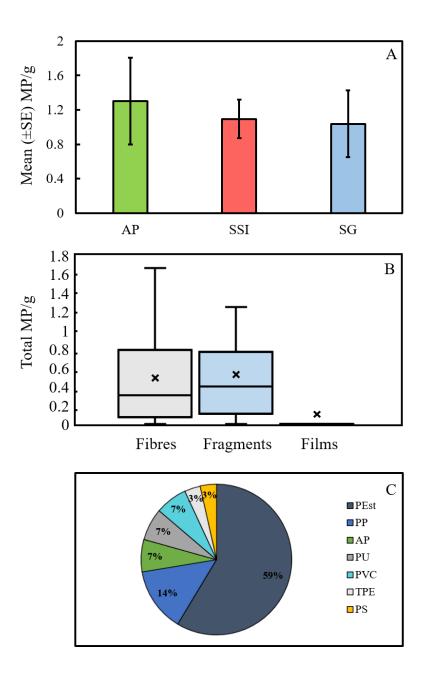


Figure 2: A) The mean (± SE) microplastics per gram of sediment from the Antarctic Peninsula (AP), South Sandwich Islands (SSI), and South Georgia (SG). B) The total number of microplastics types (fragments, fibres, and films) per gram of sediment, including the mean value (×). C) The percentage contribution of each of the seven polymer types found within the sediment cores. Polyester (PEst; including one Alkyd), Polypropylene (PP), Acrylic polymers (AP), Polyurethane (PU), Polyvinyl chloride (PVC), Rubber (TPE), and Polystyrene (PS).

263 Additionally, the mean (\pm SE) MP/g for the procedural blanks was 0.16 ± 0.08 MP/g, indicating a low level of contamination overall. Within the procedural blanks, a low amount 264 of natural fibres (n = 5) and polyester fibres (n = 3) were found; however, the polyester fibres 265 were visually different from the other polyester fibres within the sediment samples (i.e. 266 partially standing upright from the filter, or displaying different lengths and widths) and 267 therefore no adjustments were made. All natural fibres were excluded from the analysis and 268 no contamination was found in the atmospheric blanks, therefore no adjustments were made 269 270 to the results. The mean (\pm SE) sediment grain size for each of the three regions was $30.52 \pm 3.53 \,\mu m$ (AP), 271 $30.71 \pm 1.44 \,\mu m$ (SSI), and $24.82 \pm 1.61 \,\mu m$ (SG), and all cores contained mostly silt 272 sediment characteristics despite the range of depths. No significant difference was found 273 among the mean grain size at the three regions overall (ANOVA F = 2.67, P = 0.08). The 274 Spearman's correlation test showed a significant linear relationship between the mean 275 sediment grain size and the core sampling depth (S = 2317.8, P = 0.006). Additionally, core 276 sampling depth showed a strong negative correlation with the percentage of clay in each core 277 (S = 7759.1, P < 0.001), and a positive yet statistically non-significant relationship with the 278 percentage silt of each core (S = 2902.8, P = 0.054). In terms of microplastic type, higher 279 280 numbers of fragments were found in cores with higher percentages of clay (S = 2714.3, P =0.030); although, the accumulation of fibres did not correlate with higher percentages of 281

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sand/silt/clay.

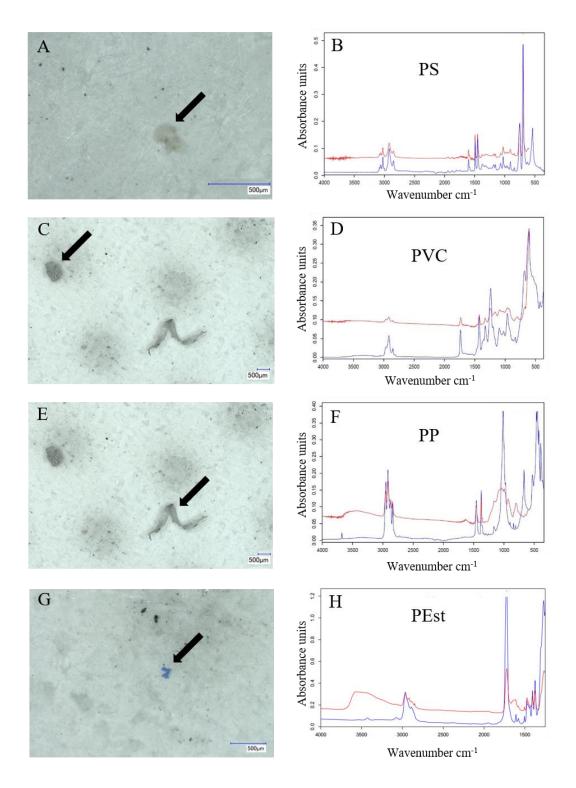


Figure 3: A subsample of the extracted microplastics including their corresponding μFTIR spectra recorded in μATR mode (B, D, F) and transmission mode (H). The red spectra represent the measured microplastic particles and the blue spectra are the reference spectra from the Bruker spectra library. All CO₂ peaks were removed from the spectra as artefacts. A-B) Clear polystyrene (PS) fragment. C-D) Grey rounded polyvinyl chloride (PVC) fragment. E-F) Grey polypropylene (PP) film. G-H) Blue polyester (PEst) fragment.

4. Discussion

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The results from our analysis demonstrate consistently high levels of microplastic pollution in 291 30 different cores from three regions within Antarctica and the Southern Ocean. This is, to 292 our knowledge, the most comprehensive study to date that highlights microplastic pollution 293 in deep-sea sediments from Antarctic regions south of the PFZ. Disturbingly, our study 294 reports very high benthic microplastic sediment loads (0 - 9.52 MP/g; Sup Table 1) that 295 resemble recent values found in Arctic deep-sea sediments (0.04 – 6.60 MP/g; Bergmann et 296 al. 2017 and 0.23 – 13.33 MP/g; Tekman et al. 2020). In addition, our samples resemble 297 similar values found in the intertidal sediments from Scapa Flow (0.1- 8.0 MP/g; 298 Blumenröder et al. 2017) and riverine sediments from Canada (0.06 – 7.56 MP/g; Crew et al. 299 2020) which one would expect to be more polluted than remote Antarctic regions. The values 300 found in our study were much higher than in another deep-sea study (0 - 1.04 MP/g; Zhang et)301 al. 2020) and many other sediment studies from shelf, intertidal, and riverine environments 302 303 (Table 2). The mean microplastic abundance in our study was also very similar to microplastic concentrations reported from shallow marine sediments (0.90 \pm 0.10 MP/g; 304 Alomar et al. 2016) and much higher than littoral sediments (0.46 ± 0.02 MP/g; Abidli et al. 305 2018) from more polluted marine habitats such as the Mediterranean Sea. Thus, Arctic and 306 307 Antarctic sediments can accumulate higher values of microplastic pollution despite their geographic distance from urbanized regions with higher levels of direct plastic pollution 308 309 input. 310 311 312 313 314 315 316 317

Table 2: Microplastic values per gram of sediment from a range of environments recorded in published sediment extraction studies. (See Supplementary Material for references)

MP/g	Environment	Region	Reference
0.04 - 6.60	Deep-sea	Arctic	Bergmann et al. 2017
0 - 0.20	Deep-sea	Arctic Central Basin	Kanhai et al. 2019
0.23 – 13.33	Deep-sea	Arctic	Tekman et al. 2020
0 - 1.04	Deep-sea	West Pacific	Zhang et al. 2020
0.04 - 0.197	Deep-sea	North Atlantic	Courtene-Jones et al. 2020
0.03 - 0.13	Deep-sea	Pacific	Peng et al. 2020
3.82	Slope	Mediterranean	Kane et al. 2020
0 - 0.07	Shelf	Arctic	Mu et al. 2019
0.04 - 0.34	Shelf	Bohai Sea	Zhao et al. 2018
0.08 - 0.28	Shelf	Northern Yellow Sea	Zhao et al. 2018
0.04 - 0.14	Shelf	Southern Yellow Sea	Zhao et al. 2018
0 - 0.26	Shelf	South Portugal	Frias et al. 2016
0.10 - 8.00	Intertidal	Scapa Flow	Blumenröder et al. 2017
0.01 - 0.06	Intertidal	Singapore	Nor & Obbard, 2014
0.14 - 0.46	Intertidal	Mediterranean	Abidli et al. 2018
0.01 - 0.52	River	Australia	He et al. 2020
0.06 - 7.56	River	Canada	Crew et al. 2020
0.03 - 0.56	River	Hong Kong	Wu et al. 2020

Once in the sediment, microplastics can be consumed by deep-sea benthic organisms, thereby entering the food web (Courtene-Jones et al. 2017). A recent study demonstrated that microplastics had been ingested by 83% of macrobenthic Antarctic invertebrates from a range of taxonomic groups, including bivalves, cnidarians, and amphipods (Sfriso et al. 2020). Furthermore, the abundance of microplastics in gentoo penguin (*Pygoscelis papua*) scat from South Georgia and the South Orkney Islands, found mean (\pm SE) levels of 0.23 \pm 0.53 microplastics per individual (Bessa et al. 2019), suggesting that microplastics may be travelling through trophic levels. Although the study was published in 2019, the scat was

represent the increase of microplastic accumulation over time. 332 There are a number of potential sources for the microplastics found within these regions. We 333 334 found unusually high levels of microplastic fragments within the sediment cores: the majority of which were polyester and blue in colour (64/82 total fragments) and represented 44% of 335 the total microplastics found overall. Notably, nearly identical blue polyester fragments were 336 also found in the scat of gentoo penguins from Bird Island, South Georgia (Bessa et al. 2019). 337 Further to this, blue polyester mainlines have been used on Patagonian toothfish 338 (Dissostichus eleginoides) fishery around South Georgia (Soeffker et al. 2015; SGSSI, 2018) 339 and Antarctic toothfish (Dissostichus mawsoni) fishery vessels operating within the Ross Sea 340 (Parker et al. 2019). Polymer ropes have shown to lose between 0.39 - 1.02 % of their mass 341 per month when degrading in seawater (Welden & Cowie, 2017), further emphasising that 342 fishing gear is another likely vector for the input of microplastic fibres or fragments in 343 344 Antarctic regions. However, we cannot attribute all of our polyester microplastics to the degradation of fishing ropes in these systems. 345 The number of fibres found within our sediment cores were lower than expected, as fibres are 346 known to be ubiquitous and therefore dominate microplastic extraction studies (Cesa et al. 347 2017). It is likely that the main source of polyester fibres within our samples derive from 348 synthetic clothing as synthetic fibres have been shown to release in high quantities during 349 washing machine cycles (Napper & Thompson, 2016). However, polyester fibres are also 350 transported atmospherically when released from synthetic clothing during everyday use, and 351 352 in quantities as high as washing machine cycles (De Falco et al. 2020). A previous study 353 suggested that with the exception of the Antarctic Peninsula, microplastic fibre release would be low in Antarctic regions due to the lack of human population (Waller et al. 2017), 354 355 although the Antarctic Peninsula showed the least number of fibres among our sediment cores. 356 357 Many marine paints are based on acrylic polymers, polyesters (like alkyds; Song et al. 2014) and polyurethane (Lacerda et al. 2019). Hence, the acrylic, polyester, alkyd and polyurethane 358 359 microplastic fragments that were found in the sampled Antarctic deep-sea sediments might derive from the varnish of ships, fishing vessels (Song et al. 2014) or marine stations. This is 360 supported by the appearance of the particles (such as the blue colour of polyester fragments 361 as well as their texture; see Fig. 3) which is similar to paint particles previously reported in 362

collected 10 years prior, and as such, our findings from the three Antarctic regions may

other studies (Song et al. 2014; Lacerda et al. 2019) and the high resemblance of our acrylic polymer µFTIR spectra with the ones of poly(acrylate/styrene) reported by Song et al. (2014). As the sediment cores did not come into contact with any ship surfaces or paint, we conclude that the microplastic fragments that we found in our samples did not derive from our ships as contamination. Recently, varnish particles were found in Arctic deep sea sediments (Tekman et al. 2020) and they were also found in Arctic sea ice cores (Peeken et al. 2018) and snow (Bergmann et al. 2019). Hence, they are prevalent even at remote locations. Although paint chips consist of high-density polymers, they can float in seawater (Song et al. 2014). Therefore, their distribution and sinking rate presumably differs from other high-density microplastics. Furthermore, paint chips may contain heavy metals and toxic antifouling substances (Song et al. 2014; Abreu et al. 2020) which can be ingested by marine organisms (Muller-Karanassos et al. 2019). Therefore, their presence in deep-sea sediments may threaten sensitive deep-sea organisms.

Most of the time it is very difficult to identify a specific plastic item as the source of a microplastic particle. Interestingly, we found two yellow polyurethane fragments (Fig. 4) in our study with a very distinctive sieve-like structure. They strongly resembled blue polyurethane fragments that were previously found in water samples along the North Western coast of Australia (Kroon et al. 2018). Their structure indicates that they may once have been part of finely woven nets or sieves.

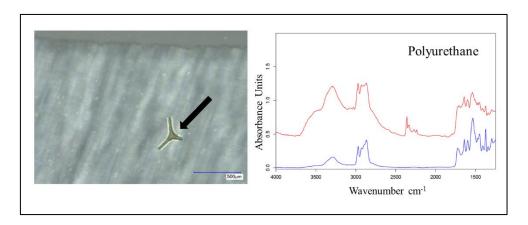


Figure 4: The sieve-like polyurethane particle with the corresponding μ FTIR spectrum (red: spectrum which was measured in transmission mode; blue: reference spectrum).

Microplastic accumulation has shown to be related to the proximity of human footfall in the past (Gewert et al. 2017). Given that high numbers of researchers are present on the Antarctic Peninsula at all times of the year (González-Alonso et al. 2017), and increasing numbers of

388 tourists are present during the summer months (Lynch et al. 2019), we would expect to see a clear increase in microplastic accumulation in this region, however, we did not find a 389 significant difference in the microplastic accumulation among the three regions. Indeed, the 390 Antarctic Peninsula had the highest average microplastic values (1.30 \pm 0.51 MP/g), and 391 392 although not statistically significant in comparison to the other regions, we expect that it is due to the increased footfall in this region (Waller et al. 2017). The most remote region, the 393 South Sandwich Islands, had the next highest mean microplastic abundance (1.09 \pm 0.22 394 MP/g), but only slightly higher than South Georgia (1.04 \pm 0.39 MP/g). Although researchers 395 396 (~ 10) are active on South Georgia all year round, and with tourists present in the summer months, the footfall is still relatively low (Gregory et al. 2017). Additionally, the South 397 Sandwich Islands have no permanent residents at any point of the year, so we suspect that 398 one of the drivers for microplastic accumulation in these regions is due to visiting research 399 and fishing vessels (Waller et al. 2017). Furthermore, it is also possible that microplastic 400 accumulation is the result of long range transportation in surface waters from neighbouring 401 402 regions or beyond (Lusher et al. 2014). Previous studies have shown no correlation between 403 human demographics and microplastic accumulation in the past, and suggest accumulation is 404 driven rather by atmospheric and oceanic currents (Kane et al. 2020; Nel et al. 2017). 405 In similarity to sediment particles, the density of microplastic polymers will alter how they are dispersed in marine environments, with low density particles maintaining positive 406 407 buoyancy on the surface for longer than high density particles (Kane & Clare, 2019). However, microplastic of both high and low densities are known to sink to the benthos 408 through a number of processes, such as biofouling (Van Cauwenberghe et al. 2013), attaching 409 to marine snow, faecal pellets and phytoplankton heteroaggregates (Tekman et al. 2020) and 410 transport to deeper waters and the sea floor through pelagic particle feeding (Choy et al 411 2019). Another recent study showed how microplastic particles positively correlate with the 412 abundance of chlorophyll a in Antarctic sea ice; and as such, highlights another pathway for 413 414 microplastic particles to enter the food chain and eventually sink (Kelly et al. 2020). These 415 studies demonstrate that microplastic pollution can be dispersed via biological means in marine systems and this link highlights the need to further investigate the biotic content of 416 417 sediment samples, which may help to explain the drivers behind sinking microplastic 418 particles. The mean sediment grain size for each core fell within the silt category $(3.9 - 62.5 \,\mu\text{m})$ 419 420 despite the range of sampling depths, however, high silt percentages of bottom sediment have

been found around the Antarctic Peninsula (Wu et al. 2019), South Georgia (Graham et al. 2017), and the South Sandwich Islands (Howe et al. 2004) in the past. The presence of finegrain sediments such as silt $(3.9 - 62.5 \mu m)$ and clay $(0.98 - 3.9 \mu m)$ are generally associated with deep-sea environments, where sediment grain size decreases with increasing depth from coarse sand at shallower to mud at deeper depths (Li et al. 2018). Clays and silts are considered the result of terrestrial erosion that can be transported long distances and are deposited in deep-sea environments through a range of means, such as aeolian transport, turbidity currents, bottom currents and resedimentation processes (Stow & Piper, 1984; Sweet & Blum, 2016). Clays will settle in low energy environments (i.e. low current velocities) such as the deep-sea, as they are continually transported from higher to lower energy environments (Ergin & Boder, 1998); however, low energy environments are also associated with shallower depths (Jackson et al. 2002) and exist at a range of depths within regions surrounding Antarctica (Harris & O'Brien, 1996; Pirrie, 1998). This would help explain why all of the sediment cores from this study were dominated by > 70% mud (silt and clay mix) despite the range of depths from 136 to 3633 m, and also explaining why sediment grain size increased significantly with depth among study samples (p = 0.006); although, the associated r² value of the correlation (0.118) showed low predictive reliability. As clay sediments are low in density and are transported through marine systems from high energy environments with higher current velocities to low energy environments (Ergin & Boder, 1998), it is likely that microplastic pollution follows a similar pattern. This study shows that microplastic fragment levels were higher in sediment samples that contained a higher percentage of clay (P = 0.03); and as a result, suggests that microplastic fragment distribution and fate in marine systems is similar to the distribution of clay particles throughout the water column. However, microplastic fibres did not correlate with an increase in any particular sediment characteristic, which suggests their presence is decoupled from sedimentary processes, perhaps reflecting the fact that they can be transported through the air over large distances (Gasperi et al. 2018). This also may be compounded by large scale disturbance in the upper Southern Ocean; their small size and irregular shape may hinder their settling rates through the water column (Kowalski et al., 2016; Martin et al. 2017). It is also likely that microplastic fibres will re-suspend in benthic environments more easily due to their shape and size (Bagaev et al. 2017). In light of these findings, a recently published study used predictive modelling to determine the fate of microplastic particles in marine systems, and found that bottom currents and particle density are the driving force behind high microplastic accumulation in the Mediterranean deep-sea (Kane et al. 2020). By calculating the critical

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455 shear stress of microplastic particles, one can determine, within reason, the settling rates of different polymer types and shapes based on density and size (Kane et al. 2020; Zhang et al. 456 2017). With the addition of further samples, the consideration of biotic content in marine 457 sediments, and the predictive modelling methods used by Kane et al. (2020), our study could 458 be developed further, and as such, provide a more comprehensive understanding of the fate of 459 microplastics in the Antarctic and Southern Ocean deep-sea. Additionally, the use of 210Pb 460 chronology may help to better describe the pollution history in our study regions and similar 461 environments from different marine systems (Chen et al. 2020; Courtene-Jones et al. 2020). 462 The Antarctic Circumpolar Current (ACC) and fronts in the Polar Frontal Zone (PFZ) are 463 464 physical barriers that enclose Antarctica and were initially thought to prevent species from travelling to and from the Southern Ocean (Brasier et al. 2017). We originally suspected our 465 466 results were deriving from within the Antarctic system due to its isolation by the PFZ and ACC, however, studies have shown that marine debris, species, and particles have crossed 467 468 these barriers in the past in certain regions (Convey et al. 2002; Galaska et al. 2017). There is also doubt that the ACC acts as a physical barrier for species or particles in the deep-sea 469 (Clarke, 2003; Brandt et al. 2007), which suggests that the microplastic particles found within 470 this study may originate from both north and south of the ACC. Although particles are known 471 to cross the ACC from the Southern Ocean, it is likely that the ACC helps to transport 472 microplastic pollution around Antarctica before subsequently being distributed by deeper 473 currents, such as the Antarctic bottom current which helps distribute sediments around 474 Antarctica (Heezen et al. 1966). Further, a recent study showed how bottom currents are 475 responsible for the transport of microplastics in the Mediterranean (Kane et al. 2020), and 476 another study described the transport of sediments via bottom currents in Antarctica 477 (Uenzelmann-Neben, 2006). Recently, surface waters sampled around the entire coastline of 478 479 Antarctica were shown to contain no microplastic pollution (Kuklinski et al. 2019); however, only three (PP, PS, TPE) of the seven polymer types found in this study were of a lower 480 density than seawater, and it is therefore likely that the majority of microplastic pollution 481 sinks rather than floats in surface waters around Antarctica where it is then transported by 482 deeper currents (Kane et al. 2020). Furthermore, microplastics were found in the sub-surface 483 waters of the Ross Sea (Cincinelli et al. 2017), and high levels were also recovered from 484 shallow sediment samples in the Ross Sea $(5 - 1705 \text{ MP/m}^2; \text{ Munari et al. 2017})$, and within 485 deep-sea sediment throughout our study. Although the reported units used by Munari et al. 486 487 (2017) are not comparable with our results, this further demonstrates that the benthos is the

488 endpoint for microplastic particles in Antarctica. As the microplastic counts in this study (0 – 9.52 MP/g or 0 – 9520 MP/Kg) were higher than other reported values in less isolated 489 systems outside Antarctica, and were similar to the high values found in the Arctic deep-sea 490 (Tekman et al. 2020), the results from this study suggest that the Antarctic and Southern 491 Ocean deep-sea accumulate much higher microplastic abundances than previously thought.. 492 493 494 495 Acknowledgments 496 EMC is supported by the Department for Agriculture, Environment, and Rural Affairs, 497 Northern Ireland. EMC gratefully thanks Dave Williams and Hazel Clarke for their technical 498 assistance, Prof Jochen Koop for facilitating the µFTIR analysis at the Federal Institute of 499 500 Hydrology, BfG, Koblenz, Germany, and Dr Jason Kirby for facilitating the microplastic analysis at Liverpool John Moores University. KL acknowledges support from the British 501 502 Antarctic Survey Polar Science for Planet Earth Programme funded by The Natural Environment Research Council NC-Science and NERC grant NE/R012296/1 for JR17003a. 503 The authors thank Prof Gerhard Bohrmann, Marum at University of Bremen, Germany for 504 the invitations to join the research cruises M134 (KL) and PS119 (KL, JDS). This work was 505 506 supported by the Hong Kong Branch of Southern Marine Science and Engineering Guangdong Laboratory (Guangzhou). The authors also thank the late Briar Dick for 507 stimulating discussion. 508 509 References 510 511 Abidli, S.; Antunes, J. C.; Ferreira, J. L.; Lahbib, Y.; Sobral, P.; Trigui El Menif, N. Microplastics in sediments from the littoral zone of the north Tunisian coast (Mediterranean Sea). Estuar. Coast. Shelf. S. 2018, 205: 1-512 9. 513 514 Abreu, F. E. L.; Lima da Silva, J. N.; Castro, Í. B.; Fillmann, G.. Are antifouling residues a matter of concern in 515 the largest South American port? J. Hazard. Mater. 2020, 398, 122937; DOI: 10.1016/j.jhazmat.2020.122937. 516 Alomar, C.; Estarellas, F.; Deudero, S. Microplastics in the Mediterranean sea: Deposition in coastal shallow 517 sediments, spatial variation and preferential grain size. Mar. Environ. Res. 2016, 115, 1-10.

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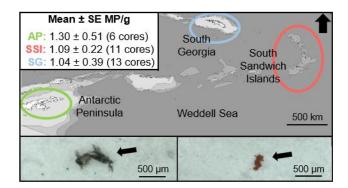
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715 For Table of Contents Only



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718 **Supporting Information**

- 719 The supplementary material provided contains:
- Supplementary Table 1
- Supplementary Table 2
- Table 2 reference list