



The role of wet wipes and sanitary towels as a source of white microplastic fibres in the marine environment

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ABSTRACT

Understanding source elements of the ocean plastic crisis is key to effective pollution reduction management and policy. The ubiquity of microplastic (MP) fibres in the oceans is considered to derive primarily from clothing fibres released in grey water. Microplastic fibres degraded from widely flushed personal care textile products (wet wipes and sanitary towels) have not been clearly identified in aquatic systems to date. Unregulated personal hygiene and sanitary product labelling fails to identify textile materials. This study demonstrated that white MP fibres in sediments adjacent to a wastewater treatment plant (WWTP) are comparable with white fibres from sewage-related waste and commercially available consumer sanitary products. Commercially available non-flushable wipes are manufactured from either polyethylene terephthalate (PET), polypropylene (PP), or a combination of PET and cellulose. Fifty percent of brands labelled flushable that were tested were comprised of a mixture of PET and cellulose and the remainder of cellulose alone. Sanitary towels are made from PP, PE, or a combination of high-density polyethylene (HDPE) and PP. The accumulation of large quantities of washed-up sewage-related macro-debris (including wet wipes and sanitary towels) intermingled with seaweed biomass adjacent to the WWTP was associated with a combined sewer overflow. Microplastic fibres extracted from this waste were similar to those extracted from intertidal sediments in close proximity to the WWTP over a ten-month period. In comparison, fibres extracted from locations spatially removed from the WWTP were primarily comprised of ABS, PP and polystyrene. The results confirm that wet wipes and sanitary towels flushed down toilets are an underestimated source of white MP fibres in the environment. Given the global distribution and projected growth of the non-woven textile industry, there is a need for increased public awareness of MP pollution in the marine environment from the inappropriate disposal of sanitary products down the toilet, instead of diversion to alternative land-based waste management.

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1. Introduction

Microplastics (MPs) <5 mm in size (maximum dimension) are a contaminant of increasing concern within aquatic systems, known to enter the food chain and act as a vector for potentially harmful contaminants (Galafassi et al., 2019; Thompson et al., 2004; Zhao et al., 2018). Primary sources of MPs in the marine environment

include micro beads as a component of cosmetic products, and secondary sources include micro particles of larger plastic debris degraded through wave action and ultraviolet light (UV) and synthetic clothing fibres (Hidalgo-Ruz et al., 2012). Benthic sedimentary cores in both coastal and continental shelf margins show ubiquitous MP contamination (Martin et al., 2017); MPs that remain buoyant follow circulatory currents, converge in oceanic gyres and reach the most remote areas of the planet, including the northern and southern polar circles (Barnes et al., 2010; Bergmann and Klages, 2012).

Understanding the possible sources of MP entering near shore marine environments is key to developing effective pollution

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

As plastic products are generally manufactured through moulding, casting and extrusion, defects and residual stress in the polymer contributes to fragmentation (Ghorbani et al., 2013). Microplastics released in wastewater treatment plants (WWTPs) are subject to shear stress forces of turbulence, pumping and mechanical mixing which increases fragmentation of MPs (Enfrin et al., 2020a). Microplastics may also impact on WWTP performance through the fouling of filtration membranes (Enfrin et al., 2020b). In addition, MPs may act as toxicological vectors, adsorbing concentrations of contaminants commonly found in WWTPs such as pharmaceuticals (Beckingham and Ghosh, 2017; Seidensticker et al., 2017; Li et al., 2018). Through these mechanisms a wide range of MP particles are released from WWTPs, including nanoplastics which may be ingested via skin diffusion during embryogenesis of fish cells, resulting in mortality of marine organisms (Enfrin et al., 2020a).

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

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

The primary objective of this study was to assess the role of sewage-related debris (flushable, non-flushable and sanitary

towels) as a source of white MP fibres in the marine environment (urban and rural). The material composition of a variety of white fibres obtained from intertidal sediments and a recurring washed-up deposit of true-to-form sanitary waste, likely associated with a combined sewage overflow on a beach in close proximity to a WWTP, were characterised. The results were compared with a variety of consumer sanitary products in order to investigate their role as a source of white MP fibres in the marine environment.

2. Methodology

2.1. Sample sites

The study area covers three locations along the mid-western Irish seaboard, on Europe's north-western, Atlantic-facing margin (Fig. 1). Weak tidal currents predominate across the near-coastal continental shelf, along which the 'Irish Coastal Current' flows northward (Fernand et al., 2006). The strong oceanic influence induces a temperate maritime climate with average annual precipitation values in excess of 1000 mm, characterized by frequent moderate winter storms. The region is marked by a strong overland and groundwater fluvial system and relatively high energy, sand to coarse sediment blanketed coastlines. Exceptions occur at the heads of the many coastal embayments, where finer grained sediment may accumulate in intertidal wedges dominated by sandy silt sized fractions.

Galway Bay, a 50 km long by 34 km wide west-facing embayment, is partially enclosed and protected from strong shelf currents and large oceanic swells by the Aran Islands topographic barrier. Oceanic flows enter the bay via the South Sound and circulate anticlockwise before exiting the Bay through the North Sound (Fig. 1). A major source of freshwater input occurs at the northeast corner of the bay, where the River Corrib, catchment size 3136.08 km², enters at the port of Galway City. The Corrib freshwater plume moves west along the east of the tidal gyre, except when strong south west winds may drive the plume to the shallower inner bay to the east (Nolan, 1997). The main WWTP for Galway City is located at Mutton Island (Fig. 1 and Fig. S1).

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

Two other estuaries were chosen as control sites for this work: Bell Harbour 16 km to the south-south-west and within Galway Bay, and Bellacragher, 100 km to the north. Bell Harbour, Co. Clare (Fig. S1c) (53°07' 20.51 N, 09°04' 23.08 W) is a sheltered tidal estuary (tidal range 4.5 m) on the south coast of Galway Bay. Inland and surrounding the site, the karst limestone plateau of the Burren area has a sparse rural population. The local catchment feeding into the estuary covers 56 km², with freshwater emerging into the centre of the estuary via a submarine outlet (McCormack et al., 2017). The sample site is located ~5 km from the wastewater outlet at Ballyvaughan and ~9 km from the outlet at Kinvara. Bellacragher (Fig. S1d) (53°57' 56.75 N, 09°49' 40.55 W) is located at the head of an enclosed tidal estuary (range 3.5 m) ~100 km north of Galway Bay adjacent to Ballycroy National Park. This control site

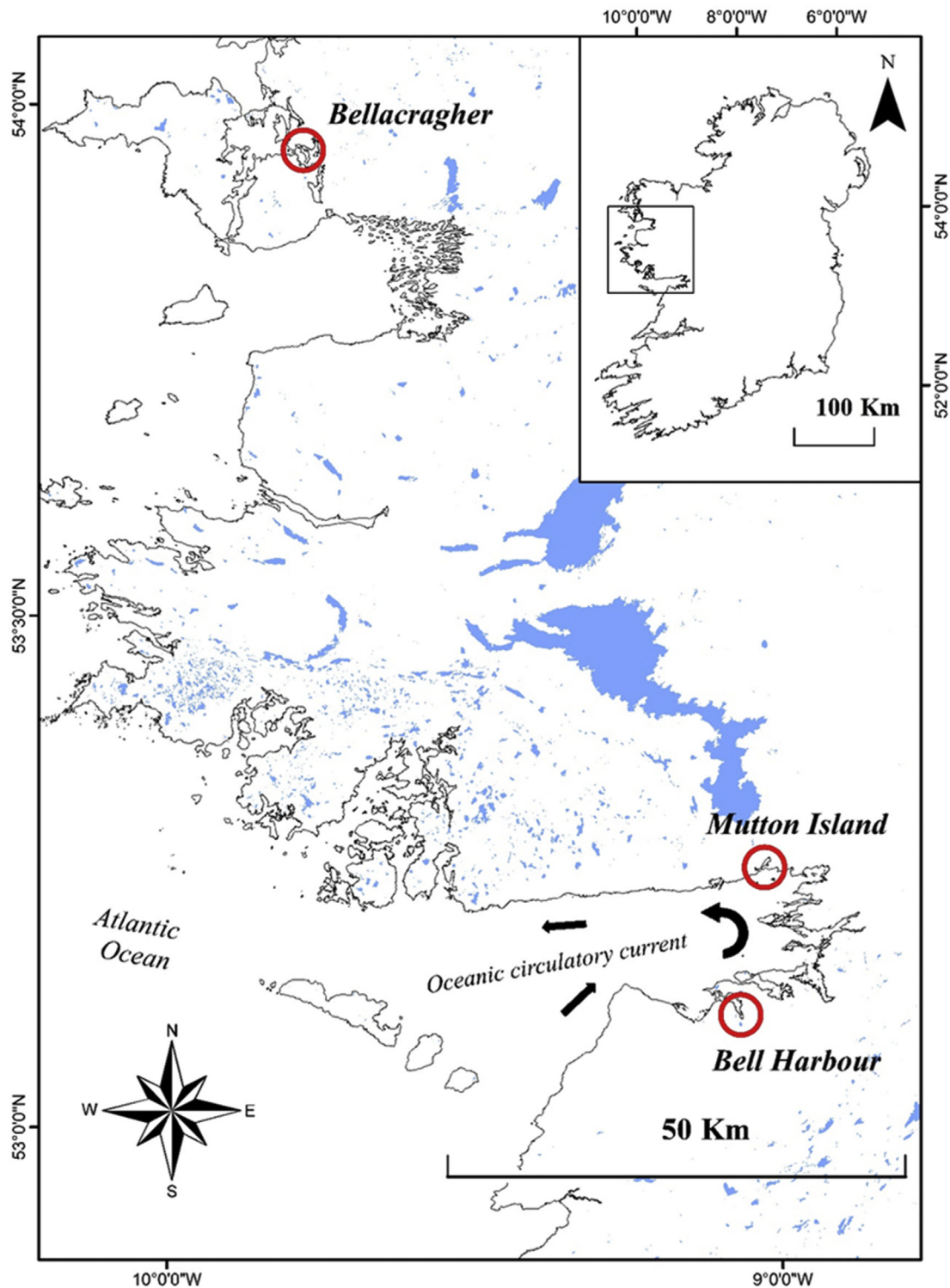


Fig. 1. Map showing locations of sampling sites (Bellacragher, Mutton Island and Bell Harbour) on the west coast of Ireland. Arrows illustrate oceanic circulatory current in Galway Bay. Image: OSi Licence number NUIG220212.

represents a sparse rural population in surrounding areas, far from urban influence, where wastewater treatment in the region is exclusively by one-off or small group septic tanks, without connection to a sewage treatment network. No direct riverine freshwater drainage adjacent to the sample site embayment is evident.

2.2. Microplastic sediment sampling

Polymethyl methacrylate (Perspex) core tubes (internal diameter 6.5 cm) were used to extract (15 cm) replicate intertidal sediment core samples from each site, on four sampling occasions, during the Spring low tides of September 2017, December 2017,

March 2018 and June 2018. Prior to field sampling, core tubes and tools were thoroughly rinsed under high pressure tap water, further rinsed with distilled water and stored in clean paper bags. Immediately prior to sampling, core tubes and tools were further rinsed with distilled water.

Coring locations were spaced 2–3 m apart along the midline of the intertidal zone by selective random sampling (Crawford and Quinn, 2017). Core tubes were manually forced into the sediments and then dug out during extraction, to ensure minimal disturbance of the sample. Tubes were immediately capped and sealed at both ends and labelled. Samples were stored upright in darkness at room temperature prior to processing.

2.3. Particle size analysis (PSA)

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

2.4. Microplastic extraction

The identification and extraction of MP particles and fibres from the upper 2.5 cm of the core was undertaken by visual examination on the 2 mm to 500 µm sediment fractions using an Olympus S251 (Tokyo, Japan) binocular microscope, at up to 40 X magnification. Plastic detritus within sieve fractions <500 µm were identified but not included in this study due to the difficulty of accurate visual identification and handling (Hidalgo-Ruz et al., 2012). Petri dishes were placed above a black background to enhance visual differentiation of white particles. Microplastics were visually identified by a set of selection criteria: no organic or cellular structures evident, except where conjoined to probable MPs; fibres are consistent in diameter with no tapering or branching; consistency of particle colour and texture; fibres remain malleable in response to manipulation. Coloured potential MP fibres and particles were extracted manually using fine tweezers and adhered to double sided tape on glass slides. White/translucent fibres were transferred into an Eppendorf tapered centrifuge tube to maintain visual clarity. Clusters frequently contained multitudes of individual fibres but were recorded as one MP find. As quality control on validity, all potential MPs were checked once again by visual examination. When biofouling hindered spectral analysis of the MP, 2 mL of hydrogen peroxide (H₂O₂) was added to the samples inside an eppendorf and left overnight. Samples were washed with Milli-Q™ water and left to dry prior to analysis.

2.5. Sampling of sewage-related debris

Post sediment sampling (Autumn 2019), a large quantity of sewage-related debris was washed-up on the shoreline at the mouth of the River Corrib in close proximity to the sediment sampling site at Mutton Island (Fig. S1a). This re-occurring accumulation consisted of a range of largely intact wipes and sanitary towels (and other personal care products) mixed and entangled in seaweed biomass (Fig. 2a–h). Samples of this material were

collected for fibre extraction and polymer identification on two different occasions.

2.6. Commercially available sanitary consumer products

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

2.7. Quality control

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

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

2.8. Raman Spectroscopy

Random samples of white fibrous field samples (n = 100) were analyzed for polymer identification by Raman Spectroscopy: 28.7% (n = 86) of total Mutton Island white/translucent fibres, 31.8% (n = 7) from Bell Harbour white fibres, and 30.4% (n = 7) from Bellacragher white fibres. Commercially available sanitary textile products (n = 17) were analyzed. Analysis was undertaken by a Raman spectrometer (Horiba LabRAM II, Horiba Jobin-Yvon, France) equipped with a 600 groove mm⁻¹ diffraction grating, a confocal optical system, a Peltier-cooled CCD detector, and an Olympus BX41 microscope (Kostrzytsia et al., 2018). The instrument was calibrated by zero-order correction on a known band of 1/520 cm etched on a crystalline silicon wafer (Jonker et al., 2015). Analysis was mostly conducted with a 532 nm laser; one highly reflective sample was identified using a 785 nm laser. The measurements were performed with acquisition times of 4–30 s over a spectral range of 100–3500 cm⁻¹. For identification purposes, the obtained spectra for each sample were compared to a spectral reference library (KnowItAll, Bio-Rad) and an in-house extension of the library with additional spectra from environmental MP collected from the intertidal zone and two clothing garments manufactured from polyester (PES) (Table 1). Replicate spectra were recorded for each sample.



Fig. 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).

2.9. Scanning electron microscopy (SEM)

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

3. Results

3.1. Wet wipe and sanitary towel polymer identification

The most common polymer used in the manufacture of

commercially available wet wipes was identified as non-woven PET fibres (Table 1). Five consumer wipe products were identified as PET and cellulose and two were identified as containing only PET; however, the main component of one wipe brand was identified as polypropylene (PP) (Table 1). Out of the eleven commercially available wipe products, four brands were labelled as flushable (Table 1) and two of these flushable wipes were observed as manufactured from a mix of PET and cellulose (Fig. 3). The remaining seven wipes were marked as non-flushable: three comprised a mix of PET and cellulose, one cellulose alone, two PET alone and one PP alone (Table 1).

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

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

Code	Description	Non-flushable	Flushable	Raman ID
Commercially available sanitary products				
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
Sewage-derived samples				
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

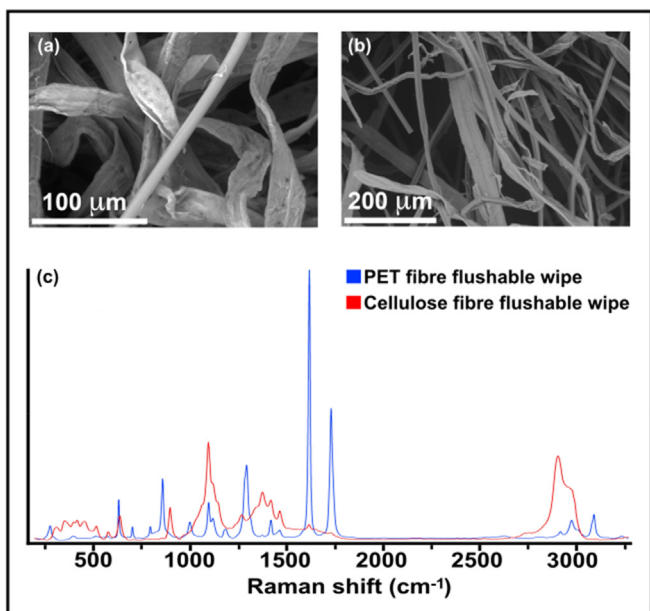


Fig. 3. SEM images of commercially available flushable wipes (a, b) with (a) identified as a combination of PET & cellulose and (b) cellulose alone using Raman Spectroscopy (c).

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

3.2. MP occurrence in intertidal sediment samples

Intertidal surficial sediments at the Mutton Island mid-tide line comprise a moderately sorted fine sand to silt, becoming well

sorted in December 2017. Bell Harbour sediments are very well sorted, indicative of silt and clay; gravel outliers evident in the summer season were largely absent in December 2017 and March 2018. Bellacragher is predominately a poorly sorted coarse medium to fine sand, becoming moderately sorted in December 2017. Microplastics were recorded in all the sediment samples, a total of 433 MPs were recorded (Tables S1–S3), of which 78% occurred at Mutton Island, 15.2% at Bell Harbour and 6.7% at Bellacragher.

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

Bell Harbour displayed a similar MP loading over the first two sampling occasions with a decrease in June 2018. In contrast, the highest MP loadings at Bellacragher occurred in June 2018 (Table 2). However, the range of MP loadings at both these rural sites were

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

Location	Month	Total Microplastic (kg^{-1})	Total White Fibres (kg^{-1})
Mutton Island	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)
Bell Harbour	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)
Bellacragher	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)

well below the levels adjacent to the WWTP at Mutton Island (Table 2).

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

While a total of 393 white fibres kg^{-1} dw were observed at Bell Harbour (Table 2), in contrast, a total of 157 MP kg^{-1} dw (white and non-white) were observed in the Bellacragher samples (Table 2).

3.3. Intertidal white microfibre polymer identification

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

3.4. Spectral analyses of white microfibres from intertidal sediments, sewage-derived waste and commercially available sanitary products

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

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

4. Discussion

The sample sites chosen for investigation in this study provide a representation of modern urban and rural European coastal environments. It is well established that WWTP effluent contributes to the MP loading of nearby shoreline sediments (Browne et al., 2011; Estahbanati and Fahrenfeld, 2016; Talvitie et al., 2015; Ziajahromi et al., 2017). Microplastic abundance and frequency at Bellacragher was consistently low with no obvious point sources. Relatively high densities of ABS fibres in June 2018 were likely an anomaly, which resulted from the presence of ABS piping and fittings in the vicinity of the sampling site. Acrylonitrile butadiene styrene is widely used in the manufacture of piping fittings due to its strength and resistance to abrasion, and the fibres at Bellacragher may be long form fragments of white plastic panelling,

which was cut for pontoon construction locally. The presence of ABS in this study is no surprise as it has been reported to be present in WWTPs, beach and lake sediments worldwide due to anthropogenic activities and dispersion to the marine environment through the sewage and freshwater systems (Hamid et al., 2018; Kang et al., 2018; Kazour et al., 2019). Bell Harbour, on the southern margins of Galway Bay, which exceeded the loading at Bellacragher, was still deemed to have a relatively low MP loading compared with the main sampling site at Mutton Island.

The Mutton Island sampling site is in an urban setting adjacent to a WWTP and is in close proximity to the outflow of the Corrib River, which adds a freshwater-derived flux to the sample area. The variability in MP loading over the 10-month period of this study was likely due to local hydrodynamics, deposition from currents, and sediment turnover in response to seasonal climatic variability as well as proximate human activity. The nearby intertidal zone is prone to the accumulation of high volumes of washed-up sewage-derived debris on a frequent basis. Excessive MP loading in December 2017 was likely induced by heavy precipitation episodes during a south-westerly storm front. Elevated debris loading on this occasion may result from combined sewer overflows, where excessive input of drainage water exceeds WWTP effluent capacity and is released untreated in the overflow. Combined sewer overflows from overloading have been reported from many other cities globally (Ellis, 2006; Gasperi et al., 2008; Kazour et al., 2019; Rathnayake and Faisal Anwar, 2019; Scoullios et al., 2020; Weyrauch et al., 2010). These overflows in turn can give rise to washed-up sewage-derived waste and debris on the surrounding coast (Krelling and Turra, 2019; MCS, 2019; Rangel-Buitrago et al., 2020). It is estimated that the quantities of wet-wipes washing up on the coastline in the UK increased by 94% since 2017 and 400% during the last decade (MCS, 2019; Pantoja-Munoz et al., 2018; The Guardian, 2015).

Blumenröder et al. (2017) report high MP loadings in intertidal sediments (6500 MPs per kg^{-1} dw) adjacent to a WWTP in Scotland (Stromness, Orkney Islands). The sampling regime, as described in the Orkney Islands, was comparable to the field sampling methodology in the current study, i.e. intertidal sediments collected from relatively sheltered depositional coastal embayments; 6083 MPs per kg^{-1} dw are reported for Mutton Island which serves a population some 40 times that of Stromness. This may be a function of the experimental design, as the current study extracted MPs from the 2 mm, 1 mm, and 500 μm sieved fractions, and therefore MPs identified in our study are likely an underestimation as the greatest numbers of MPs occur in finer grained sediments (Corcoran et al., 2020; Mintenig et al., 2017).

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

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

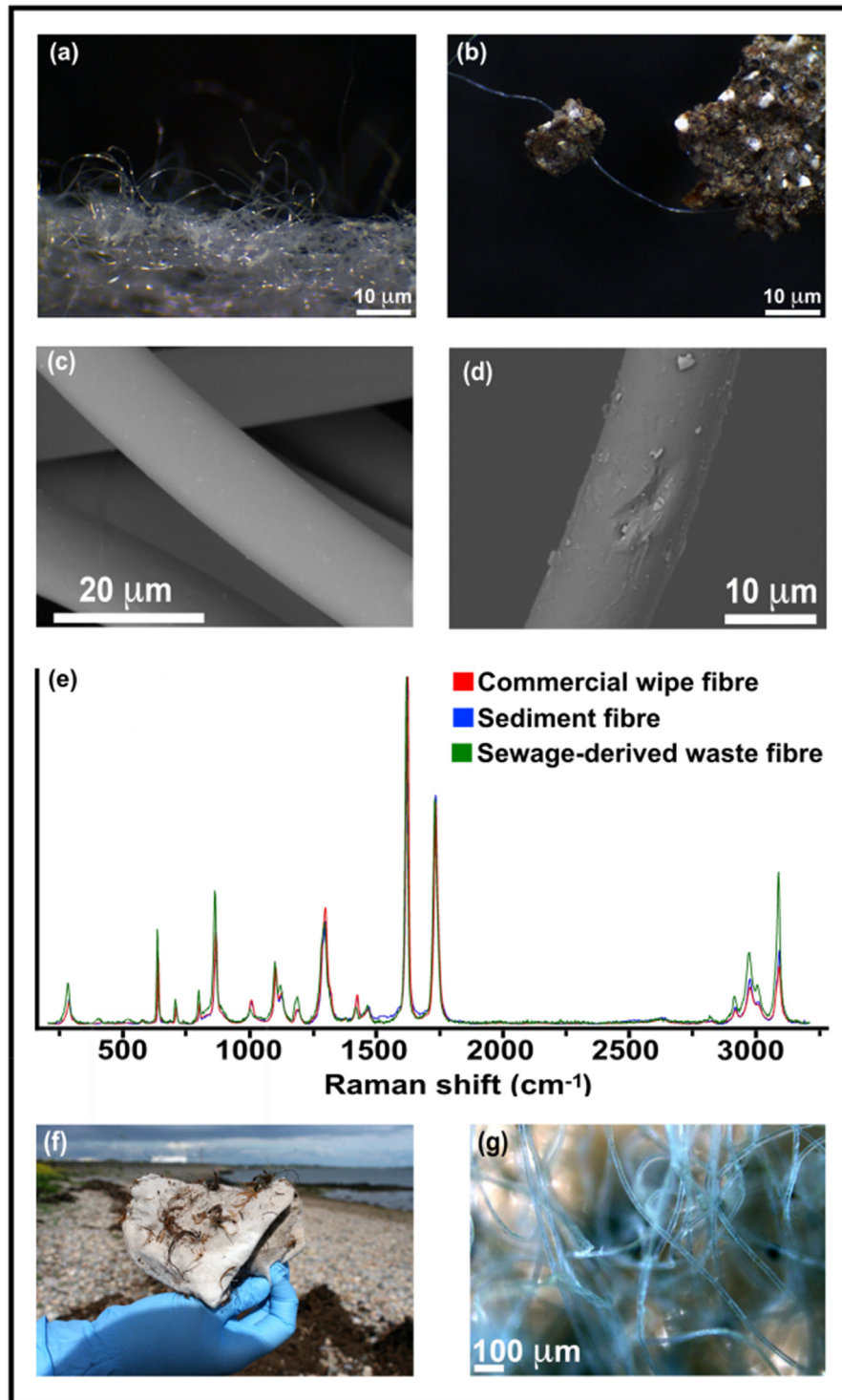


Fig. 4. Images and Raman spectra of PET white fibres extracted from commercially available wipes and intertidal sediments; a) microscope image of commercial wipe MicroFibre (MF), b) microscope image of MF extracted from the intertidal sediments, c) scanning electron microscope (SEM) image of a commercially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

from the intertidal zone near the WWTP following a heavy rainfall event were mostly comprised of PET, with only a quarter of the samples analyzed presenting as a mix of PET and cellulose, and over 80% of the wipes in the shoreline waste were identified as non-flushable due to their polymer composition following the International Water Services Flushability Group and non-woven textile

industry guidelines (INDA/EDANA, 2018; IWSFG, 2018). The flushable wipes market has increased significantly in value and is currently estimated to have reached \$2.7 billion (Atasağun and Bhat, 2019), with the entire global personal care wipes market valued at \$15.8 billion in 2018 (Grand View Research, 2019). Consequently, this rapid growth globally has resulted in an increase

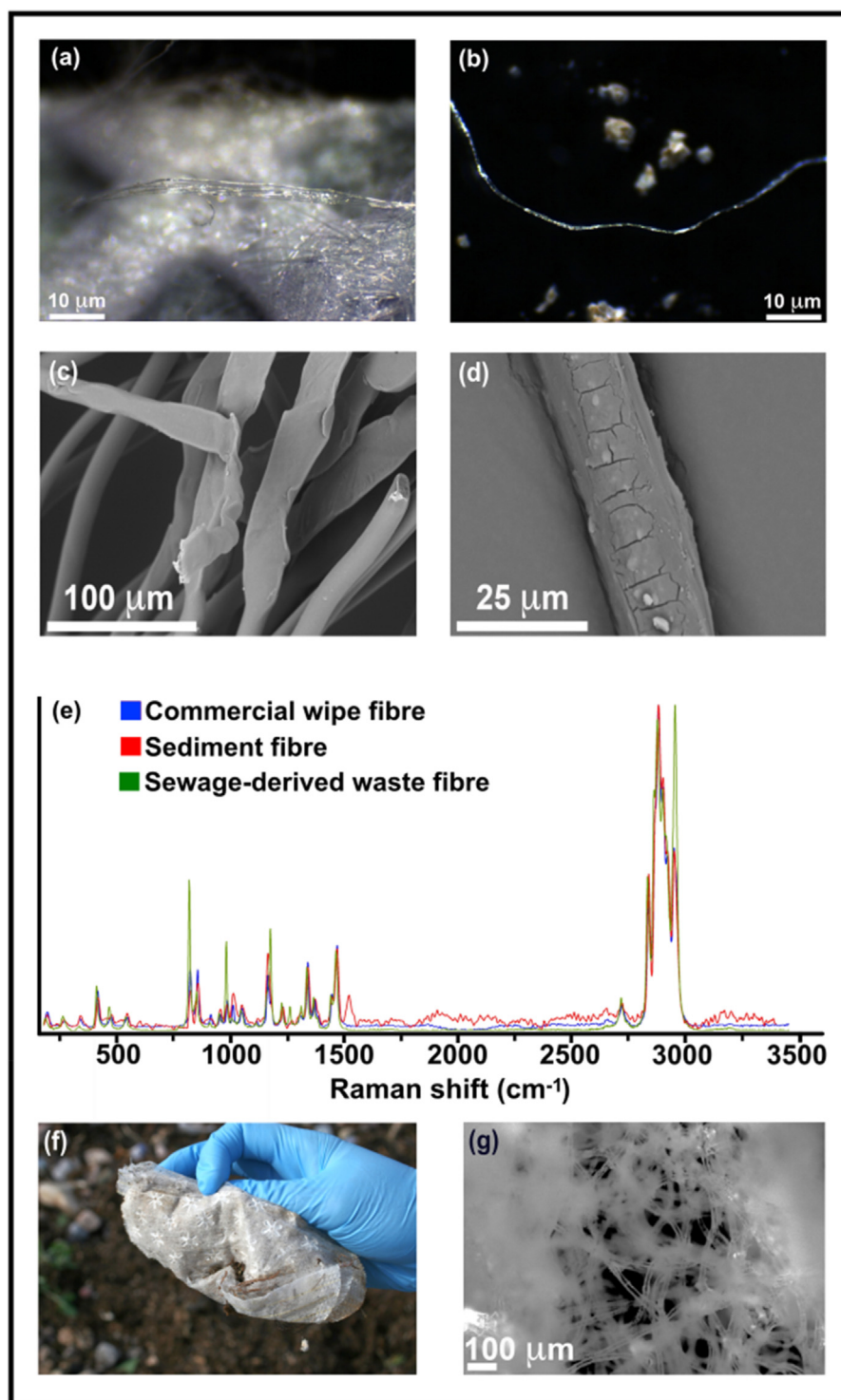


Fig. 5. Images and Raman spectra of PP white fibres extracted from commercially available sanitary towels and intertidal sediments; a) microscope image of a commercially available sanitary towel MicroFibre (MF), b) microscope image of MF extracted from the intertidal sediments, c) scanning electron microscope (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.

in the quantity of wipes found on the UK coastline, a 400% increase in the last decade (MCS, 2016; MCS, 2019). This is a phenomenon likely occurring globally in concurrence with combined sewage overflows. Sanitary towels contributed significantly to the washed-up sewage-related debris in the present study along with wipes and both may fragment into white MP fibres, which potentially pose a

hazard to marine organisms.

The analyses of commercially available flushable and non-flushable wipes revealed that over 90% contained either cellulose, a combination of PET and cellulose, or PET alone in their matrix, which was also reported in other studies investigating fibre composition of wipes (Durkan and Karadagli, 2019; Pantoja-Munoz

et al., 2018; Khan et al., 2019). Microscopic and spectroscopic analysis of flushable and non-flushable wipes in this study revealed a similar structure/construction and fibre composition. It was microscopically possible to identify and separately analyse cellulose and MP fibres (Figs. 3a and 5c). Cellulose fibres presented a more rough and ribbon-like appearance than PET, PE and PP, which presented a more uniform and three-dimensional shape, although this is not always the case (Dyachenko et al., 2017; Lares et al., 2018; Norén, 2007). The presence of white MP fibres in the wipes reinforces their strength and renders them more durable than wipes manufactured from cellulose alone (MESUGC, 2019).

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

Manual manipulation of the wipes in the current study also highlighted that wipes manufactured from cellulose alone were more easily torn, fragmented and disintegrated when submerged and agitated in water following the guidelines of the International Water Services Flushability Group (IWSFG PAS1, 2018). By contrast, wipes made of PET or PET/cellulose combinations were more difficult to tear apart and less susceptible to disintegration when submerged and agitated in water, rendering them unsuitable to be flushed down toilets (INDA/EDANA, 2018; IWSFG PAS2, 2018; Khan et al., 2019). Cellulose degradation under aerobic and anaerobic conditions is well-documented (Demain et al., 2005; Leschine, 1995; McDonald et al., 2012; Smith, 1994; Song et al., 2013), unlike PES and PET fibres which are not subject to bacterial breakdown in the environment and in WWTPs (Zambrano et al., 2020). Although flushable wipes comprising cellulose fibres alone degrade in the wastewater system, the current study has shown that more than 50% of wipes labelled as flushable contain PET and hence are non-degradable and produce MP fibres that are available to aquatic organisms when released to the marine environment.

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

The presence of PET and PET/cellulose wipes among sewage-related waste washed-up on the beach are indicative of the inappropriate disposal of non-flushable wipes. This would suggest that

some consumers are unlikely to follow packaging disposal instructions and flush these products indiscriminately. On the other hand, the inappropriate disposal of flushable wipes may be more a function of incorrect labelling resulting in a lack of awareness by consumers.

Two billion single-use menstrual products are flushed down the toilet each year and tampons, pads and applicators generate 200,000 tonnes of waste per annum in the UK alone (Wen, 2020). Furthermore, manufacturers of female hygiene products fail to adequately reveal the precise composition of sanitary towels (which are 90% plastic) as part of the labelling (Wen, 2020). Although the market for reusable and biodegradable female products is growing, disposable towels are still the most common product used due to convenience and costings, their presence in WWTPs and beaches should be of concern not only for protection of wastewater systems but also for environmental and public health hazards.

5. Conclusion

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

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.watres.2020.116021>.

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