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## 6 7 **The role of wet wipes and sanitary towels as a source of white** 8 **microplastic fibres in the marine environment**

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18

### 19 **Abstract**

20 Understanding source elements of the ocean plastic crisis is key to effective pollution reduction  
21 management and policy. The ubiquity of microplastic (MP) fibres in the oceans is considered  
22 to derive primarily from clothing fibres released in grey water. Microplastic fibres degraded

23 from widely flushed personal care textile products (wet wipes and sanitary towels) have not  
24 been clearly identified in aquatic systems to date. Unregulated personal hygiene and sanitary  
25 product labelling fails to identify textile materials. This study demonstrated that white MP  
26 fibres in sediments adjacent to a wastewater treatment plant (WWTP) are comparable with  
27 white fibres from sewage-related waste and commercially available consumer sanitary  
28 products. Commercially available non-flushable wipes are manufactured from either  
29 polyethylene terephthalate (PET), polypropylene (PP), or a combination of PET and cellulose.  
30 Fifty percent of brands labelled flushable are comprised of a mixture of PET and cellulose and  
31 the remainder of cellulose alone. Sanitary towels are made from PP, PE, or a combination of  
32 high-density polyethylene (HDPE) and PP. The accumulation of large quantities of washed-up  
33 sewage-related macro-debris (including wet wipes and sanitary towels) intermingled with  
34 seaweed biomass adjacent to the WWTP and was associated with a combined sewer overflow.  
35 Microplastic fibres extracted from this waste were similar to those extracted from intertidal  
36 sediments in close proximity to the WWTP over a ten-month period. In comparison, fibres  
37 extracted from locations spatially removed from the WWTP were primarily comprised of ABS,  
38 PP and polystyrene. The results confirm that wet wipes and sanitary towels flushed down toilets  
39 are an underestimated source of white MP fibres in the environment. Given the global  
40 distribution and projected growth of the non-woven textile industry, there is a need for  
41 increased public awareness of MP pollution in the marine environment from the inappropriate  
42 disposal of sanitary products down the toilet, instead of diversion to alternative land-based  
43 waste management.

44

45 **Keywords:** microplastic, sewage, non-woven textiles, wipes, sanitary towels, wastewater.

46

## 47 **1. Introduction**

48 Microplastics (MPs) <5mm in size (maximum dimension) are a contaminant of increasing  
49 concern within aquatic systems, known to enter the food chain and act as a vector for potentially  
50 harmful contaminants (Galafassi et al., 2019; Thompson et al., 2004; Zhao et al., 2018).  
51 Primary sources of MPs in the marine environment include micro beads as a component of  
52 cosmetic products, and secondary sources include micro particles of larger plastic debris  
53 degraded through wave action and ultraviolet light (UV) and synthetic clothing fibres (Hidalgo-  
54 Ruz et al., 2012). Benthic sedimentary cores in both coastal and continental shelf margins show  
55 ubiquitous MP contamination (Martin et al., 2017); MPs that remain buoyant follow circulatory  
56 currents, converge in oceanic gyres and reach the most remote areas of the planet, including  
57 the northern and southern polar circles (Barnes et al., 2010; Bergmann & Klages, 2012).

58 Understanding the possible sources of MP entering near shore marine environments is key to  
59 developing effective pollution reduction policies. Domestic washing machines can release  
60 thousands of synthetic fibres per wash cycle (Napper & Thompson, 2016) and, although some  
61 of this material is retained in the sludge during the wastewater treatment process, a certain  
62 portion bypasses the treatment process (Carr et al., 2016). This has been estimated to be as  
63 much as 160,000,000 MP particles per day as a result of the release of grey water into coastal  
64 water bodies (Horton et al., 2017; Leslie et al., 2017; Magni et al., 2019; Murphy et al., 2016).  
65 This predominant reporting of multiple coloured particles supports the view that the principal  
66 source of marine MP pollution are clothing fibres and fragmented marine ropes (Almroth et  
67 al., 2018; Browne et al., 2011; Dris et al., 2015; GESAMP, 2015; Mahon et al., 2017; Reed et  
68 al., 2018; Wieczorek et al., 2018).

69 As plastic products are generally manufactured through molding, casting and extrusion, defects  
70 and residual stress in the polymer contributes to fragmentation (Ghorbani et al., 2013).

71 Microplastics released in wastewater treatment plants (WWTPs) are subject to sheer stress  
72 forces of turbulence, pumping and mechanical mixing which increases fragmentation of MPs  
73 (Enfrin et al., 2020a). Microplastics may also impact on WWTP performance through the  
74 fouling of filtration membranes (Enfrin et al., 2020b). In addition, MPs may act as toxicological  
75 vectors, adsorbing concentrations of contaminants commonly found in WWTPs such as  
76 pharmaceuticals (Beckingham and Ghosh, 2017; Seidensticker et al., 2017; Li et al., 2018).  
77 Through these mechanisms a wide range of MP particles are released from WWTPs, including  
78 nanoplastics which may be ingested via skin diffusion during embryogenesis of fish cells,  
79 resulting in mortality of marine organisms (Enfrin et al., 2020a).

80 Non-woven textiles form the base material of many sanitary products (including wet wipes and  
81 sanitary towels). A recent study identified the material composition of ‘baby/wet wipes’ as  
82 white micro-polyethylene terephthalate (PET) fibres used in the manufacture of products  
83 labelled as flushable and other components such as high-density polyethylene (HDPE) and  
84 polyethylene/vinyl acetate (PEVA/EVA) (Pantoja-Munoz et al., 2018). European production of  
85 non-woven textiles for hygiene products and personal care wipes reached over 1M tonnes in  
86 2016 alone (INDA/EDANA, 2018). These products form a significant component of global  
87 sewerage system blockages (Patchell, 2014), incurring significant technical and financial costs  
88 to wastewater utilities (Irish Water, 2018; Mitchell et al., 2017; Morrison, 2015; Thames Water  
89 2018). Twenty percent of the 8490 items of subsurface debris trapped over a three-month period  
90 in the River Thames were identified as macro components (intact or partially intact) of sanitary  
91 products, with contamination highest adjacent to a WWTP (Morritt et al., 2014). Many  
92 personal care products form an increasingly persistent feature of global coastal plastic pollution  
93 surveys (Coastwatch, 2016). In comparison with clothes fibres that are generally coloured or  
94 multi-coloured, fibres from sanitary products are white in colour. To date, the role of MP fibres

95 in the marine environment, emanating from these products as a significant component of  
96 WWTP effluent, appears to remain unconsidered and unconstrained.

97 In most MP studies to date, white fibres are likely underestimated, because of the commonly  
98 used filtration procedure to capture MP particles, as filters are commonly white, making visual  
99 identification of microscopic white fibres against a white background difficult (Blumenröder  
100 et al., 2017; Dekiff et al., 2014; Horton et al., 2017; Leslie et al., 2017; Murphy et al., 2016;  
101 Nor & Obbard, 2014; Pagter et al., 2018; Reed et al., 2018; Vianello et al., 2013). This is  
102 significant given the global growth of non-woven synthetic fibre products and their ubiquity in  
103 wastewater.

104 The primary objective of this study was to assess the role of sewage-related debris (flushable,  
105 non-flushable and sanitary towels) as a source of white MP fibres in the marine environment  
106 (urban and rural). The material composition of a variety of white fibres obtained from intertidal  
107 sediments and a recurring washed-up deposit of true-to-form sanitary waste, likely associated  
108 with a combined sewage overflow on a beach in close proximity to a WWTP, were  
109 characterised. The results were compared with a variety of consumer sanitary products in order  
110 to investigate their role as a source of white MP fibres in the marine environment.

111

## 112 **2. Methodology**

### 113 *2.1 Sample sites*

114 The study area covers three locations along the mid-western Irish seaboard, on Europe's north-  
115 western, Atlantic-facing margin (Fig. 1). Weak tidal currents predominate across the near-  
116 coastal continental shelf, along which the 'Irish Coastal Current' flows northward (Fernand et  
117 al., 2006). The strong oceanic influence induces a temperate maritime climate with average  
118 annual precipitation values in excess of 1000 mm, characterized by frequent moderate winter

119 storms. The region is marked by a strong overland and groundwater fluvial system and  
120 relatively high energy, sand to coarse sediment blanketed coastlines. Exceptions occur at the  
121 heads of the many coastal embayments, where finer grained sediment may accumulate in  
122 intertidal wedges dominated by sandy silt sized fractions.

123 Galway Bay, a 50 km long by 34 km wide west-facing embayment, is partially enclosed and  
124 protected from strong shelf currents and large oceanic swells by the Aran Islands topographic  
125 barrier. Oceanic flows enter the bay via the South Sound and circulate anti-clockwise before  
126 exiting the Bay through the North Sound (Fig. 1). A major source of freshwater input occurs at  
127 the northeast corner of the bay, where the River Corrib, catchment size 3136.08 km<sup>2</sup>, enters at  
128 the port of Galway City. The Corrib freshwater plume moves west along the east of the tidal  
129 gyre, except when strong south west winds may drive the plume to the shallower inner bay to  
130 the east (Nolan, 1997). The main WWTP for Galway City is located at Mutton Island (Figs. 1  
131 and Fig. S1).

132 Mutton Island, the primary sample site (Fig. S1a, b) (53°15'45.37 N, 09°03'17.74 W), is located  
133 just south of Galway City within the headwater reaches of Galway Bay. The sample site is  
134 located on an intertidal flat (tidal range 4.5 m) at the mainland end of a non-breached access  
135 causeway 0.8 km from Mutton Island WWTP, to the lee of prevailing winds and downstream  
136 of the Corrib river outflow (0.6 km). The area is dominated by extensive, relatively fine-grained  
137 sediment veneer (generally <1 m) overlying eroded undulating Pleistocene glacial  
138 sediments. This is a regionally important wastewater infrastructure with a population  
139 equivalent of 170,000 (local population = 80,000; annual overseas visitors in 2016 = 1.67  
140 million; one annual summer week-long festival alone accounts for an additional influx of  
141 145,000 visitors).

142 Two other estuaries were chosen as control sites for this work: Bell Harbour 16 km to the south-  
143 south-west and within Galway Bay, and Bellacragher, 100 km to the north. Bell Harbour, Co.  
144 Clare (Fig. S1c) (53 07' 20.51 N, 09 04' 23.08 W) is a sheltered tidal estuary (tidal range 4.5  
145 m) on the south coast of Galway Bay. Inland and surrounding the site, the karst limestone  
146 plateau of the Burren area has a sparse rural population. The local catchment feeding into the  
147 estuary covers 56 km<sup>2</sup>, with freshwater emerging into the centre of the estuary via a submarine  
148 outlet (McCormack et al., 2017). The sample site is located ~5 km from the wastewater outlet  
149 at Ballyvaughan and ~9 km from the outlet at Kinvara. Bellacragher (Fig. S1d) (53 57' 56.75  
150 N, 09 49' 40.55 W) is located at the head of an enclosed tidal estuary (range 3.5 m) ~100 km  
151 north of Galway Bay adjacent to Ballycroy National Park. This control site represents a sparse  
152 rural population in surrounding areas, far from urban influence, where wastewater treatment in  
153 the region is exclusively by one-off or small group septic tanks, without connection to a sewage  
154 treatment network. No direct riverine freshwater drainage adjacent to the sample site  
155 embayment is evident.

156

## 157 *2.2 Microplastic sediment sampling*

158 Polymethyl methacrylate (Perspex) core tubes (internal diameter 6.5 cm) were used to extract  
159 (15 cm) replicate intertidal sediment core samples from each site, on four sampling occasions,  
160 during the Spring low tides of September 2017, December 2017, March 2018 and June 2018.  
161 Prior to field sampling, core tubes and tools were thoroughly rinsed under high pressure tap  
162 water, further rinsed with distilled water and stored in clean paper bags. Immediately prior to  
163 sampling, core tubes and tools were further rinsed with distilled water.

164

165 Coring locations were spaced 2-3 m apart along the midline of the intertidal zone by selective  
166 random sampling (Crawford & Quinn, 2017). Core tubes were manually forced into the

167 sediments and then dug out during extraction, to ensure minimal disturbance of the sample.  
168 Tubes were immediately capped and sealed at both ends and labelled. Samples were stored  
169 upright in darkness at room temperature prior to processing.

170

### 171 *2.3 Particle size analysis (PSA)*

172 The top 2.5 cm of sediment was removed from replicate samples using a clean stainless-steel  
173 spoon, placed in individual petri trays and immediately covered. Sediment wet weight was  
174 recorded, and then following drying for 48 hrs at 70°C and for a further 24 hrs at 20°C, the dry  
175 weight was recorded. Dry sieving of sediment samples was undertaken in a sieve stack of 2  
176 mm, 1 mm, 500 µm and 250 µm, sealed with parafilm above a 1 L glass beaker. The sieve stack  
177 was shaken manually for 60 seconds. Each sediment size fraction was then placed in a labelled  
178 petri dish, weighed and covered. The < 500 to > 250 µm fractions were then placed in an  
179 additional sieve stack, comprising 125 µm, 100 µm, 63 µm and 45 µm sieves, and shaken for  
180 60 seconds. Each sediment sample fraction was again weighed and covered as above.

181

### 182 *2.4 Microplastic extraction*

183 The identification and extraction of MP particles and fibres from the upper 2.5 cm of the core  
184 was undertaken by visual examination on the 2 mm to 500 µm sediment fractions using an  
185 Olympus S251 (Tokyo, Japan) binocular microscope, at up to 40 X magnification. Plastic  
186 detritus within sieve fractions < 500 µm were identified but not included in this study due to  
187 the difficulty of accurate visual identification and handling (Hidalgo-Ruz et al., 2012). Petri  
188 dishes were placed above a black background to enhance visual differentiation of white  
189 particles. Microplastics were visually identified by a set of selection criteria: no organic or  
190 cellular structures evident, except where conjoined to probable MPs; fibres are consistent in  
191 diameter with no tapering or branching; consistency of particle colour and texture; fibres

192 remain malleable in response to manipulation. Coloured potential MP fibres and particles were  
193 extracted manually using fine tweezers and adhered to double sided tape on glass slides.  
194 White/translucent fibres were transferred into an Eppendorf tapered centrifuge tube to maintain  
195 visual clarity. Clusters frequently contained multitudes of individual fibres but were recorded  
196 as one MP find. As quality control on validity, all potential MPs were checked once again by  
197 visual examination. When biofouling hindered spectral analysis of the MP, 2 mL of hydrogen  
198 peroxide (H<sub>2</sub>O<sub>2</sub>) was added to the samples inside an eppendorf and left overnight. Samples  
199 were washed with Milli-Q™ water and left to dry prior to analysis.

200

### 201 *2.5 Sampling of sewage-related debris*

202 Post sediment sampling (Autumn 2019), a large quantity of sewage-related debris was washed-  
203 up on the shoreline at the mouth of the River Corrib in close proximity to the sediment sampling  
204 site at Mutton Island (Fig. S1a). This re-occurring accumulation consisted of a range of largely  
205 intact wipes and sanitary towels (and other personal care products) mixed and entangled in  
206 seaweed biomass (Fig. 2a - h). Samples of this material were collected for fibre extraction and  
207 polymer identification on two different occasions.

208

### 209 *2.6 Commercially available sanitary consumer products*

210 Commonly available flushable and non-flushable wet wipes and sanitary towels (n=17), both  
211 own brand and other brands, were purchased from the most common national and international  
212 retailers on the high street in Ireland for comparative purposes. A plastic polymer material  
213 component was not listed on any product packaging and the wipes were marketed under an  
214 array of descriptions, including 99% water / purified water and biodegradable. Samples of the  
215 sanitary products were air dried and a quadrat of 2 x 2 cm was removed for microscopic and  
216 spectroscopic characterization and analysis.

217

## 218 *2.7 Quality Control*

219 All laboratory surfaces and equipment were thoroughly cleaned with ethanol. Laboratory trays  
220 and petri dishes were new from pre-sealed bags and were carefully checked under a microscope  
221 before use for potential contaminants. During particle size analysis (PSA) sieving, 2 x 200 ml  
222 jars of water were placed in the laboratory working area each day to capture airborne  
223 contaminants, which were then filtered through Whatman™ 0.45 µm hydrophilic nylon  
224 membrane filters. During the MP extraction phase, petri dishes were placed either side of the  
225 microscope each day to capture airborne contaminants. Filters and petri dishes were examined  
226 by binocular microscope for contaminants. Whenever possible, clothing made of cotton  
227 material was used throughout the study, but in either case base material and colour were  
228 registered for control purposes.

229 Sediment loss due to airborne scatter during the PSA process was 1%. Potential airborne  
230 contaminants captured during the sieving phase (n = 76) and total airborne contamination  
231 during the MP extraction phase (n=7) were very low. Contaminants were considered negligible  
232 and well within the error of the quantification (Tables S1-S3).

233

## 234 *2.8 Raman Spectroscopy*

235 Random samples of white fibrous field samples (n = 100) were analysed for polymer  
236 identification by Raman Spectroscopy: 28.7% (n = 86) of total Mutton Island white/translucent  
237 fibres, 31.8% (n = 7) from Bell Harbour white fibres, and 30.4% (n = 7) from Bellacragher  
238 white fibres. Commercially available sanitary textile products (n = 17) were analysed. Analysis  
239 was undertaken by a Raman spectrometer (Horiba LabRAM II, Horiba Jobin-Yvon, France)  
240 equipped with a 600 groove mm<sup>-1</sup> diffraction grating, a confocal optical system, a Peltier-

241 cooled CCD detector, and an Olympus BX41 microscope (Kostrzytsia et al., 2018). The  
242 instrument was calibrated by zero-order correction on a known band of 1/520 cm etched on a  
243 crystalline silicon wafer (Jonker et al., 2015). Analysis was mostly conducted with a 532 nm  
244 laser; one highly reflective sample was identified using a 785 nm laser. The measurements were  
245 performed with acquisition times of 4-30 seconds over a spectral range of 100-3500 cm<sup>-1</sup>. For  
246 identification purposes, the obtained spectra for each sample were compared to a spectral  
247 reference library (KnowItAll, Bio-Rad) and an in-house extension of the library with additional  
248 spectra from environmental MP collected from the intertidal zone and two clothing garments  
249 manufactured from polyester (PES) (Table 1). Replicate spectra were recorded for each sample.

250

### 251 *2.9 Scanning Electron Microscopy (SEM)*

252 Field samples and commercial textile samples were gold coated (Emitech SC500, Quorum  
253 Technologies Ltd, West Sussex, United Kingdom) and subjected to scanning electron  
254 microscopy (SEM) in backscatter mode using a Hitachi model S-2600N (Hitachinaka, Japan).  
255 The analyses were performed at an acceleration voltage of 15 kv, an emission current range  
256 (Ie) of 68-106  $\mu$ A, and a working distance range of 6.8-11 mm (Morrison et al., 2009). The  
257 images were acquired under variable pressure mode at 50 Pa.

258

## 259 **3. Results**

### 260 *3.1 Wet wipe and sanitary towel polymer identification*

261 The most common polymer used in the manufacture of commercially available wet wipes was  
262 identified as non-woven PET fibres (Table 1). Five consumer wipe products were identified as  
263 PET and cellulose and two were identified as containing only PET; however, the main

264 component of one wipe brand was identified as polypropylene (PP) (Table 1). Out of the eleven  
265 commercially available wipe products, four brands were labelled as flushable (Table 1) and  
266 two of these flushable wipes were observed as manufactured from a mix of PET and cellulose  
267 (Fig. 3). The remaining seven wipes were marked as non-flushable: three comprised a mix of  
268 PET and cellulose, one cellulose alone, two PET alone and one PP alone (Table 1).

269 Six female hygiene products (sanitary pads/towels) were primarily composed of non-woven  
270 PP or HDPE/PP fibres, while one of these contained HDPE fibres at the outer edge of the pad.  
271 One other brand of sanitary towel was identified as a polyethylene (PE) film (Table 1).

272 Sanitary towels and wet wipes collected as macro debris from the intertidal zone adjacent to  
273 Mutton Island WWTP were also identified as non-woven PP and PET MP fibres, respectively.

274

### 275 *3.2 MP occurrence in intertidal sediment samples*

276 Intertidal surficial sediments at the Mutton Island mid-tide line comprise a moderately sorted  
277 fine sand to silt, becoming well sorted in December 2017. Bell Harbour sediments are very  
278 well sorted, indicative of silt and clay; gravel outliers evident in the summer season were  
279 largely absent in December 2017 and March 2018. Bellacragher is predominately a poorly  
280 sorted coarse medium to fine sand, becoming moderately sorted in December 2017.  
281 Microplastics were recorded in all the sediment samples, a total of 433 MPs were recorded  
282 (Tables S1-S3), of which 78% occurred at Mutton Island, 15.2% at Bell Harbour and 6.7% at  
283 Bellacragher.

284 Total MP accumulation ( $\text{kg}^{-1}$  dry weight [dw]) at Mutton Island remained reasonably  
285 consistent in September 2017, March 2018 and June 2018 (Table 2), with an average total MP  
286 content of  $534 (\pm 48) \text{ kg}^{-1} \text{ dw}$ ; however, in December 2017, a three-fold increase ( $1441 \pm 631.2$   
287  $\text{MP kg}^{-1}$ ) in MPs levels was observed (Table 2).

288 Bell Harbour displayed a similar MP loading over the first two sampling occasions with a  
289 decrease in June 2018. In contrast, the highest MP loadings at Bellacragher occurred in June  
290 2018 (Table 2). However, the range of MP loadings at both these rural sites were well below  
291 the levels adjacent to the WWTP at Mutton Island (Table 2).

292 Microplastic fibres were the most common MP type identified at the three sampling locations,  
293 an average total of 2768 white fibres  $\text{kg}^{-1}$  dw were reported from Mutton Island samples,  
294 corresponding to 91% of the average sum of MP identified in intertidal sediments adjacent to  
295 the WWTP at Mutton Island (Table 2).

296 While an average sum of 393 white fibres  $\text{kg}^{-1}$  dw were observed at Bell Harbour (Table 2), in  
297 contrast, an average sum of 157 MP  $\text{kg}^{-1}$  dw (white and non-white) were observed from on the  
298 Bellacragher samples (Table 2).

299

### 300 *3.3 Intertidal white microfibre polymer identification*

301 Randomly selected white fibres extracted from the intertidal sediments ( $n = 100$ ) were analyzed  
302 for polymer type. White fibres from the sediments at Mutton Island ( $n = 86$ ) were identified as  
303 PET ( $n = 51$ ; 59.3%), PP ( $n = 23$ ; 26.7%), PES ( $n = 9$ ; 10.4%), polystyrene (PS;  $n = 2$ ; 2.3%)  
304 and acrylonitrile butadiene styrene (ABS;  $n = 1$ ; 1.1%), while in Bell Harbour, ( $n = 7$ ) white  
305 fibres were identified as ABS ( $n = 4$ ; 57.1%), PP ( $n = 2$ ; 28.5%) and PS ( $n = 1$ ; 14.2%); and in  
306 Bellacragher ( $n = 7$ ) as ABS ( $n = 6$ ; 85.7%) and PS ( $n = 1$ ; 14.2%).

307

### 308 *3.4 Spectral analyses of white microfibres from intertidal sediments, sewage-derived waste and* 309 *commercially available sanitary products*

310 Comparative images at 35X magnification of white/translucent field samples and product  
311 fibres appear to show similar characteristics of texture, shape and colour (Figs. 4-5). White  
312 microfibrils extracted from the sediment cores at the sampling site adjacent to Mutton Island  
313 WWTP, identified as PET, appear to share a very similar morphology to commercially available  
314 wipes manufactured from PET (Fig. 4a-e). Figure 4 highlights the similarity in microfibrils  
315 from commercial baby wipes and microfibrils collected from the same location at Mutton  
316 Island (from sediment cores and washed-up sewage-derived waste), both identified as PET  
317 (Fig. 4e). Polyethylene terephthalate white microfibre field samples (at a magnification of  
318 X5000 under the SEM) appear to display robust characteristics in response to marine  
319 environmental conditions, appearing resistant to degradation and fragmentation (Fig. 4d).

320 Polypropylene microfibrils in the sediments near the WWTP appear identical to PP fibres from  
321 a commercially available sanitary pad (Fig. 5a-d). SEM images of samples of MP fibres from  
322 the sewage-derived waste from this location appear to display brittle characteristics, like  
323 cracking and fragmenting in response to seawater exposure (Fig. 5d). The PP fibres from the  
324 sediments are also very similar to another commercially available female hygiene product (Fig.  
325 5f-g).

326

#### 327 **4. Discussion**

328 The sample sites chosen for investigation in this study provide a representation of modern  
329 urban and rural European coastal environments. It is well established that WWTP effluent  
330 contributes to the MP loading of nearby shoreline sediments (Browne et al., 2011; Estahbanati  
331 and Fahrenfeld, 2016; Talvitie et al., 2015; Ziajahromi et al., 2017). Microplastic abundance  
332 and frequency at Bellacragher was consistently low with no obvious point sources. Relatively  
333 high densities of ABS fibres in June 2018 were likely an anomaly, which resulted from the

334 presence of ABS piping and fittings in the vicinity of the sampling site. Acrylonitrile butadiene  
335 styrene is widely used in the manufacture of piping fittings due to its strength and resistance to  
336 abrasion, and the fibres at Bellacragher may be long form fragments of white plastic panelling,  
337 which was cut for pontoon construction locally. The presence of ABS in this study is no surprise  
338 as it has been reported to be present in WWTPs, beach and lake sediments worldwide due to  
339 anthropogenic activities and dispersion to the marine environment through the sewage and  
340 freshwater systems (Hamid et al., 2018; Kang et al., 2018; Kazour et al., 2019). Bell Harbour,  
341 on the southern margins of Galway Bay, which exceeded the loading at Bellacragher, was still  
342 deemed to have a relatively low MP loading compared with the main sampling site at Mutton  
343 Island.

344 The Mutton Island sampling site is in an urban setting adjacent to a WWTP and is in close  
345 proximity to the outflow of the Corrib River, which adds a freshwater-derived flux to the  
346 sample area. The variability in MP loading over the 10-month period of this study was likely  
347 due to local hydrodynamics, deposition from currents, and sediment turnover in response to  
348 seasonal climatic variability as well as proximate human activity. The nearby intertidal zone is  
349 prone to the accumulation of high volumes of washed-up sewage-derived debris on a frequent  
350 basis. Excessive MP loading in December 2017 was likely induced by heavy precipitation  
351 episodes during a south-westerly storm front. Elevated debris loading on this occasion may  
352 result from combined sewer overflows, where excessive input of drainage water exceeds  
353 WWTP effluent capacity and is released untreated in the overflow. Combined sewer overflows  
354 from overloading have been reported from many other cities globally (Ellis, 2006; Gasperi et  
355 al., 2008; Kazour et al., 2019; Rathnayake 2019; Scoullou, 2020; Weyrauch et al., 2010). These  
356 overflows in turn can give rise to washed-up sewage-derived waste and debris on the  
357 surrounding coast (Krelling and Turra, 2019; MCS, 2019; Rangel-Buitrago, et al., 2019). It is  
358 estimated that the quantities of wet-wipes washing up on the coastline in the UK increased by

359 94% since 2017 and 400% during the last decade (MCS, 2019; Pantoja-Munoz et al., 2018;  
360 The Guardian, 2015)

361 Blumenröder et al. (2017) report high MP loadings in intertidal sediments (6,500 MPs per kg<sup>-1</sup>  
362 dw) adjacent to a WWTP in Scotland (Stromness, Orkney Islands). The sampling regime, as  
363 described in the Orkney Islands, was comparable to the field sampling methodology in the  
364 current study, i.e. intertidal sediments collected from relatively sheltered depositional coastal  
365 embayments; 6,083 MPs per kg<sup>-1</sup> dw are reported for Mutton Island which serves a population  
366 some 40 times that of Stromness. This may be a function of the experimental design, as the  
367 current study extracted MPs from the 2 mm, 1 mm, and 500 µm sieved fractions, and therefore  
368 MPs identified in our study are likely an underestimation as the greatest numbers of MPs occur  
369 in finer grained sediments (Corcoran et al., 2020; Mintenig et al., 2017).

370 Given the preponderance of white MP fibres in marine sediments adjacent to the Mutton Island  
371 WWTP, it is reasonable to assume some of the fibres were derived from clothing items released  
372 from grey water discharges (Kang et al., 2018; Talvitie et al., 2015; Yang et al., 2019). Although  
373 many clothing items are manufactured from PET/Polyester, no white MP fibres derived from  
374 polymers, also associated with clothing materials such as acrylic fabrics and poly-cotton  
375 blends, were detected in the current study (Napper & Thompson, 2016).

376 While it is considered that most modern WWTPs effectively retain 95 to 99% of MP particles  
377 and that wastewater effluents account for the release of the remainder (Gies et al., 2018; Habib  
378 et al., 2020; Mintenig et al., 2017), sewer overflows and the subsequent shoreline deposition  
379 of sanitary waste have not previously been thoroughly investigated as a source of white MP  
380 fibres in the marine environment. The white MP fibre loading of sediments at Mutton Island  
381 was almost four times that of Bell Harbour and eighteen times that observed at Bellacragher.  
382 The samples of sanitary-related macro debris (wipes and sanitary towels) collected from the

383 intertidal zone near the WWTP following a heavy rainfall event were mostly comprised of PET,  
384 with only a quarter of the samples analyzed presenting as a mix of PET and cellulose, and over  
385 80% of the wipes in the shoreline waste were identified as non-flushable due to their polymer  
386 composition following the International Water Services Flushability Group and non-woven  
387 textile industry guidelines (INDA/EDANA, 2018; IWSFG, 2018). The flushable wipes market  
388 has increased significantly in value and is currently estimated to have reached \$2.7 billion  
389 (Atasağun and Bhat, 2019), with the entire global personal care wipes market valued at \$15.8  
390 billion in 2018 (Grand View Research, 2019). Consequently, this rapid growth globally has  
391 resulted in an increase in the quantity of wipes found on the UK coastline, a 400% increase in  
392 the last decade (MCS, 2016; MCS, 2019). This is a phenomenon likely occurring globally in  
393 concurrence with combined sewage overflows. Sanitary towels contributed significantly to the  
394 washed-up sewage-related debris in the present study along with wipes and both may fragment  
395 into white MP fibres, which potentially pose a hazard to marine organisms.

396 The analyses of commercially available flushable and non-flushable wipes revealed that over  
397 90% contained either cellulose, a combination of PET and cellulose, or PET alone in their  
398 matrix, which was also reported in other studies investigating fibre composition of wipes  
399 (Durkan and Karadagli 2019; Pantoja-Munoz et al., 2018; Khan et al., 2019). Microscopic and  
400 spectroscopic analysis of flushable and non-flushable wipes in this study revealed a similar  
401 structure/construction and fibre composition. It was microscopically possible to identify and  
402 separately analyse cellulose and MP fibres (Figs. 4a and 6c). Cellulose fibres presented a more  
403 rough and ribbon-like appearance than PET, PE and PP, which presented a more uniform and  
404 three-dimensional shape, although this is not always the case (Dyachenko et al., 2017; Lares et  
405 al., 2018 Norén, 2007). The presence of white MP fibres in the wipes reinforces their strength  
406 and renders them more durable than wipes manufactured from cellulose alone (MESUGC,  
407 2019).

408 In order for a wipe to be considered flushable, there is a requirement that the product is  
409 manufactured from natural polymers (plant-based source, i.e. cellulose, cotton) which degrade  
410 during the wastewater treatment process without affecting the system or downstream  
411 environment (IWSFG, 2018). Wipes (both flushable and non-flushable) comprising a mixture  
412 of cellulose and PET, or those made from petrochemical derivative polymers (i.e. PET, PP,  
413 HDPE) alone, are less susceptible to degradation but subject to fragmentation through defects,  
414 residual stress and sheer stress forces of mechanical mixing in a WWTP (Enfrin et al., 2020a;  
415 Ghorbani et al., 2013). Wet wipes and other macro debris cause operational problems in sewer  
416 system through the formation of long length fibres that lead to the formation of ropes (Fig. 2c-  
417 f). These structures block the normal sewage pathway, causing adverse effects to microbial  
418 communities and biological processes in the wastewater system (Atasağun and Bhat, 2019;  
419 Durukan & Karadagli, 2019; Scolz and Sigmund, 2012).

420 Manual manipulation of the wipes in the current study also highlighted that wipes  
421 manufactured from cellulose alone were more easily torn, fragmented and disintegrated when  
422 submerged and agitated in water following the guidelines of the International Water Services  
423 Flushability Group (IWSFG PAS1, 2018). By contrast, wipes made of PET or PET/cellulose  
424 combinations were more difficult to tear apart and less susceptible to disintegration when  
425 submerged and agitated in water, rendering them unsuitable to be flushed down toilets  
426 (INDA/EDANA, 2018; IWSFG PAS2, 2018; Khan et al., 2019). Cellulose degradation under  
427 aerobic and anaerobic conditions is well-documented (Demain et al., 2005; Leschine, 1995;  
428 McDonald et al., 2012; Smith, 1994; Song et al, 2013), unlike PES and PET fibres which are  
429 not subject to bacterial breakdown in the environment and in WWTPs (Zambrano et al., 2020).

430 Although flushable wipes comprising cellulose fibres alone degrade in the wastewater system,  
431 the current study has shown that more than 50% of wipes labelled as flushable contain PET

432 and hence are non-degradable and produce MP fibres that are available to aquatic organisms  
433 when released to the marine environment.

434 Out of the 11 commercially available wipes analysed, 36% were distinctly labelled as flushable  
435 and included secondary instructions for disposal, the remaining 64% of these products (non-  
436 flushable) were clearly marketed as non-flushable, but disposal instructions were lacking.  
437 While all non-flushable wipe packages displayed the “Do Not Flush” logo, none adhered to  
438 industry guidelines for presentation on the packaging (placement, colour and size). According  
439 to these guidelines, there is a requirement to position the “Do Not Flush” symbol on the front  
440 panel, beside where individual wipes are removed from the package prior to use, as opposed  
441 to the rear or obscured by packaging seals or folds. In addition, the logo should contrast with  
442 the background with a diameter of 4-6% the size of the packaging panel (INDA/EDANA,  
443 2018). All the non-flushable brands tested in this study failed to meet this criterion and only  
444 one flushable brand adhered to the correct labelling guidelines.

445 The presence of PET and PET/cellulose wipes among sewage-related waste washed-up on the  
446 beach are indicative of the inappropriate disposal of non-flushable wipes. This would suggest  
447 that some consumers are unlikely to follow packaging disposal instructions and flush these  
448 products indiscriminately. On the other hand, the inappropriate disposal of flushable wipes may  
449 be more a function of incorrect labelling resulting in a lack of awareness by consumers.

450 Two billion single-use menstrual products are flushed down the toilet each year and tampons,  
451 pads and applicators generate 200,000 tonnes of waste per annum in the UK alone (Wen, 2020).  
452 Furthermore, manufacturers of female hygiene products fail to adequately reveal the precise  
453 composition of sanitary towels (which are 90% plastic) as part of the labelling (Wen, 2020).  
454 Although the market for reusable and biodegradable female products is growing, disposable  
455 towels are still the most common product used due to convenience and costings, their presence

456 in WWTPs and beaches should be of concern not only for protection of wastewater systems  
457 but also for environmental and public health hazards.

458

## 459 **5. Conclusion**

460 The marine sediments adjacent to a WWTP have been shown to be consistently strewn with  
461 white MP fibres that were comparable with the white fibres from sewage-related waste and  
462 commercially available consumer sanitary products (wet wipes and sanitary towels). Nearly  
463 every MP fibre identified had a profile (spectra, shape, and size) similar to the white fibres  
464 present in the commercial wipes and sanitary towels analyzed. Although the WWTP process  
465 removes almost all MPs and fibres, the release of sewage-related waste containing wipes and  
466 sanitary towels through combined sewage overflows impacts public health and the  
467 environment. This demonstrates the downstream consequence of the misleading marketing of  
468 non-woven textile products, which in fibrous form may have been underestimated in studies to  
469 date. The results of this study show that wet wipes and sanitary towels are a source of  
470 unaccounted white MP fibres in the marine environment, not all flushable wipes are  
471 biodegradable and sanitary towels contain MP fibres. There is a need for increased public  
472 awareness of MP pollution in the marine environment from the inapt disposal of sanitary  
473 products down the toilet instead of diversion to alternative land-based waste management.

474

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483

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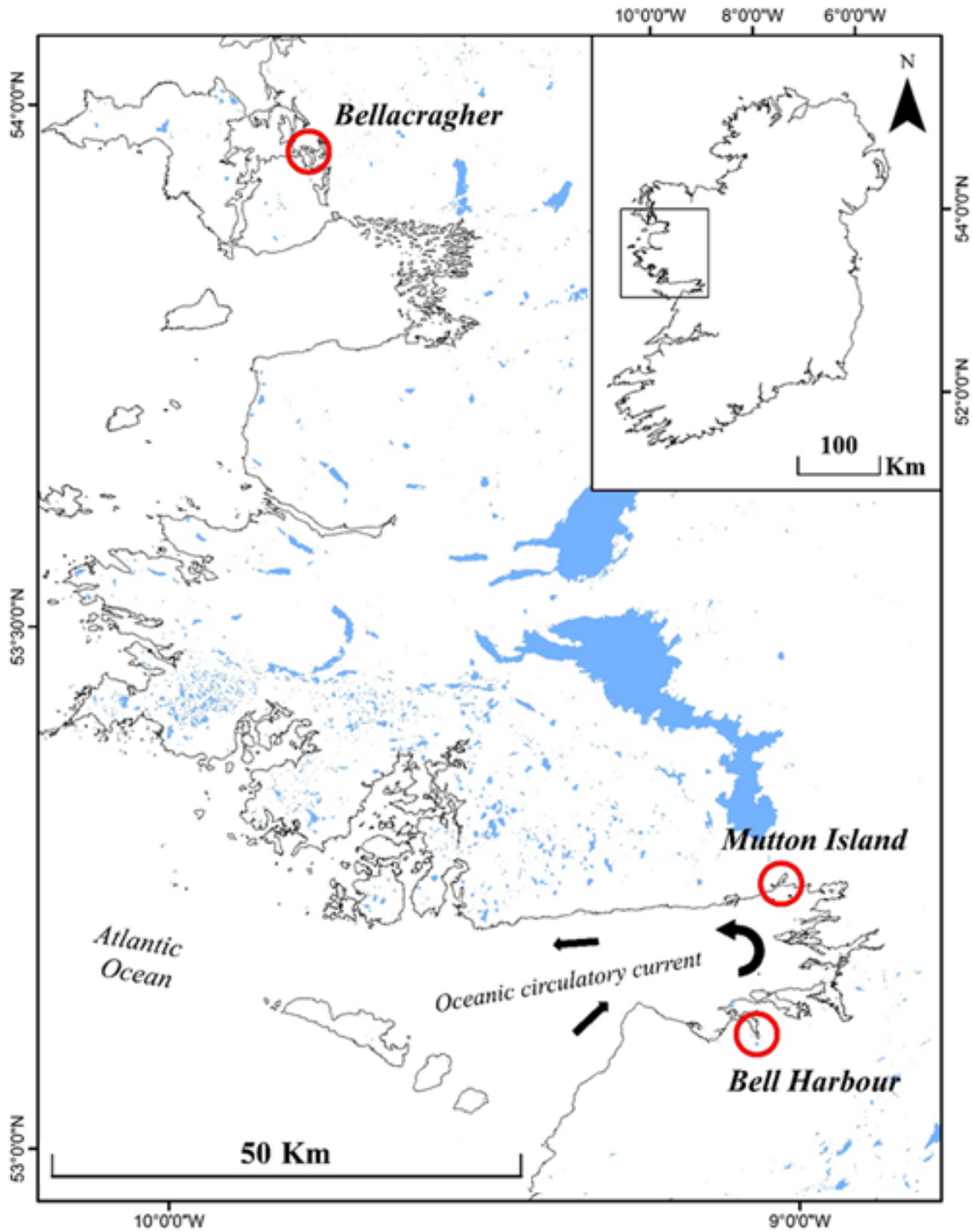
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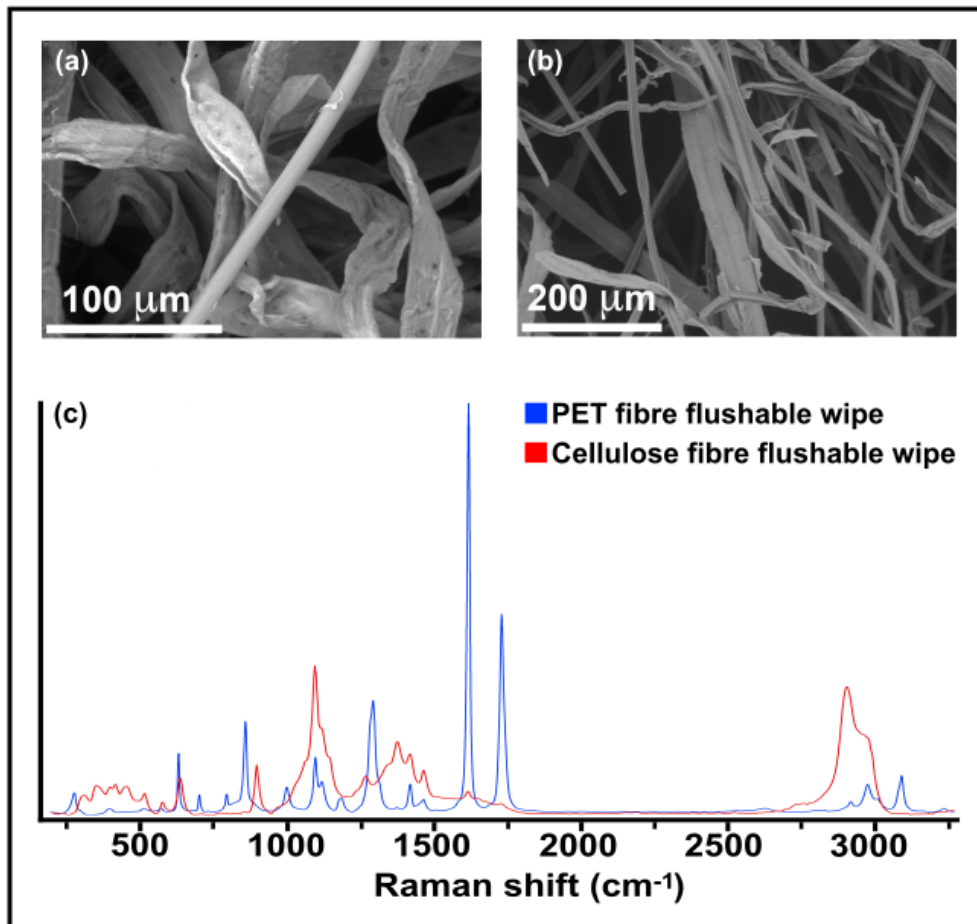
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**Figure 1.** Map showing locations of sampling sites (Bellacragher, Mutton Island and Bell Harbour) on the west coast of Ireland. Arrows illustrate oceanic circulatory current in Galway Bay. Image: OSi Licence number NUIG220212.



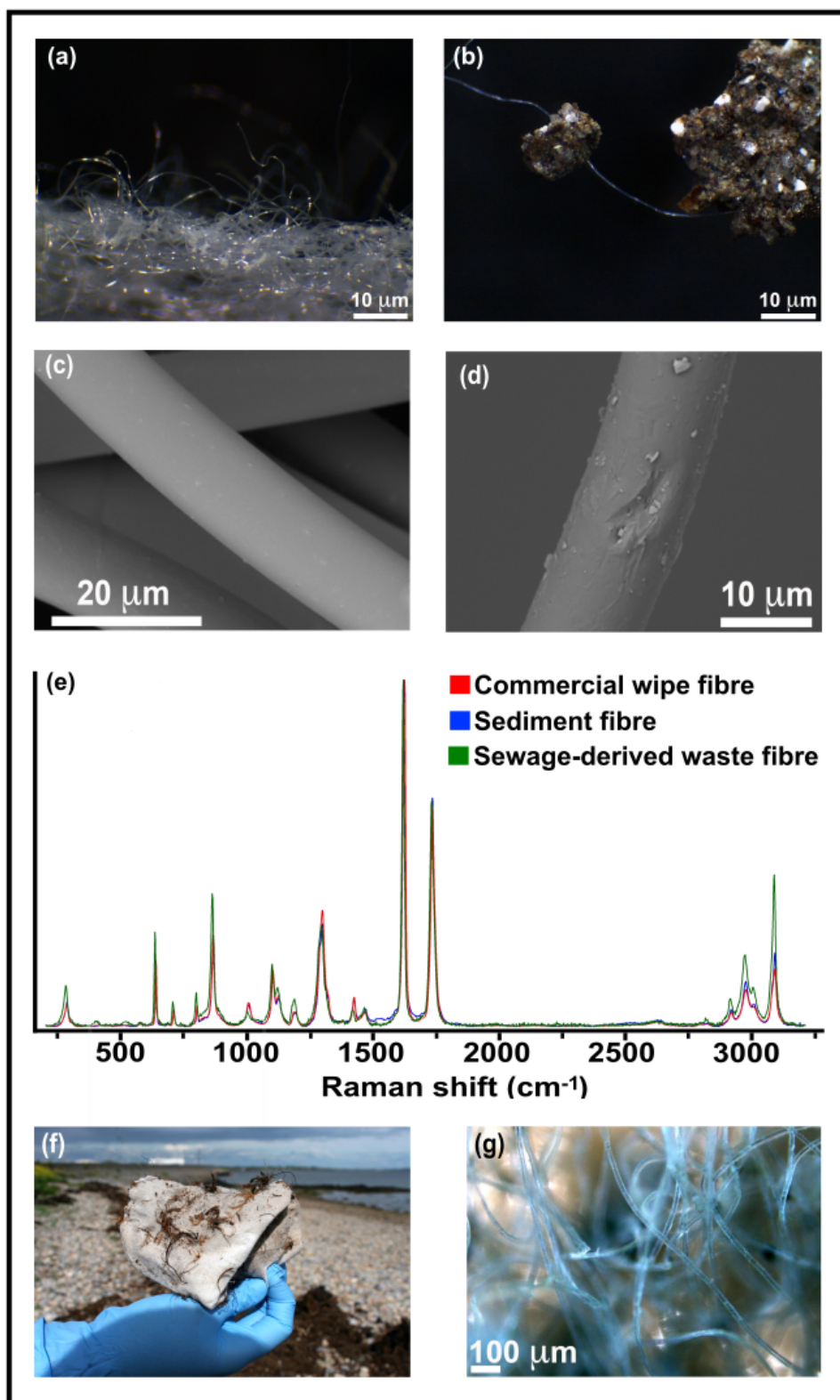
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**Figure 2.** (a-f); Washed up deposit of Sewage-derived debris intertangled with seaweed biomass littering the coastline in the vicinity of Mutton Island, including wipes (g) and sanitary pads (h).

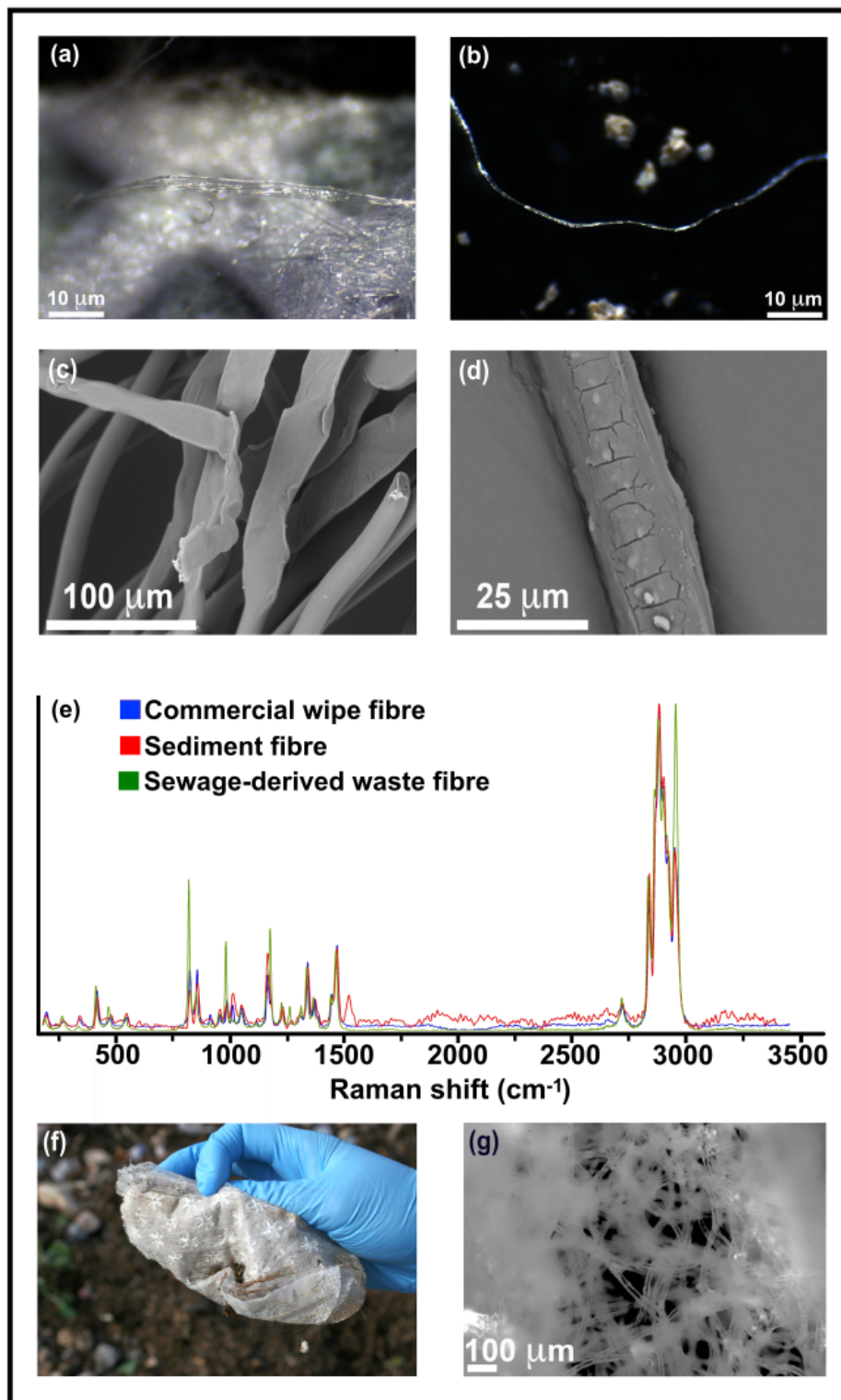


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**Figure 3.** SEM images of commercially available flushable wipes (a, b) with (a) identified as a combination of PET & cellulose and (b) cellulose alone using Raman Spectroscopy (c).



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 822 **Figure 4** Images and Raman spectra of PET white fibres extracted from commercially available  
 823 wipes and intertidal sediments; a) microscope image of commercial wipe MicroFibre (MF), b)  
 824 microscope image of MF extracted from the intertidal sediments, c) scanning electron  
 825 microscope (SEM) image of a commercially available wipe, d) SEM image of MF from the  
 826 intertidal sediments, e) Raman spectra of commercially available wipes, sediment and field  
 827 sample identified as white PET fibres; f, g) macro and micro image of field sample of wipe  
 828 from the washed up deposit of sewage derived waste, identified as white PET fibres.



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**Figure 5.** Images and Raman spectra of PP white fibres extracted from commercially available sanitary towels and intertidal sediments; a) microscope image of a commercially available sanitary towel MicroFibre (MF), b) microscope image of MF extracted from the intertidal sediments, c) scanning electron microscope (SEM) image of a commercially available sanitary towel, d) SEM image of MF from the intertidal sediments, e) Raman spectra of commercially available sanitary towel, sediment and field sample identified as white PP fibres; f, g) macro and micro image of field sample of sanitary towel from the washed up deposit of sewage derived waste, identified as white PP fibres.

839 **Table 1.** Polymer composition of commercial non-woven sanitary products and their  
 840 flushability and environmental samples collected from the washed-up deposit of sewage-  
 841 derived waste near Mutton Island.

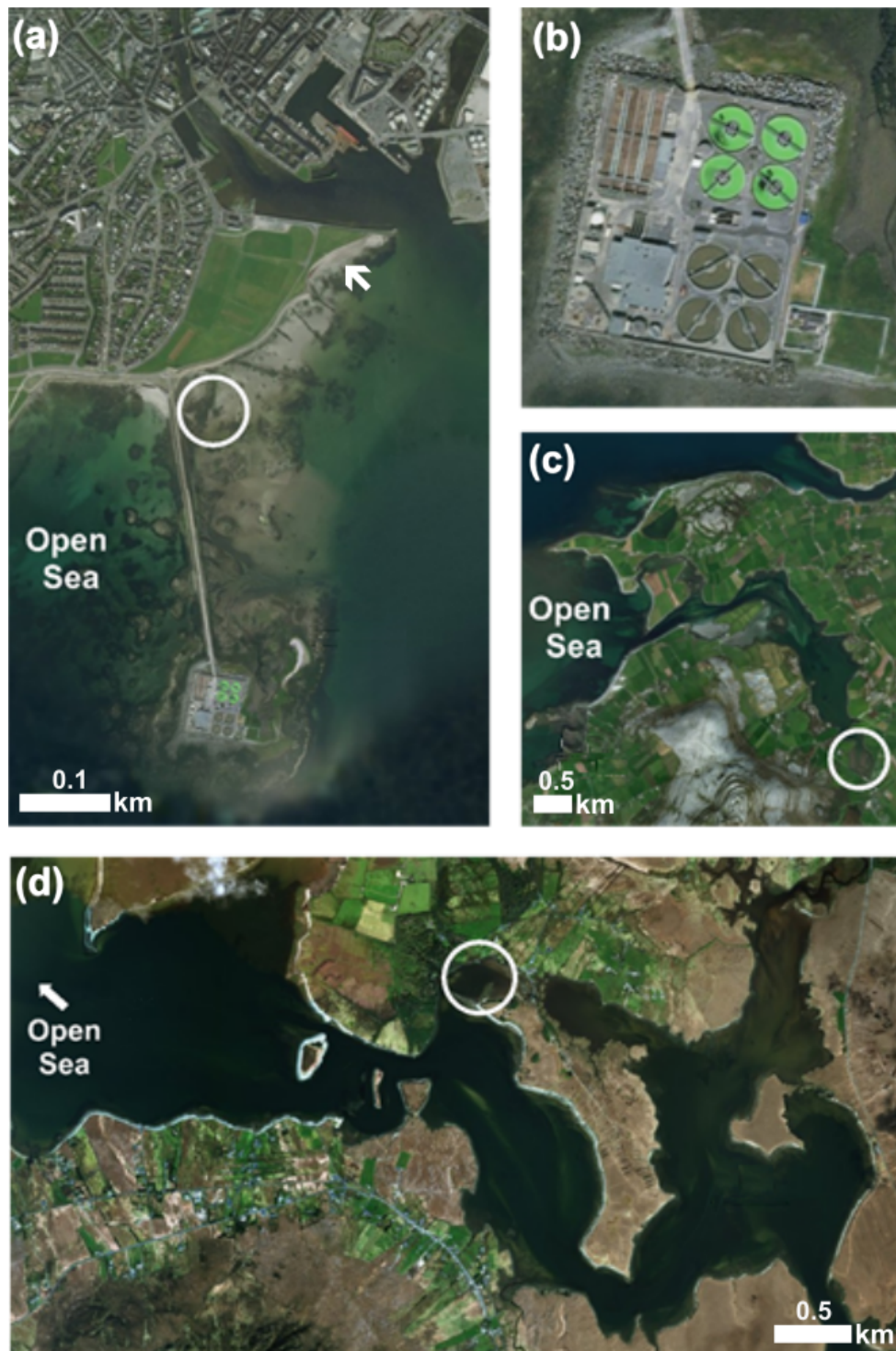
Code	Description	Non-flushable	Flushable	Raman ID
<b>Commercially available sanitary products</b>				
P1	Flushable toilet wipes		✓	Cellulose
P2	Flushable moist toilet tissue wipes		✓	PET & Cellulose
P3	Toilet wipes (fragrance free)		✓	Cellulose
P4	Toilet wipes		✓	PET & Cellulose
P5	99% water, organic cotton wipe	✓		PET & cellulose
P6	Ultra-soft wipes	✓		PET
P7	Baby wipes	✓		PET & cellulose
P8	Baby wipes, 100% biodegradable	✓		Cellulose
P9	Baby wipes, chemical free	✓		PET & cellulose
P10	Baby wipes, Cotton soft fragrance free	✓		PET
P11	Baby wipes	✓		PP
P12	Sanitary Pad 1	✓		PP
P13	Sanitary Pad 2	✓		PE
P14	Sanitary Pad 3	✓		HDPE & PP
P15	Sanitary Pad 4	✓		PP
P16	Sanitary Pad 5	✓		PP
P17	Sanitary Pad 6	✓		HDPE & PP
P18	Control clothing item 1	✓		PES
P19	Control clothing item 2	✓		PES
<b>Sewage-derived samples</b>				
Ev1	Washed-up wipe			PET & Cellulose
Ev2	Washed-up wipe			PET
Ev3	Washed-up wipe			PET
Ev4	Washed-up wipe			PET
Ev5	Washed-up wipe			PP
Ev6	Washed-up wipe			PET
Ev7	Washed-up sanitary towel			PET
Ev8	Washed-up sanitary towel			PP

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844 **Table 2.** Spatial and temporal quantity of total microplastics and white fibres found per kg of  
 845 sediment.

	<b>Month</b>	<b>Total Microplastic ( kg<sup>-1</sup>)</b>	<b>Total White Fibres (kg<sup>-1</sup>)</b>
<b>Mutton Island</b>	Sep	553 (± 101.5)	524 (±97.3)
	Dec	1441 (± 631.2)	1323 (± 651.8)
	Mar	479 (268.6)	444 (± 238.9)
	Jun	569 (154.4)	477 (142.5)
<b>Bell Harbour</b>	Sep	295 (± 274.5)	102 (± 97.3)
	Dec	248 (± 44.4)	130 (± 31.2)
	Mar	158 (± 13.5)	95 (± 75.3)
	Jun	113 (± 102.5)	66 (± 55.9)
<b>Bellacragher</b>	Sep	41 (± 6.7)	23 (± 32.6)
	Dec	7 (± 10.3)	0 (± 0.0)
	Mar	32 (± 29.7)	32 (± 29.7)
	Jun	77 (± 31.9)	77 (± 31.9)

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878 **Figure S1.** Sample sites, (a) Mutton Island, Galway City Ireland primary sample site adjacent  
879 to wastewater treatment plant (WWTP), with arrow indicating location of re-occurring  
880 deposition of washed-up sewage derived waste; (b) closer view of Mutton Island WWTP; (c)  
881 Bell Harbour, Co. Clare Ireland, control site; (d) Bellacragher, Co. Mayo Ireland, control site  
882 (Image source: Esri, DigitalGlobe "Imagery" [basemap]. 0.5m resolution. "World Imagery  
883 Map". Captured March 2012).

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886 **Table S1:** Spatial and temporal quantity of total microplastics, white fibres and controls per  
 887 dry sediment sample for Mutton Island sampling site.

Month	Sample	Samples Weight (g)	Total MP	Control MP	Total White Fibres	Control white
Sep-17	Replica 1	76.92	37		35	
	Replica 2	62.44	39		37	
	<b>Total</b>	<b>139.36</b>	<b>76</b>	<b>10</b>	<b>72</b>	<b>2</b>
Dec-17	Replica 1	38.68	73		69	
	Replica 2	45.24	45		39	
	<b>Total</b>	<b>83.92</b>	<b>118</b>	<b>5</b>	<b>108</b>	<b>3</b>
Mar-18	Replica 1	53.82	36		33	
	Replica 2	72.65	21		20	
	<b>Total</b>	<b>126.47</b>	<b>57</b>	<b>7</b>	<b>53</b>	<b>3</b>
Jun-18	Replica 1	79.65	54		46	
	Replica 2	71.8	33		27	
	<b>Total</b>	<b>151.45</b>	<b>87</b>	<b>6</b>	<b>73</b>	<b>2</b>

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889 **Table S2:** Spatial and temporal quantity of total microplastics, white fibres and controls per  
 890 dry sediment sample for Bell Harbour sampling site.

Month	Sample	Samples Weight (g)	Total MP	Control MP	Total White Fibres	Control white
Sep-17	Replica 1	40.91	20		7	
	Replica 2	59.61	6		2	
	<b>Total</b>	<b>100.52</b>	<b>26</b>	<b>4</b>	<b>9</b>	<b>1</b>
Dec-17	Replica 1	39.35	11		6	
	Replica 2	55.36	12		6	
	<b>Total</b>	<b>94.71</b>	<b>23</b>	<b>7</b>	<b>12</b>	<b>2</b>
Mar-18	Replica 1	23.88	4		1	
	Replica 2	20.22	3		3	
	<b>Total</b>	<b>44.1</b>	<b>7</b>	<b>13</b>	<b>4</b>	<b>3</b>
Jun-18	Replica 1	74.86	3		2	
	Replica 2	37.82	7		4	
	<b>Total</b>	<b>112.68</b>	<b>10</b>	<b>7</b>	<b>6</b>	<b>2</b>

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894 **Table S3:** Spatial and temporal quantity of total microplastics, white fibres and controls per  
 895 dry sediment sample for Bellacragher sampling site.

Month	Sample	Samples Weight (g)	Total MP	Control MP	Total White Fibres	Control white
<b>Sep-17</b>	Replica 1	81.6	3		0	
	Replica 2	86.63	4		4	
	<b>Total</b>	<b>168.23</b>	<b>7</b>	<b>5</b>	<b>4</b>	<b>2</b>
<b>Dec-17</b>	Replica 1	80.25	0		0	
	Replica 2	68.81	1		0	
	<b>Total</b>	<b>149.06</b>	<b>1</b>	<b>7</b>	<b>0</b>	<b>2</b>
<b>Mar-18</b>	Replica 1	90.63	1		1	
	Replica 2	94.31	5		5	
	<b>Total</b>	<b>184.94</b>	<b>6</b>	<b>6</b>	<b>6</b>	<b>2</b>
<b>Jun-18</b>	Replica 1	109.76	6		6	
	Replica 2	90.24	9		9	
	<b>Total</b>	<b>200</b>	<b>15</b>	<b>5</b>	<b>15</b>	<b>1</b>

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