



Welden, N. A. and Cowie, P. (2017) Degradation of common polymer ropes in a sublittoral marine environment. *Marine Pollution Bulletin*, 118(1-2), 248 - 253. (doi:[10.1016/j.marpolbul.2017.02.072](https://doi.org/10.1016/j.marpolbul.2017.02.072))

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Deposited on: 20 September 2018

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1 Degradation of Common Polymer Ropes in a Sublittoral Marine Environment

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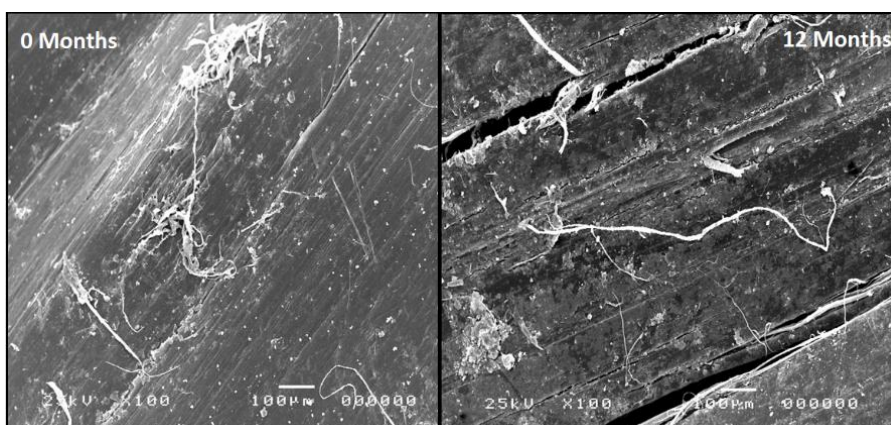
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7

8 ABSTRACT

9 Contamination by microplastic particles and fibres has been observed in sediment and animals
10 sampled from the Firth of Clyde, West Scotland. In addition to microplastics released during clothes
11 washing, a probable source is polymer ropes in abandoned, lost and discarded fishing and recreational
12 sailing gear. The fragmentation of polypropylene, polyethylene, and nylon exposed to benthic
13 conditions at 10 m depth over 12 months was monitored using changes in weight and tensile
14 properties. Water temperature and light levels were continuously monitored. The degree of biofouling
15 was measured using chlorophyll *a*, the weight of attached macroalgae, and colonizing fauna. Results
16 indicate microplastic fibres and particles may be formed in benthic environments despite reduced
17 photodegradation. Polypropylene, Nylon, and polyethylene lost an average of 0.39%, 1.02%, and
18 0.45% of their mass per month respectively. Microscope images of the rope surface revealed notable
19 surface roughening believed to be caused by abrasion by substrate and the action of fouling
20 organisms.

21



22

23

24 Capsule: By exposing polymer ropes to benthic conditions over a year we calculated that the initial
25 production of microplastic fibres would average 0.427 grams per meter per month.

26

27 Highlights:

- 28 • Polymer ropes lost between 0.39 and 1.02% of their mass per month
- 29 • Average production of microplastics was 0.427 grams per meter per month
- 30 • Of the polymers, polyethylene showed the largest reduction in mechanical properties
- 31 • Light intensity and temperature were linked to reduction in mechanical properties

32

33 Keywords: microplastic; microfibre; marine pollution; biofouling; tensile strength; fragmentation

34

35 INTRODUCTION

36 Plastic pollution, once regarded as a primarily aesthetic issue, is now recognised as physically
37 damaging to both marine organisms and habitats (Barnes et al., 2009; Browne et al., 2011). One of the
38 major concerns regarding plastic debris is its high durability, referred to as recalcitrance (Alexander,
39 1999). This durability is caused by strong bonds within the polymer and its high molecular weight,
40 which confer resistance to degradation (Palmisano and Pettigrew, 1992; Zheng et al., 2005). As a result
41 plastics persist in the environment for long periods, causing them to build up to high levels in both
42 terrestrial and marine habitats (Barnes et al., 2009). While the structure of polymers is highly durable,
43 plastics are susceptible to embrittlement, cracking, and reduction in mechanical properties (Massey,
44 2006). This weathering leads to fragmentation and the formation of secondary microplastics (Arthur,
45 2009). Secondary microplastic particles – less than 5 mm in size – have been seen to negatively impact
46 the marine environment as they are more difficult to remove from the environment and available for
47 uptake by a wider range of organisms. Microplastic ingestion has been observed in cetaceans (Lusher
48 et al., 2015a), wading and pelagic bird species (Connors and Smith, 1982; Ryan, 2008), benthic and
49 mid-water fish (Lusher et al., 2013; Lusher et al., 2015b), and benthic, pelagic and intertidal
50 crustaceans (Devriese et al., 2015; Murray and Cowie, 2011; Ugolini et al., 2013; Welden and Cowie,
51 2016).

52 Fragmentation of plastic debris can occur as a result of direct damage to the plastic product (Reddy et
53 al., 2006), or by weakening of the polymer structure (Alexander, 1999; Andrady, 2011). Physical

54 damage to plastic debris may occur via weathering action of waves and abrasion by either sediment
55 or other rough benthic substrates. This abrasive action may directly cause the formation of
56 microplastic, or enable settlement of colonising organisms by changing the surface texture of the
57 polymer (ter Halle, 2016). Physical degradation may also affect plastics in use in the marine
58 environment that may not otherwise be considered debris, for example the abrasion of trawl nets,
59 plastic pontoon components or ropes.

60 Weakening of a polymer is the result of breaking bonds in the molecule's backbone (Massey, 2006).
61 This may occur via a number of reactions, for example hydrolysis (Andrady, 2011; Kinmonth 1964).
62 For hydrolysis to occur a polymer must contain labile functional groups, for example esters which form
63 ionized acids. The first stage of the degradation process involves non-enzymatic, random hydrolytic
64 ester cleavage and its duration is determined by the initial molecular weight of the polymer as well as
65 its chemical structure (Pitt et al., 1981). Hydrolysis is especially important in the design of degradable
66 polymers and many have increased numbers of hydrolysable bonds and a more hydrophilic structure,
67 which increases the rate of polymer breakdown (Göpferich, 1996). Polymer backbones may also be
68 shortened by the production of highly reactive free radicals, units containing unpaired electrons or an
69 open electron shell. This breakdown is a three stage process. The first stage, initiation, is characterised
70 by the scission of the polymer chain either at the chain-end or randomly throughout its length,
71 resulting in the formation of two free radicals. Free radicals are units containing unpaired electrons or
72 an open electron shell; this makes the resulting molecules highly reactive. During the second reaction
73 phase, propagation, radical groups act upon the hydrocarbon chain to cause further breakdown of the
74 polymer (Leonas and Gorden, 1993; Muasher and Sain, 2006). The initial scission of a polymer chain
75 may be caused by a number of factors, the most common of which are ultra violet light (UV) and
76 temperature (Albertsson and Karlsson, 1988; Singh and Sharma, 2008).

77 The rate of weathering depends on the availability of these factors; as a result polymer degradation is
78 highly dependent on location and environmental conditions. This can be seen in the different rates of
79 degradation observed at different latitudes, linked to the level of UV radiation (Statz and Doris, 1987).
80 This scission of the bond causes the formation of free radicals, which may affect adjacent polymers
81 causing a secondary breakdown in the polymer structure. The final step in the process is the
82 stabilisation of the remaining radicals, which either join to form a new product, or form two separate
83 stable species (Albertsson and Karlsson, 1988; Geuskens and David, 1979). This process reduces a
84 polymers molecular weight and increases its susceptibility to degradation by biota. Fouled plastic
85 materials are then susceptible to fragmentation by biodegradation, or biodeterioration. Colonising
86 organisms and their by-products may cause direct damage to plastics (Andrady, 2011), for example,

87 through the mechanical action of borers (Davidson, 2012). Fouled debris may also be subject to
88 increased solubility and hydrolysis (Göpferich, 1996; Singh and Sharma, 2008).

89 The Clyde Sea Area receives plastic inputs from the highly populated and industrialized catchment
90 area and numerous marine activities. This plastic may enter the environment by numerous routes
91 including local littering, transport via wind and currents, landfill run-off, overboard disposal, and the
92 accidental loss of fishing gear (Lattin et al., 2004). Estimates of the relative importance of plastic
93 sources suggest that approximately 80% of marine litter originates from terrestrial sources (Teuten et
94 al., 2009) Of the remaining 20% of plastic litter believed to originate at sea, industrial maritime
95 activities are one of the main sources (Brown and Macfadyen, 2007; Kaiser et al., 1996; Kiessling, 2003;
96 Macfadyen, 2009; Otley and Ingham, 2003). Modern rigging and gear is almost solely of polymer
97 construction, and gear can be accidentally lost even with carefully handling. Fragmentation of
98 abandoned, lost and discarded fishing gear (ALDFG) in the marine environment will result in the
99 release of large quantities of microplastic particles and fibres; in this study the degradation of
100 commonly used polymer ropes is monitored to establish a rate of microfiber formation in benthic
101 environments.

102

103 MATERIALS AND METHODS

104 Nylon, polypropylene and polyethylene ropes were chosen due to their common use in both
105 recreational and industrial maritime activities around the Firth of Clyde. Sisal rope was selected as a
106 natural comparison, historically used in maritime activities. Construction varied between the three
107 rope types; polypropylene was a four strand twisted film, whilst polyethylene and nylon were four
108 strand braided ropes. All samples were 10mm in diameter, to ensure that the same surface area was
109 available. The initial mechanical properties of the samples were determined by tensile testing, carried
110 out using an Zwick-Roell Z250 tensile testing machine with a capacity of 100kN (Breslin and Li, 1993;
111 McKeen, 2008). Tensile strength was calculated by determining the force per unit area required to
112 fracture the sample. Elongation was recorded as both the total increase in length and the ratio
113 between the change in length and the original length. Sample ropes were cut to 50mm lengths and
114 randomly assigned spaces on acrylonitrile butadiene styrene (ABS) frames. Frames were then fixed on
115 the seabed from an existing pier, at 10 meters depth on silty sediment. The deployment site was
116 situated in a tidal eddy off an existing pier, however tidal currents in the Clyde Sea are predominantly
117 weak. Water movement is modulated by wind-driven surface currents, and the site was chose due to
118 its south easterly aspect, facing into the prevailing winds (Wilding et al., 2005).

119 **Abiotic Causes of Degradation**

120 Over the sample period water temperature and light levels were measured using HOBO UA-002-64
121 loggers. Frames were lifted every two months and three samples of each rope type removed. Where
122 rope samples were to be weighed, biofouling organisms were removed using forceps and a scalpel
123 and gentle rinsing with a wash-bottle. Where removal of attached biota would result in damage to the
124 rope surface care was employed to cut away as much of the attached biomass as possible. Once dried,
125 samples were weighed and the new buoyancy determined. Ropes to be subjected to tensile testing
126 were rinsed with deionised water, air dried, and stored in foil at 20°C until analysis. The tensile
127 strength and elongation at break of each was determined.

128 **Biotic Causes of Degradation**

129 Samples were examined for evidence of colonisation by biofouling organisms. Levels of chlorophyll α ,
130 the weight of attached algae, and the number and diversity of invertebrates and attached diatoms
131 were examined for each sample. Chlorophyll α was determined by refrigerating 10mm lengths of rope
132 with 10ml of 90% acetone for 24 hours, after extraction each sample was centrifuged for 10 minutes,
133 and the resulting supernatant transferred to a 1 ml cuvette. The chlorophyll concentration was
134 determined by using a Thermospectronic HeliosY spectrophotometer, with readings taken at 750, 664,
135 647, and 630 μm , using a 90% acetone blank.

136 The abundance and type of fouling organisms were determined by enumerating the number of
137 colonising animals and the weight of attached biomass. Biofouling by macro-organisms was
138 determined examining 50mm sections of rope under a binocular microscope. Algae and organisms
139 were removed for identification. Attached macro-algae were removed and oven dried at 40°C
140 overnight to determine the dry weight.

141 **Statistical Analysis**

142 Monthly changes in the weight of the sample, its tensile strength and elongation at break were
143 compared using Kruskal-Wallis analysis, carried out in Minitab 15. The measured factors responsible
144 for variation in tensile strength and elongation at break were subjected to GLM analysis in R (version
145 3.0.2). Prior examination of the data revealed that a number of variables were found to inter-
146 correlate; these variables were included in sequential models to determine which had the greatest
147 impact on model fit. After running a GLM using all environmental variables, the model was reduced
148 using in a stepwise process in order to improve of the resulting model, this was determined by the
149 relative size of the Akaike information criterion (AIC).

150

151 RESULTS

152 **Abiotic Causes of Degradation**

153 Over the exposure period the average sea temperature was 9.73 °C. The recorded temperature range
154 was between 5.7 °C, recorded in March, and 17.9 °C, recorded in July. Average light intensity over the
155 exposure period was 122 Lux, reaching a maximum of 17222 Lux in June. The highest monthly light
156 intensity took place throughout June and July.

157 **Biotic Causes of Degradation**

158 Sisal samples recovered two, four, and six months into the sample period had low chlorophyll *a*
159 readings and less than 0.01g dry weight of macroalgae – which was first observed after four months.
160 Higher levels of chlorophyll *a* were observed on polymer ropes (Table 1). All rope samples also
161 exhibited macroalgal growth, which greatly increased over the course of the year. The two most
162 commonly identified macroalgal species were *Alaria esculenta* and *Palmaria palmata*. *P. palmata* was
163 observed on all rope types, occurring after eight months on polymer ropes. *A. esculenta* was only
164 observed on polypropylene and polyethylene ropes, first recorded on polyethylene at six months,
165 followed by polypropylene at eight months. The high algal dry weights observed on these two
166 polymers were the result of large *A. esculenta* fronds.

167

168 Table 1. Colonisation of samples by macroalgae over the experimental period

	Max Chlorophyll <i>a</i> mg cm ⁻²	Max Dry Algal Biomass (g per 50 cm)	First macroalgae observed	Macroalgal Species observed
Nylon	1.98	2.1	Eight months	<i>P. palmata</i>
Polypropylene	1.84	10.7	Four months	<i>P. palmata</i> , <i>A. esculenta</i>
Polyethylene	0.76	9.3	Six months	<i>P. palmata</i> , <i>A. esculenta</i>
Sisal	0.20	<0.01	Four months	<i>P. palmata</i>

169

170 Table 2. Colonisation of samples over the experimental period. Values indicate number of months
171 after which the species was observed

	Nylon	Polypropylene	Polyethylene	Sisal
Chorophium sp.	2	2	2	2
Stenula sp.	-	-	8	-
Bryozoan	10	8	10	-
Eliminus modestus	-	8	-	-
Mesotigmata	-	-	6	-
Littorina littorea	8	-	6	-
Polychaete	10	10	6	-
Chironomid	-	-	-	6
Mytilus edulis	8	8	6	-
Annelida	-	-	10	-
Pomatoceros triqueter	8	-	-	-
Ascidian	8	-	-	-

172

173

174 The number and type of invertebrate organisms observed varied over the course of the study period
 175 (Table 2). After two months all polymer ropes were colonised by *Corophium* sp., which formed tubes
 176 on the rope surface. Between six and eight months the number of grazers was observed to increase,
 177 with the appearance of the periwinkle, *Littorina littorea*. A late coloniser of polyethylene was *Stenula*,
 178 an amphipod commonly found in sublittoral algae, possibly attracted by increasing macroalgal cover.

179 Prolonged exposure to benthic conditions resulted in fouling by larger encrusting organisms. The
 180 barnacle, *Eliminus modestus*, was found on samples of polypropylene exposed for over eight months,
 181 and the blue mussel, *Mytilus edulis*, was found on all polymers between six and eight months
 182 exposure. After 12 months polypropylene had the most recorded species with five per sample, while
 183 Nylon had only four species recorded per sample.

184

185 **Mechanical Properties**

186 Analysis of the change in the mechanical properties of sisal revealed significant reductions in both
 187 elongation and breaking strain. Of the three polymer ropes, polyethylene showed the highest average
 188 change in elongation at break, followed by polypropylene and Nylon (Table 2). The mean change in
 189 tensile strength was highest in polypropylene, followed by Nylon and polyethylene. For all rope types,

190 the rate of degradation was greatest in the first months, slowing during the 12 month experimental
 191 period. Statistical analysis of the factors related to both elongation at break and tensile strength
 192 exhibited different responses, however, a number of factors were common between the three
 193 polymers. The most commonly observed significant factors were maximum temperature, average
 194 temperature, average light intensity and the weight of macro algae.

195

196 **Fragmentation**

197 All ropes demonstrated a reduction in mass over the twelve month exposure period (Table 2);
 198 however, the scale of this loss was highly variable. Statistical analysis of the monthly reduction in
 199 sample weight indicated significant differences in the rate of microplastic formation by each rope type
 200 ($H = 18.23$, $df = 3$, $P < 0.001$). The natural sisal rope had degraded completely by month eight but even
 201 the fastest degrading polymer had not yet lost 13% of their mass.

202 The polymer ropes displayed differing levels of wear when examined under SEM. Fibrous Nylon line
 203 showed indications of there was obvious increasing fraying over the twelve month period; fibres did
 204 not lay as flat to the rope surface, and there were notable breakages of individual strands. Extruded
 205 polyethylene filament rope revealed increased surface scratching and roughening, and polypropylene
 206 twisted film rope developed many visible cracks and fissures, as well as the formation of fine surface
 207 fibres - particularly apparent in areas of animal attachment.

208 GLM analysis was used to compare the average mass lost per month with the observed changes in the
 209 tensile properties of the sample and the colonising organisms. The fragmentation of both Nylon and
 210 polyethylene was linked to elongation at break. The fragmentation of polypropylene rope samples
 211 was not significantly related to any of the measured variables, although a number of weak
 212 relationships were apparent.

213

214 Table 3. Variation in Mechanical Properties over the Experimental Period

	Average Mass lost per Month	Percentage Mass lost per Month	Percentage Mass lost	Max reduction in Elongation (mm)	Max reduction in Tensile Strength (N)
Nylon	0.422 g	1.02%	12.24%	- 15.265	- 339.60
Polypropylene	0.086 g	0.39%	4.68%	- 11.850	- 544.06

Polyethylene	0.132 g	0.45%	5.4%	- 43.620	- 562.53
Sisal	0.657 g	12.50%	100%	- 9.3866	- 1125.34

215

216 DISCUSSION

217 The results of this study indicate that large masses of microplastic can be formed quickly, even in the
 218 low energy benthic habitat in the Firth of Clyde. When the numerous sources of rope debris are
 219 considered, this indicates that an alarming volume of microplastic is formed annually by ropes alone.
 220 The reduction in mechanical properties and the fragmentation rates of the polymer ropes varied over
 221 the 12 month experimental period. Nylon rope was observed to fragment the fastest, whilst
 222 polypropylene rope fragmented the slowest. The rate of fragmentation may have been partially
 223 influence by differences between the ropes constructions; however, whilst there was an apparent
 224 difference in the monthly rate of mass lost, a number of common factors were identified as
 225 significantly affecting degradation and fragmentation.

226 **Abiotic Causes of Degradation**

227 Of the environmental factors included in the GLM analysis, only maximum and average temperature
 228 and average light were identifies as significant in analyses. Light is often identified as a driving factor
 229 of polymer degradation. Samples of LDPE exposed to UV irradiation showed three stages of
 230 degradation. These are believed to represent a rapid change within the material followed by a reduced
 231 rate of degradation, and finally a collapse of the polymer structure (Albertsson and Karlsson, 1988). In
 232 a study of the degradation of polymer films in the marine environment, reduced UV transmission was
 233 found to result in lower rates of degradation over time (O’Brine and Thompson, 2010). Because of the
 234 low penetration of light, most photochemical reactions take place on the surface of the plastic; even
 235 so, photodegradation is considered to be the primary cause of plastic break down (Singh and Sharma,
 236 2008).

237 Previous observations of the degradation rates of polyethylene films has demonstrated that exposure
 238 to increased temperature significantly reduces a plastic’s tensile properties (Whitney et al., 1993).
 239 Temperature influences the degradation of plastics by exciting electrons within the polymer structure
 240 causing bond scission and the shortening of the polymer chain (Singh and Sharma, 2008). Increased
 241 temperature also affects other forms of degradation. If the temperature is increased this reduces the
 242 energy required to break vulnerable bonds in the polymer backbone (Singh and Sharma, 2008).

243 One abiotic factor that could not be controlled within this study was the impact of abrasion by
244 sediment. The exposure trial was carried out using fixed ropes in a comparatively low energy
245 environment. Abrasion against rocks, encrusting organisms and sediment would increase the rate of
246 fragmentation and, therefore, microplastic formation.

247 **Biotic Causes of Degradation**

248 Weak relationships were observed between the number of fouling organisms and the reduction in
249 mechanical properties/sample mass. This may be the result of the constitutive enzymes of sessile
250 invertebrate colonisers on the surface of the polymer. These chemicals act on the bonds in the
251 polymer to weaken them (Flemming, 1998; Göpferich, 1996; Gu and Gu, 2005). Damage to the bonds
252 in the polymer backbone enables biodegradation by other organisms (Bonhomme et al., 2003), as well
253 as an increasing in the surface solubility of the polymer, enabling attachment by other organisms.

254 The action of grazing colonisers may also influence rope fragmentation. Numerous grazing crustaceans
255 have been seen to consume plastic. Mesocosm experiments have shown that *Gammarus fossarum*
256 ingest both microspheres and fibres (Blarer and Burkhardt-Holm, 2016). More notably, wild caught
257 gammarids from the Dutch marine environment have shown microplastic uptake at approximately 11
258 per g (dry weight)(Leslie et al., 2013). One of the most common invertebrate colonisers of all rope
259 types were *Corophium*. These grazing invertebrates may rasp the rope surface whilst feeding on algae
260 and, in the process, may take in microplastic particles and fibres, establishing a primary route for
261 microplastics entering the food chain.

262 The colonisers observed here exist within a defined area of intertidal and shallow subtidal waters, to
263 which they are adapted; outside this niche, the impact of increased polymer biodegradation by fouling
264 organisms may be highly variable. Changing abiotic variables will also influence biotic degradation.
265 One example of such an impact would be that of increasing depth. With increased depth there is a
266 reduction in light penetration which will limit photosynthesis, and therefore, algal biomass as well as
267 the number of grazers.

268 **Variation in Degradation**

269 Over the experimental period the average monthly air temperature was between 4.9 – 11.4 °C, and
270 1271.3 sunshine hours were recorded. These conditions are consistent with local monthly averages
271 recorded in the Firth of Clyde between 1981 and 2010 (between 2.6 – 19.6 °C, and 1320.0 hours of
272 sunshine). The average water temperature at the experimental site was found to be 9.81°C, and

273 ranged from 5.6 to 17.5 °C; this range is similar to that previously reported by Slesser and Turrell
274 (2005). This suggests that levels of plastic degradation in the CSA will be similar between years.

275 Globally, the factors linked to plastic degradation in the marine environment are subject to high
276 variability. The rate of microplastic formation from ropes, nets etc. may be expected to vary over the
277 course of the year due to the changing influence of light on both degradation and fragmentation rates.
278 This study indicates that there will be increased microplastic formation during summer months, when
279 light levels are at their highest. This proposed seasonality in microplastic generation does not account
280 for the influence of winter storm events, during which there would be increased mechanical abrasion.
281 Similarly, different latitudes receive light at different intensities over the course of the year. Areas at
282 lower latitudes may be expected to experience higher rates of degradation due to their
283 comparatively high light levels.

284 The action of organisms on the plastic may increase the rate of degradation; however, encrusting
285 animals may reduce the amount of light reaching the polymer - either by covering the surface and
286 preventing light penetrating the polymer or by causing the plastic to sit lower in the water column. At
287 increasing depth plastic in would experience lower light levels and fewer extremes of temperature,
288 reducing the overall rate of degradation.

289 The dimensions of the plastic litter will also greatly influence degradation rate. The surface area to
290 volume ratio of rope samples is small when compared to that of films; as a result, much of the mass
291 of the sample would be protected from environmental conditions. Observations of the fragmentation
292 rates of polyethylene films were faster than those observed here (Whitney et al., 1993). The average
293 rate of degradation observed over the twelve month experiment may increase as the rope fragments
294 and the surface area to volume ratio increases the rate of degradation would be expected to increase
295 accordingly (Andrady, 2011).

296 **Impact on Microplastic Formation**

297 The mass of microplastic particles and fibres in the marine environment is a combination of that from
298 existing marine debris and those produced from other sources including clothes washing, seen to
299 produce around 1900 fibres in a single wash (Browne et al., 2011). A single washing of a polyester
300 garment has been seen to produce up to 2 g of microfibers, approximately 0.3% of the total garment
301 weight (Hartline et al., 2016). Understanding the rate of rope degradation sheds more light on the
302 pool of secondary microplastics. Previous analysis of non-degradable plastic films in the marine
303 environment revealed that only 2% of the material was lost over 40 weeks (O'Brine and Thompson,
304 2010). Over the time span, the polymer ropes in during this study demonstrated a reduction in mass

305 of 5.72%. Whilst the conditions influencing microplastic generation are highly variable, the ability to
306 categorise areas in which the abiotic conditions favour plastic degradation will enable the
307 identification of habitats at risk from locally formed secondary microplastics.

308 The conservative estimates of plastic fragmentation in a temperate environment which are presented
309 here indicate that there will be vast amounts of microplastic formed from the degradation of global
310 abandoned lost and discarded fishing gear. The FAO estimate that 640,000 tonnes of fishing gear are
311 lost to the marine environment every year. Assuming an equal mix of the three polymers observed in
312 this study the degradation rate of this plastic would be 0.62% per month. Using this conservative
313 estimate, the monthly mass of microplastic generated would be 3,968 tonnes, totalling 47,616 tonnes
314 in the first year. This calculation only refers to annual gear losses, and not to the mass of plastic already
315 in the environment. In addition to ALDFG lost at sea, trawl nets and other ropes used in both
316 commercial and recreational boating will also release both thick strands and finer fibres to the
317 environment, greatly increasing this figure. In order to reduce ongoing microplastic production, there
318 must be an active effort to reduce the volume of parent material in the marine environment. Increased
319 incentives must be made to “fish of litter” and to recover and recycle plastic at port facilities. Costs
320 currently levied on the correct disposal of wastes from industrial maritime activities need to be
321 reassessed to make the reduction of marine litter a priority for maritime industry.

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