Characterisation, quantity and sorptive properties of microplastics extracted from cosmetics

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A B S T R A C T

Cosmetic products, such as facial scrubs, have been identified as potentially important primary sources of microplastics to the marine environment. This study characterises, quantifies and then investigates the sorptive properties of plastic microbeads that are used as exfoliants in cosmetics. Polyethylene microbeads were extracted from several products, and shown to have a wide size range (mean diameters between 164 and 327 μm). We estimated that between 4594 and 94,500 microbeads could be released in a single use. To examine the potential for microbeads to accumulate and transport chemicals they were exposed to a binary mixture of ^{3}H-phenanthrene and ^{14}C-DDT in seawater. The potential for transport of sorbed chemicals by microbeads was broadly similar to that of polythene (PE) particles used in previous sorption studies. In conclusion, cosmetic exfoliants are a potentially important, yet preventable source of microplastic contamination in the marine environment.

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1. Introduction

Plastics provide a diverse range of inexpensive, lightweight, strong, durable and corrosion-resistant products (Thompson et al., 2009b). The success of plastics as materials has been substantial and they are used in a wide range of applications. This versatility, together with their low cost, has resulted in the annual worldwide production of around 300 million tonnes (Plastics Europe, 2014). Approximately 50% of production is used to make packaging, much of which is used in disposable applications. This creates a major waste management problem, with plastics accounting for approximately 8–10% of all the waste generated in the UK (Barnes et al., 2009; Hopewell et al., 2009).

Around 700 species of marine organism have been reported to encounter marine debris in the natural environment, with plastic debris accounting for over 90% of these encounters (Gall and Thompson, 2015). Large plastic items, such as discarded fishing rope and nets, can cause entanglement of invertebrates, birds, mammals, and turtles (Carr, 1987; Eerkes-Medrano et al., 2015; Fowler, 1987; Laist, 1997) but the marine environment is also contaminated with much smaller microplastics particles (defined by NOAA as <5 mm). These have been reported at the sea surface (Law and Thompson, 2014), on shorelines (Claessens et al., 2011), and on the sea bed (Van Cauwenberghe et al., 2013). The sources of microplastics include fragmentation of larger items (secondary sources), and direct inputs of microplastic sized particles, such as microbeads used in cosmetics and pre-production pellets (primary sources). It is important to understand the relative importance of these sources as well as the size and abundance of microplastic particles released, since this will influence encounter rate and availability to biota (Teuten et al., 2007; Thompson et al., 2009a; Cole et al., 2011).

There is growing evidence that the amount of microplastics in marine waters is increasing, with unknown ecotoxicological consequences (Goldstein et al., 2012). Fendall and Sewell (2009) reported on microbeads used as "scrubbers" in cosmetics products, which they described as being up to 500 μm in diameter, being released into the natural environment and potentially available to organisms. Ingestion of microplastics, has been reported for a wide range of marine organisms including deposit and suspension feeders (Browne et al., 2008; Graham and Thompson, 2009), crustaceans (Murray and Cowie, 2011), fish (Boerger et al., 2010), marine mammals (Denuncio et al., 2011), and seabirds (Avery-Gomm et al., 2012; Van Franeker et al., 2011). However, the extent, if any, to which chemicals sorbed onto, or incorporated into plastics can desorb from plastic particles, and transfer to the tissues of marine organisms is less clear. Recent experimental trials provide evidence for the role of plastics in the transfer of chemicals with

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subsequent adverse physiological effects (Besseling et al., 2013; Rochman et al., 2013), but studies based on bioaccumulation models concluded that the transfer of contaminants from plastics to marine organisms upon ingestion is of limited importance compared to other pathways (Gouin et al., 2011; Koelmans et al., 2013).

Microplastics have been used to replace natural exfoliating materials (for example, pumice, oatmeal, apricot or walnut husks) in cosmetics and have been reported in a variety of products such as hand-cleansers, soaps, toothpaste, shaving foam, bubble bath, sunscreen, shampoo and facial scrubs (Fendall and Sewell, 2009; Gregory, 1996; Zitko and Hanlon, 1991; UNEP, 2015).

Industry uses the terms 'microbeads' to describe microplastic particles present as ingredients in personal care and cosmetic products; they may also be called microspheres, nanospheres, plastic particulates (UNEP, 2015). Around 93% of the 'microbeads' used in cosmetics are polyethylene (PE), but they can also be made of polypropylene (PP), PE terephthalate (PET), polymethyl methacrylate (PMMA) and nylon (Gouin et al., 2015; Eriksen et al., 2013; UNEP, 2015). Microbeads are likely to be transported to wastewater treatment plants, where some will be captured in oxidation ponds or sewage sludge. However, due to their small size, it is anticipated that a substantial proportion will pass through filtration systems and enter aquatic environments (Fendall and Sewell, 2009).

Leslie et al. (2013), examined wastewater treatment plants that discharge into the North Sea, the Oude Maas River or the North Sea Canal and reported that the treated effluent contained on average 52 pieces of microplastics/L. Eriksen et al. (2013) also reported substantial amounts of multi-coloured microplastic spheres in surface waters of the Laurentian Great Lakes of the United States which were suspected to originate from consumer products. This provides evidence that microplastics are not all captured in sewage sludge of wastewater treatment plants and is of broad concern, since treated effluent from sewage disposal sites is discharged into a range of water bodies, including into inland waters, estuaries and the sea (DEFRA, 2002).

Gouin et al. (2011) estimated that the per capita consumption of microplastic used in personal care products for the U.S. population, based on the usage of PE microplastic beads used in personal care products, was approximately 2.4 mg per person^{-1} per d^{-1}, indicating that the U.S. population may be emitting an estimated 263 tonnes per yr^{-1} of PE microplastic (Gouin et al., 2011). To set this into perspective, in terms of its contribution to marine litter, this annual quantity is approximately equivalent to 25% of the total mass of plastic that is estimated to have accumulated in the North Atlantic Subtropical Gyre (Law et al., 2010; Gouin et al., 2011).

Facial scrubs are one type of cosmetic which contains microplastics as exfoliating agents. Due to this, such products could contribute microplastics contamination to the marine environment. Despite concerns about the potential for products containing microbeads to represent a major source of microplastics to the environment, only one study has measured microplastics in facial scrubs (Fendall and Sewell, 2009), and there are no peer reviewed publications confirming the type or quantity of microplastic polymers used in facial scrubs. Here we examined six brands of facial scrubs manufactured by three companies and describe the microplastics (plastic microbeads) present, in terms of polymer type, colour, size, weight and abundance. We also investigated the sorptive properties of the microplastics in relation to the potential for transport of the POPs phenanthrene (Phe) and dichlorodiphenyltrichloroethane (DDT) and compared them with commercially available PE particles previously used in adsorption/desorption studies of persistent organic pollutants (POPs) (Bakir et al., 2012, 2014a,b; Teuten et al., 2007).

2. Methods

2.1. Sample preparation

Six major brands of facial scrubs were chosen, based on their prevalence in major supermarkets close to Plymouth UK. All of the products listed in their ingredients that they contained PE. Four replicates of each product were purchased, with each replicate sourced from a different supermarket to provide a representative sample.

Since the specific brand names of the products are not of particular relevance, they were labelled A–F.

Each facial scrub was a viscous liquid (A–D contained 150 mL of product, E contained 125 mL). The contents were subjected to vacuum filtration to obtain the plastic particles. The procedure required mixing each product in approximately 1 L of boiling water, followed by vacuum filtration over Whatman N°4 filter paper, then drying at 30 °C to constant weight. Once dry, the particles were weighed by Precisa 2200C weighing scales and the residues were transferred into separate glass vials. A Kruskal–Wallis test was performed on the data, using R studio, to test whether the amount of microplastics per unit volume extracted differed between products ($p < 0.05$). This was followed by a *post-hoc* Nemenyi-Test to find which specific products significantly differed.

2.2. Visualisation and identification

Microplastics from each product were identified using Fourier transform infra-red spectroscopy (FTIR), using a Hyperion 1000 microscope (Bruker) coupled to an IFS 66 spectrometer (Bruker). The spectra obtained were compared to a spectral database of synthetic polymers (Bruker I26933 Synthetic fibres ATR library).

Some non-plastic residues were extracted and separated from the plastic particles using Endecotts woven wire sieves of varying mesh size. The mass of plastic particles was recorded.

A Malvern Mastersizer 2000 laser particle sizer (MM2) was used to measure the size-frequency distributions (SFDs) of the extracted plastic into sixty-eight different sized bands with logarithmic spacing (range 0.015–2000 µm; Woolfe and Michibayashi, 1995). The resultant particle size distributions were expressed as a volume weighted mean from an average of twenty five measurements per product. The mean for each product was then calculated.

The number of plastic particles in each product, N, was estimated, assuming the particles were of spherical shape, using the following equations:

$$Vt = \frac{Mt}{D} \tag{i}$$

$$V \text{ (avg particle)} = \frac{4}{3}\pi r^3 \tag{ii}$$

$$N = \frac{Vt}{V \text{ (avg particle)}} \tag{iii}$$

where Vt is the total volume of plastic extracted, Mt is the total mass of plastic extracted, D is the density, $V(avg.p)$ is the mean volume of one particle, N is number of particles, and r is the radius.

For each product: Eq. (i) allowed calculation of the total volume of microplastic extracted; Eq. (ii) allowed calculation of the average volume of a microplastic particle from each product; by dividing the total volume of microplastic by the average volume of a microplastic particle, Eq. (iii) allowed calculation of the approximate number of particles in each product. Particles were then

visualised by scanning electron microscopy (JEOL, 7001F), imaging to describe both whole particles and their topography.

2.3. Sorption of pollutants to plastics

As part of a separate, but related study, microbead exfoliants were extracted from shower gel and used to examine the adsorption of POPs by microbeads. The microbeads from the shower gel products were extracted and identified by FTIR following the same methods in Sections 2.1 and 2.2. As these microbeads were extracted from different brands of exfoliant products, they are labelled X, Y & Z. These microbeads were exposed to Phe and DTT; the results were then compared with sorption to ultra-high-molecular-weight (UHMW) PE particles used in a previous sorption study (Bakir et al., 2014a,b; Bakir et al., 2012).

Adsorption experiments were conducted in an ISO9001 accredited radioisotope facility at the Plymouth University. ^{3}H-Phe and ^{14}C-DDT were selected as contaminants in this study to allow simultaneous quantification and to compare with past studies (Bakir et al., 2012). 10 mg of either UHMW PE or the extracted microbeads were placed into three glass centrifuge tubes (50 mL) and 5 μL of ^{14}C-DDT and 16 μL of ^{3}H-Phe were added to the walls of the tubes. The solvent was allowed to evaporate and 25 mL of seawater (35 psu, 59.3 ± 0.26 mS) was added and the tubes were equilibrated for 48 h (Bakir et al., 2014a) in the dark at 18 °C under continuous horizontal, rotary agitation at 220 rpm. All experiments were carried out in triplicate. The concentration of contaminant was determined in the aqueous and solid phase by counting the β decay from the ^{14}C-contaminant by liquid scintillation counting (LSC) as outlined in Bakir et al. (2012). The amount of contaminant in each phase was quantified using a calibration curve prepared by counting known amounts of the contaminant.

The single point distribution coefficient, single point K_d, was calculated using the equation:

$$K_d = [q_e]_{solid}/[C_e]_{aq} \qquad (iv)$$

where q_e is the amount of contaminant adsorbed onto plastic (μg kg^{-1}) at equilibrium and C_e is the contaminant concentration in the aqueous phase at equilibrium (μg L^{-1}).

2.4. Statistical analysis

A two-factor ANOVA, with contaminants and the microbead type considered as fixed factors, was used to characterise any significant differences ($p < 0.05$) between the distribution coefficients calculated from the sorption of Phe and DDT onto microbeads. Cochran's test was used to ensure that the data fulfilled the pre-requisites for parametric analysis and the appropriate data were $\ln(x + 1)$ transformed. Student–Newman–Keuls (SNK) tests were then used to identify any significant terms. The tests were carried out using GMAV5 software (Underwood et al., 2002) and are presented in the Supplementary information.

3. Results

3.1. Extraction and identification

All of the products contained microplastic particles of PE, which was in agreement with their stated ingredients. Product C also contained green and yellow particles that were slightly larger than the PE microbeads. These could not be identified by FTIR using the Bruker spectral database and were removed from the samples via sieving and are not included in any of the calculations. The collected solids from product C also contained micro-'glitter'. These 'glitter' particles were small and could not be removed from the

filter paper for further analysis. However, 'glitter' is commonly manufactured from plastic, such as PE.

The weight of microplastic extracted varied significantly between products (Kruskal–Wallis test, $p = 0.0012$, Fig. 1); the products which were significantly different from each other were C and E ($p = 0.0009$); D and E ($p = 0.0463$) (*post hoc* Nemenyi-Test).

3.2. Size-frequency distributions

Microplastics from the facial scrubs showed polydispersed size ranges, each with logarithmic bimodal distributions (Fig. 2). Product B had the largest size range (10 μm to >2000 μm), whereas product A was the most homogenous, ranging from 8 μm to 56 μm, with the largest proportion of smaller particles. Size frequency by volume distributions were used to calculate the mean diameters for each product. Products D–F had similar volume-weighted mean diameters, which were 288.80 μm, 289.63 μm and 293.48 μm respectively. The particles in product B and C were larger, with mean diameters of 326.83 μm and 317.91 μm, while product A was much smaller with a mean diameter of 163.82 μm. The volume-weighted mean diameters were used to estimate the number of particles in each product. Since the absolute density of the extracted plastics was not known, we calculated estimates using a range of standard densities. For PE these were, high (0.959 g/cm^3), medium (0.940 g/cm^3) and low density (0.910 g/cm^3).

Particle diameter, rather than the average weight in each product, was found to have the greatest effect on abundance estimates. Product E had on average 11.47 g of PE in each bottle, with a mean particle size of 289.63 μm, resulting in an estimated 6423 particles per mL. Whereas product A had less PE by weight with, on average, 6.11 g in each bottle, but resulted in an estimate of 18,906 particles per mL because the mean size was smaller (163.82 μm); being the highest quantity in any of the products. Product C had the second largest PE particles (317.91 μm), but the lowest particle abundance, with only 919 particles per mL. This data implies that the products tested could each contain between 137,000 and 2,800,000 microparticles (Fig. 3). The
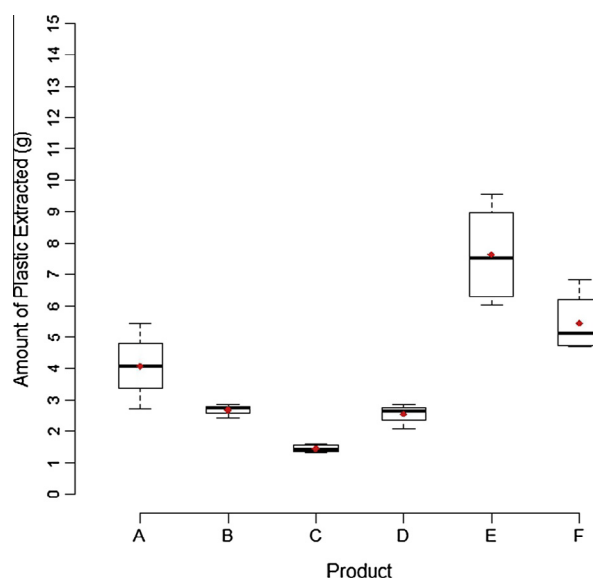

Fig. 1. Total mass of plastic microbeads extracted from six facial scrubs (A–F) per 100 mL. Diamond symbol indicates $\bar{x}$ ($n = 4$). The tails show both the maximum and minimum mass obtained, and the box represents the upper and lower quartiles. There were significant differences between the amount of microplastic in each of the products ($p < 0.05$).
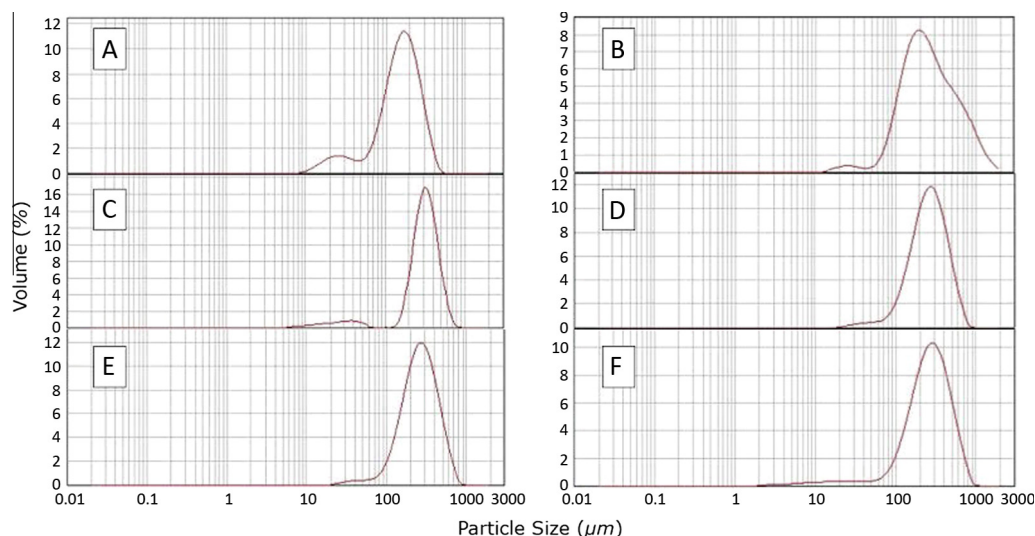
Fig. 2. Particle size distribution of PE microbead particles extracted from six facial scrubs (A–F). Determined using a Malvern Mastersizer 2000, laser particle sizer.
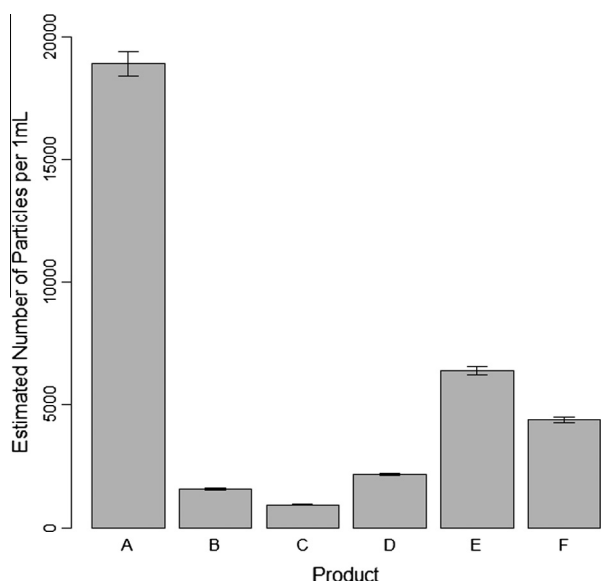

Fig. 3. Estimates for the number of PE microbead particles in six brands of facial scrubs per 1 mL. Calculated using data from the volume weighted mean ($n = 3$, ±SD; correlating to the spread of the different amounts of particles calculated for high, medium and low density PE).

quantity of particles was calculated using data for the volume mean diameter, however the size particle distribution had a tail of smaller particles, hence the particle abundances calculated are likely to be underestimates.

The shape and surface topography of the extracted microplastic particles was visualised by scanning electron microscopy. For all the brands, the extracted microplastics had a variety of shapes, including ellipses, ribbons, and threads, as well as irregular fragments (Fig. 4). An exception was product F, which in addition to irregular shaped pieces, also contained smooth, blue, PE spheres that were substantially larger than the rest of the particles, but represented a small proportion of the total amount of plastics present. Some of these spheres were fragmenting (Fig. 4).

The colour of microplastics used in the different products also varied (Table 1). All products contained white microplastics, but products A, D, E and F also contained coloured particles. The coloured microplastics in products D–F were larger than the white

plastics, but were less abundant. The white and pink microplastics in product A were of similar size to each other.

3.3. Sorption of persistent organic pollutants

Visualisation of microbeads extracted from products X, Y, and Z showed they could be differentiated between "smooth" and "rough" forms. This particle shape differentiation was also observed in products A–F, where A–E contained "smooth" particles and product F contained both "smooth" and "rough" forms (Fig. 4). Therefore, we considered sorption onto both morphologies. Results showed that microbeads extracted from the cosmetic products were able to sorb Phe and DDT from seawater (Fig. 5). Sorption capacity for all plastics was significantly higher for DDT compared to Phe ($p < 0.05$, Table 2). The "rough" microbeads were more efficient at adsorbing POPs from seawater than "smooth" ones, probably due to increased surface area. The "rough" microbeads were also more similar in shape, surface texture and sorptive property for POPs to PE particles used in previous experiments (e.g. Bakir et al., 2012, 2014a,b; Teuten et al., 2007). There were some significant differences between adsorption by microbeads and adsorption by PE particles and the direction of these effects was that microbeads from cosmetics tended to adsorb lower concentrations of POPs then PE particles. However, broadly speaking, it would appear that results from previous studies on transport of chemicals by sorption on to plastic are comparable with the transport potential on microbeads.

4. Discussion

Microplastics found within cosmetics such as facial scrubs, will routinely be washed into sewers as a direct consequence of consumer use. Due to their size, a considerable proportion is likely to pass through preliminary sewage treatment screens (typically coarse, >6 mm, and fine screens, 1.5–6 mm) (Water Environment Federation, 2003). Effluent containing the microplastics would then be discharged into inland waters, estuaries and the oceans. A recent study reported that treated effluent from three sample sites in the Netherlands contained on average 52 microplastic particles/L (Leslie et al., 2013). Microbeads used as exfoliants in facial scrubs are likely to be an important primary source of microplastics contamination, due to the quantity of plastic used in each product.
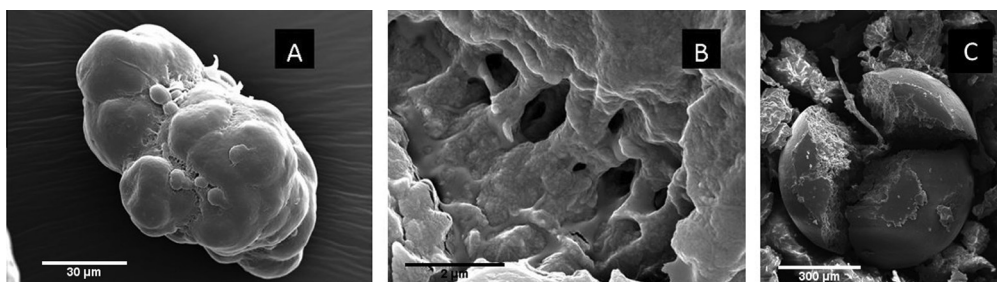
Fig. 4. (A) Scanning electron microscopy (SEM) of a typical rough facial scrub plastic microbead particle (9000× magnification). (B) SEM of surface microbead topography (16,000× magnification). (C) SEM of a broken smooth spherical plastic microbead from 'product F' (900× magnification).

Table 1
Colour of microplastics found within six facial scrub products.

Product	Colour of microplastic present
A	White and pink
B	White
C	White
D	White and light blue
E	White and dark blue
F	White and dark blue

When considering the potential consequences of the release of microbeads to the environment, if any, it is important to consider both the mass of plastic, and the number and size of the particles; the latter will have direct influence on the probability of encounters with wildlife.

The common application of facial scrub exfoliants is once per day, and it has been estimated that they are used by around 1.1 million women in the UK (Statista, 2013). Focussing on the products used in this study (A–F), and assuming that the typical daily amount used is 5 mL, between 4594 and 94,500 microplastic particles would have the potential to pass into the sewage system per use.

In terms of the mass of plastic entering the marine environment, previous work by Gouin et al. (2011) estimated that users in the U.S emit 2.4 mg of PE person^{-1} d^{-1}, amounting to an emission of 263 tonnes yr^{-1}. This estimate is calculated from data on liquid soap consumption, and assumes that only 15% of the

market is shared by companies that use microplastic beads in their liquid soaps. However, many brands do use exfoliating microbeads. Assuming that three out of four body exfoliants contain microplastics (Marine Conservation Society, 2012), and that an estimate that 25% of the microplastic is caught by the sewage system, the UK population could emit to the natural environment 40.5–215 mg of PE person^{-1} d^{-1}, or between 16 and 86 tonnes yr^{-1} (population of the UK in 2013: 64.1 million, (The World Bank, 2013) just from facial exfoliants. In order to set these quantities into context, by way of comparison, between 2009 and 2014 inclusive, in its annual weekend beach clean, MCS typically collect around 9 tonnes of litter per year (over an average length of 115 km of UK shoreline).

The presence of microplastics in sewage sludge has been reported previously by Browne et al. (2011), who found that former sewage disposal-sites on the seabed in UK waters contained more microplastics than non-disposal reference sites, highlighting the potential for microplastics to accumulate in aquatic habitats. The occurrence of microplastics within the marine environment is now well documented in the water column, at the sea surface and sediments (Law and Thompson, 2014). Microplastics also account for around 10% of all reports of ingestion of marine debris, highlighting their importance as a component of marine debris (Gall and Thompson, 2015). Their size makes them accessible to organisms with a range of feeding methods, including: filter feeders (mussels, barnacles), deposit feeders (lugworms) and detritivores (amphipods, sea cucumbers) and zooplankton (Wright et al., 2013a; Graham and Thompson, 2009; Thompson et al., 2009a,b; Browne et al., 2008). However, studies that quantify the
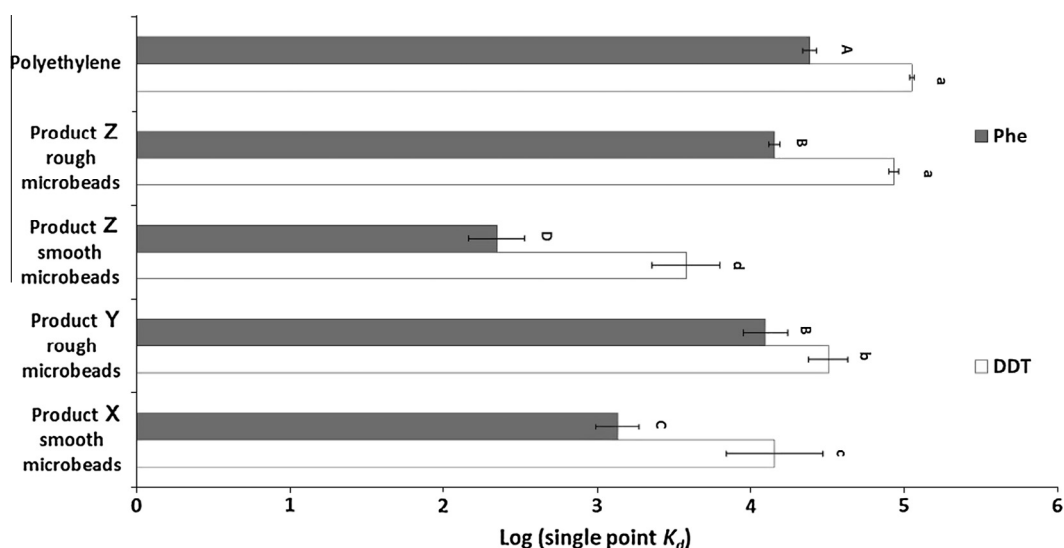

Fig. 5. Single point distribution coefficients (K_d) for the sorption of a mixture of phenanthrene (Phe) and DDT onto PE particles and rough and smooth PE-microbeads extracted from cosmetic products (n = 3, ± SD). For each contaminant, treatments with the same letters (A–C for Phe and a–d for DDT) were not significantly different (p < 0.05).

Table 2
Recovery (%) of phenanthrene (Phe) and DDT following sorption experiments onto PVC and PE (average values displayed, n = 3).

Particle type	POP	Aqueous phase	Glass wall	Solid phase	Total recovery
Product X beads	DDT	12	8	59	78
	Phe	43	1	24	68
Product Y particles	DDT	7	8	91	106
	Phe	13	3	65	81
Product Z beads	DDT	20	26	33	79
	Phe	64	2	6	73
Product Z particles	DDT	3	8	90	101
	Phe	11	5	60	75
UHMW PE	DDT	2	6	87	94
	Phe	7	2	80	89

abundance of microplastic predominately report elongated fibres. This may in part be due to the relative ease of detection of pieces with these shapes, since they differ from many natural particles found in sediments. Hence, the prevalence of microplastics with non-fibrous shapes (Fig. 4), for example microbeads from facial scrubs, may be under-reported in environmental sampling (Desforges et al., 2014; Lusher et al., 2014; Gallagher et al., 2015).

There is no way of effectively removing microplastic contamination once it is in the environment. The materials are too dispersed, the scale is too vast, ecological damage would be caused by any remediation (tiny organisms would likely be removed along with the microplastics), and the costs would be extremely high (UNEP, 2015). Since plastic is highly resistant to degradation, the abundance of microplastics in the ocean is assumed to be increasing, thus increasing the probability of ingestion by biota (Law and Thompson, 2014). The majority of microplastics extracted from the facial products herein were white or blue. It has been suggested by Wright et al. (2013b) that these colours are similar to various types of plankton, a primary food source for surface feeding fish, which are visual predators.

A further potential problem associated with microplastics contamination is the possibility of transport of hydrophobic contaminants by microplastics: such contaminants have been found to sorb onto their surface of plastics and may transfer to biota upon ingestion (Avio et al., 2015; Bakir et al., 2014b; Teuten et al., 2007). Previous studies have shown that PE particles have the potential to sorb and concentrate a range of hydrophobic contaminants. This is of interest because these contaminants can be released in conditions resembling those in the gut of an organism (Bakir et al., 2014b). However, at present, the environmental importance of plastics as a vector in the transport of contaminants is not known. Here we show that microbeads were able to adsorb greater amounts of DDT than Phe when both chemicals were present in a mixture. This was in agreement with previous work indicating that plastic showed a preferential affinity for DDT when present with Phe in a binary mixture (Bakir et al., 2012). The size and shape of microbeads was also found to be an important factor in their sorptive property for POPs and smooth microbeads were found to adsorb lower concentrations of POPs than rough ones. Rough microbeads were found to be most similar in their sorptive properties for POPs to commercially available PE used in chemical transport studies (e.g. Bakir et al., 2012, 2014b,a; Teuten et al., 2007). However, both types of microbeads were broadly similar in their sportive properties to the microplastics used in previous studies. Hence, on the basis of the experimental work here, it seems likely that conclusions regarding the potential role of microplastics as possible vectors in the transport of POPs in the environment could also be applied to transport by microbeads from cosmetics.

Rochman et al. (2013) investigated the transfer of hydrophobic organic compounds (PAHs, PCBs and PBDEs) from PE to the fish, Japanese medaka (*Oryzias latipes*) and the subsequent health effects. Plastic particles were exposed to natural marine conditions, as opposed to laboratory exposures used in most previous studies. Environmental exposure will be highly dependent on the sites selected, which can be prone to variation. Results suggested the ingestion of virgin PE particles caused physiological stresses. However, the ingestion of contaminated PE particles led to the transfer of adsorbed contaminants, causing liver toxicity and pathology (Rochman et al., 2013). Laboratory studies using microplastic particles of polystyrene (Besseling et al., 2013) and PVC (Browne et al., 2013) have also indicated the potential for transfer of harmful chemicals with subsequent effects on biota. The present study showed that plastic particles present in cosmetics can be of varying size and shape and have differential affinities for sorption of POPs. Further work would be needed investigate the presence of chemicals such as pigments and dyes in microbeads, and their potential, if any, for migration from the polymer in either water or gut conditions.

The uneven topography of microplastics used in cosmetics could also provide habitats for diverse communities of microorganisms. A study by Zettler et al. (2013) described the presence of a rich eukaryotic and bacterial microbiota living on PE microplastic samples collected from the North Atlantic subtropical Gyre. Scanning electron microscope (SEM) images showed microbial cells embedded in pits on the plastic surface, and suggested that some members of this community could be accelerating the physical degradation of plastic; however this remains to be confirmed. The communities found on the plastic particles were distinct from surrounding surface water, indicating that plastic provides a novel habitat. Other studies have highlighted the potential for microplastic to act as vectors for microbial pathogens (Harrison et al., 2014).

Currently, there are reported to be eighty facial scrubs in the UK market, which according to their product labelling, contain plastic material amongst their ingredients (Beat the Microbead, 2015). However, some companies have indicated that they will voluntarily phase out microplastics from their products. This could possibly be due to research indicating the negative consequences of microplastics within the environment; Fendall and Sewell (2009) stated that the presence of microplastics in facial cleansers, and their potential use by millions of consumers world-wide, should be of increasing concern, whilst Andrady (2011) also reported that there is an urgent need to assess the future impact of increasing microplastics levels on the world's oceans. There have also been associated public awareness campaigns (eg. Beat the Microbead and Scrub it Out), urging consumers to boycott such products.

However, for the global market, usage statements vary within and between companies, with some stating they will remove all microplastics from all their products, while others say only PE will be removed. In some regions, legislation has been introduced; for example, Illinois and California (U.S.A.) have banned the manufacture and sale of cosmetics that contain plastic microbeads, with similar legislation being proposed for New York, Michigan, and Ohio (but not yet adopted) (Driedger et al., 2015).

In conclusion, the present work characterised the microplastics in facial scrubs by describing the polymer type, colour, size, weight and abundance. This allowed for estimation that between 4594 and 94,500 particles could be released into the environment per use. We also estimate that the UK population is emitting 40.5–215 mg of PE person^{-1} d^{-1}, resulting in a total of 16–86 tonnes yr^{-1}. Particle size, rather than the average weight in each product, was found to be important as it had the greatest effect on abundance estimates. Their small size also renders microbeads accessible to a wide range of organisms, and may facilitate the

transfer of waterborne contaminants or pathogens. There are alternatives to the use of plastics as exfoliating particles (UNEP, 2015); hence these emissions of microplastic are avoidable. Given the quantities of plastic particles reported here, and current concerns about the accumulation of microplastics in the ocean, it is important to monitor the extent to which manufacturers do voluntarily opt to remove microplastics from their products. Such monitoring will help to establish whether there is a need for further legislation.

Acknowledgements

Appendix A. Supplementary material

Supplementary data associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.marpolbul.2015.07.029.

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Release of synthetic microplastic plastic fibres from domestic washing machines: Effects of fabric type and washing conditions

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ABSTRACT

Washing clothes made from synthetic materials has been identified as a potentially important source of microscopic fibres to the environment. This study examined the release of fibres from polyester, polyester-cotton blend and acrylic fabrics. These fabrics were laundered under various conditions of temperature, detergent and conditioner. Fibres from waste effluent were examined and the mass, abundance and fibre size compared between treatments. Average fibre size ranged between 11.9 and 17.7 μm in diameter, and 5.0 and 7.8 mm in length. Polyester-cotton fabric consistently shed significantly fewer fibres than either polyester or acrylic. However, fibre release varied according to wash treatment with various complex interactions. We estimate over 700,000 fibres could be released from an average 6 kg wash load of acrylic fabric. As fibres have been reported in effluent from sewage treatment plants, our data indicates fibres released by washing of clothing could be an important source of microplastics to aquatic habitats.

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1. Introduction

Microplastics have accumulated in marine and freshwater environments, and in some locations outnumber larger items of debris (Browne et al., 2011; Thompson et al., 2004; Wagner et al., 2014). The sources of microplastic include the fragmentation of larger plastic items once they have entered the environment (secondary sources), and also the direct input of microplastic sized particles, such as microbeads used in cosmetics and pre-production pellets (Napper et al., 2015), or particles and fibres resulting from the wear of products while in use (primary sources). Microplastics can be ingested by a wide range of species both in marine (Anastasopoulou et al., 2013; Gall and Thompson, 2015; Lusher et al., 2013) and freshwater environments (Sanchez et al., 2014; Eerkes-Medrano et al., 2015). Laboratory studies indicate the potential for physical harm to biota from the result of ingestion (Wright et al., 2013). Ingestion could also facilitate the transfer of chemicals to organisms, however the relative importance of plastic debris as a vector in the transport for chemicals is not certain (Besseling et al., 2013; Rochman et al., 2013; Koelmans et al., 2013; Koelmans et al., 2014). Encounter rate, as well as polymer type and any associated chemicals (sorbed or additives), will influence the potential for effects in the environment (Teuten et al., 2007; Bakir et al., 2012; Koelmans et al., 2014; Bakir et al., 2014), therefore it is important to understand the relative abundance, as well as the sources of various types of microplastic.

Microplastic has been reported in a wide range of aquatic habitats, including beaches, surface waters, the water column and subtidal sediments (Lattin et al., 2004; Thompson et al., 2004), and there is evidence that the abundance is increasing (Thompson et al., 2004). They are also reported in some of the most remote environments, including the deep sea and the arctic, indicating their ubiquity and the need for further understanding about the potential environmental consequences (Obbard et al., 2014; Woodall et al., 2014).

Release of microplastic sized fibres as a result of washing of textiles has been widely reported as a potential source of microplastic (Browne et al., 2011; Dris et al., 2015; Essel et al., 2015; GESAMP, 2015; Wentworth and Stafford, 2016), however there has been little quantitative research on the relative importance of this source or on the factors that might influence such discharges. This is the focus of the research described here. In this context we consider microplastics as particles of plastic < 5 mm in their smallest dimension. While some fibres may be longer than 5 mm they will usually have a diameter considerably less than 5 mm. There is a lack of clarity on the formal definition for the lower size limit of microplastic and in environmental studies this has tended to relate more to the method of capture; e.g. mesh size of plankton nets used to sample water, or the method of identification such as spectroscopy. At present the smallest particles identified form the environment are around 20 μm in their smallest dimension.

Textiles have the potential to release fibres into the environment, and one pathway is via laundering in washing machines. A range of fibres are used in the production of textiles; these include natural fibres (such as cotton and wool), synthetic fibres (such as nylon) and some are blends of natural and synthetic (such as polyester-cotton). Synthetic

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fibres have been used to supplement cotton, wool and linen in textiles for >50 years, and fabrics such as polyester and acrylic are now widely used in clothing, carpets, upholstery and other such materials. Washing of clothing has been suggested as a potentially important source of microplastic fibres (Browne et al., 2011).

Synthetic microplastic fibres are frequently reported in samples from sediments, the water column and biota (Browne et al., 2011). Waste effluent from washing machines, containing released fibres, will then travel via wastewater to sewage treatment plants (Leslie et al., 2013; Dris et al., 2015). Due to the small size of the fibres, a considerable proportion could then pass through preliminary sewage treatment screens (typically coarse, >6 mm, and fine screens, 1.5–6 mm) (Water Environment Federation, 2003), and be released into aquatic environments. As synthetic fibres are not readily decomposed by aerobic or anaerobic bacteria, any that are intercepted in the sewage treatment plant will accumulate in sewage sludge, and may subsequently be released back to the environment; for example if the sludge is returned to the land or dumped at sea (Habib et al., 1998). Hence, there is a considerable potential for fibres from synthetic textiles to accumulate in the environment; Gallagher et al. (2016) found predominately fibres when surveying the Solent estuarine complex (U.K.) for microplastic. Similarly Dris et al. (2015), found considerable quantities of fibres in the River Seine. There is evidence that some of this material can be transported as airborne particulates (Dris et al., 2015); however it would appear that considerable quantities enter directly from sewage treatment (Browne et al., 2011). To date, there has been limited research to establish the importance of clothing as a source of microplastic contamination to the environment.

A study by Browne et al. (2011), sampled wastewater from domestic washing machines and suggested that a single garment could produce >1900 fibres per wash (Browne et al., 2011). To examine the role of the sewage system as a pathway to the environment, Browne extracted microplastic from effluent discharged by treatment plants, and also examined the accumulation of microplastic in sediments from sewage sludge disposal sites. On average, the effluents contained one particle of microplastic per litre, including polyester (67%) and acrylic (17%) and polyamide (16%); these proportions were similar to the relative proportions found on shorelines and disposal-sites (Browne et al., 2011). Similarly, a high number of plastic fibres were observed in the sediments near to a sewage outfall in Amsterdam (Leslie et al., 2013), and have been reported even 15 years after application in terrestrial soils that have received sewage sludge (Zubris and Richards, 2005). Unless the release of microplastics to waste water or sewage treatment practices change, the release of microplastic to the environment via sewage is likely to increase, as the human population grows. It is anticipated, for example, that reductions in emissions of microbeads via sewage will be reduced as a consequence of legislation to prohibit their use in cosmetics (Napper et al., 2015).

However, there are currently no peer reviewed publications that compare the quantity of fibres released from common fabrics due to laundering. In addition, the potentially important influence of washing practices including temperature, the use of detergent and fabric conditioners have not been examined. Here we tested three different fabrics that are commonly used to make clothes; polyester, polyester-cotton blend, and acrylic. These fabrics were then laundered at two temperatures (30 °C and 40 °C), using various combinations of detergent and fabric conditioner. The fibres extracted from the waste effluent were examined to determine the typical size, and to establish any differences in the mass/abundance of fibres among treatments.

2. Method

Three synthetic fabric types were selected based on their prevalence in high-street retail stores close to Plymouth, UK. The chosen fabric types were all from jumpers (Fig. 2), with each being a different colour so they could be readily distinguished after fragmentation; 100%

polyester (black), 100% acrylic (green) and 65% polyester/35% cotton blend (blue). Four replicates of each garment were purchased, with each replicate sourced from a different retail outlet to provide a representative sample. The identity of each fabric type was confirmed by Fourier transform infra-red spectroscopy (FTIR), using a Hyperion 1000 microscope (Bruker) coupled to an IFS 66 spectrometer (Bruker). The spectra obtained were compared to a spectral database of synthetic polymers (Bruker I26933 Synthetic fibres ATRlibrary). As each garment varied in overall size, 20 cm × 20 cm squares were cut from the back panel of the garments and the edges hemmed by 0.5 cm using black and white cotton thread to deter the excess loss of fibres.

A Whirlpool WWDC6400 washing machine was used to launder the garment samples. While it would be valuable to compare a range of washing machines, this was beyond the budget of the current research. This machine was selected as it is a popular brand used for domestic laundry. The number of fibres released from the wastewater outlet, as a result of laundering, was recorded. To achieve this, a nylon CellMicroSieve™ (Fisher Scientific), with 25 μm pores, was attached to the end of the drain hose. Once a cycle was complete, the CellMicroSieve™ was removed and the fibres collected. Due to the potential build-up of detergent or conditioner on the collected fibres, they were washed using 2 L of water and filtered again over Whatman No. 4 filter papers, and then dried at 30 °C to constant weight. Once dry, the fibres were weighed by a Cubis® precision balance (Sartorius). The weight of fibres were compared across four factors: Factor one, (fabric type, fixed factor, 3 levels: 100% polyester, 100% acrylic, and 65% polyester/35% cotton blend); Factor two wash temperature (fixed factor, 2 levels; 30 °C and 40 °C); Factor three, detergent (3 levels; detergent absent, 20 mL bio-detergent present (contains enzymes), 20 mL non-bio detergent present); Factor four, conditioner (2 levels; 20 mL conditioner absent or present). Factors gave a total of 36 treatments (Fig. 1).

In this study the main factors of interest were: fabric type, temperature, presence of detergent and/or conditioner. The duration of each wash and the rotations per minute are also factors of potential relevance, but were beyond the scope of this study. Therefore, in order not to confound the experimental design they were kept constant (Duration, 1 h 15 min and 1400 rotations per minute (R.P.M)). Each treatment had four replicates.

Cross-contamination was minimized to <8 fibres per wash between washes, by running the washing-machine at 30 °C, 1400 R.P.M for 45 min between washes with no fabric present. Any initial spike in fibre loss from new clothes was reduced by washing each fabric four times before recording any data. Care was taken to ensure any potential sources of airborne contamination were minimized during the analysis (Woodall et al., 2015). The number of fibres released in the effluent from each wash, N, was then estimated from the weight of captured fibres using the following equations and assuming the fibres were of cylindrical shape:

$i) Vt = \frac{Mt}{D}$ $ii) V(avg.fibre) = \pi r^2 l$ $iii) N = \frac{Vt}{V(avg.fibre)}$ where Vt is the total volume of fibres collected, Mt is the total mass of fibres collected, D is the density, $V(avg.fibre)$ is the mean volume of one fibre, N is number of fibres, l is the length and r is the radius.

For each product: equation i) allowed calculation of the total volume of fibres collected; equation ii) allowed calculation of the average volume of a fibre from each garment; by dividing the total volume of fibres by the average volume of a single fibre, equation iii) allowed estimation of the approximate number of fibres released in the effluent from each wash.

Fibres were visualised by scanning electron microscopy (JEOL, 7001F); images taken were used to measure the width of the fibres, and also to analyse their topography. Images of the fibres were also taken by using LEICA M205C light microscope and analysed by Image J to measure their length (Rasband, 2015). For each fabric type, a mean
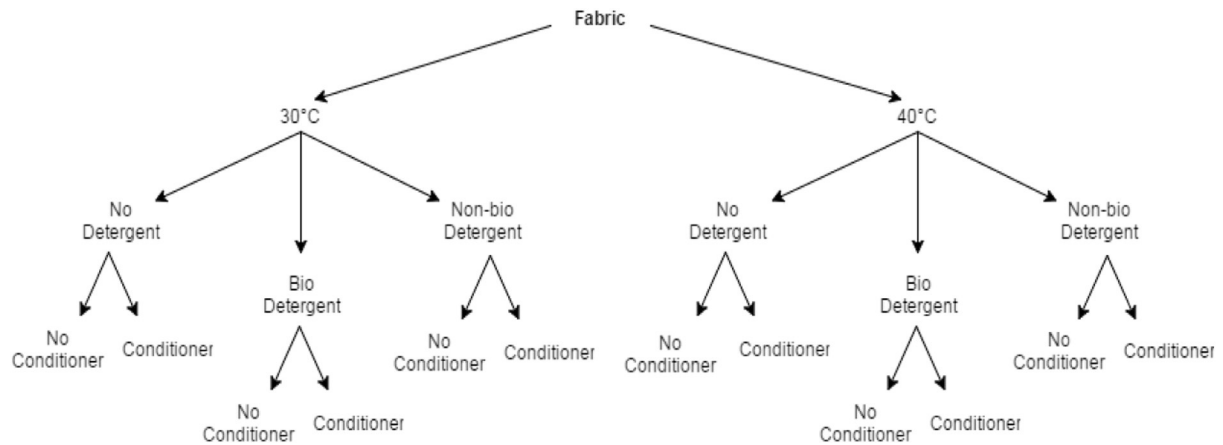
Fig. 1. Experimental design showing factors used for each fabric type (acrylic, polyester, polyester-cotton blend).

size was calculated for length and width based on data from 10 individual fibres.

Using GMav for windows, 4-Way Analysis of Variance (ANOVA) was used to establish any significant effects ($p < 0.05$) between treatments. Post-hoc SNK tests were then used to identify the location of any significant effects.

3. Results

Substantial numbers of microplastic fibres (smallest dimension, <5 mm) were collected from jumpers made out of all three of the common man-made fabrics (polyester, acrylic and polyester-cotton blend) examined (Fig. 2). These were discharged into wastewater from a generic cycle of a domestic washing machine. The fibres were confirmed to be the material type stated on the garment by Fourier transform infra-red spectroscopy. Loss of fibres during the first 4 washes were recorded (Fig. 3), but not included in the data analysis. Polyester showed a steady decrease in fibre loss overall: 1st wash (2.79 mg) to 5th (1.63 mg). Acrylic followed a similar pattern, but the fibre loss decreased more rapidly: 1st wash (2.63 mg) to 4th (0.99 mg). Polyester-cotton blend had the least variation, and showed little decrease between subsequent washes: 1st wash (0.45 mg) to 4th (0.30 mg). Since there was little change in fibre release between the 4th and 5th wash data, data from the 5th wash was used for formal analysis.

While there was a consistent trend between fabric types, ANOVA revealed significant complex interactions between the 4 Factors (Table 1). Focussing on the type of fabric, polyester-cotton blend was consistently found to shed fewer fibres than both the other fabric types, regardless of
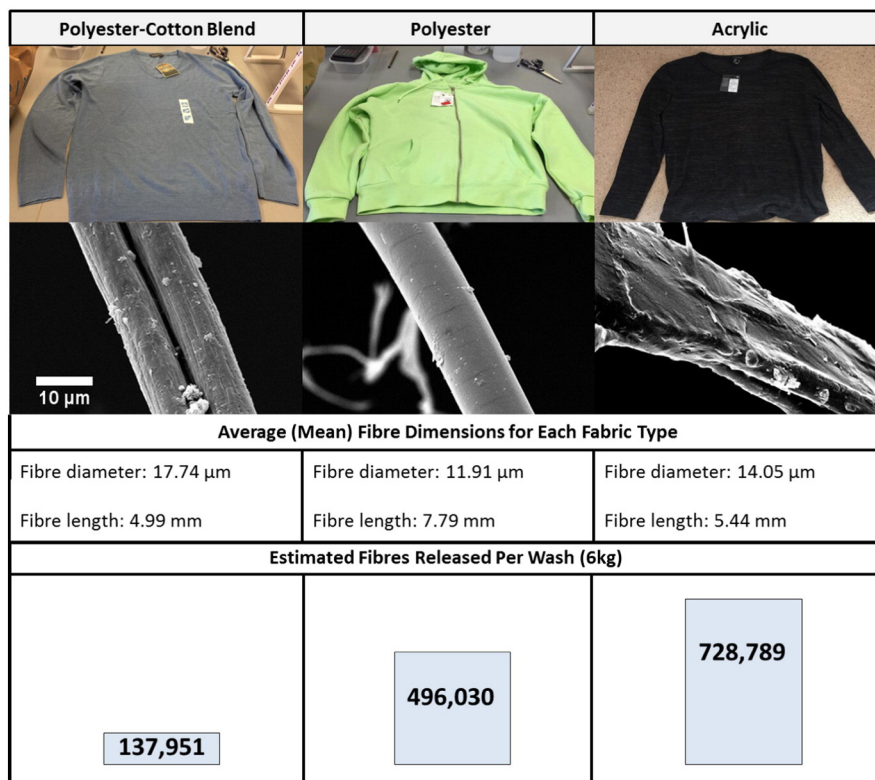

Fig. 2. Images to show the original garments (each representing a different fabric), and a scanning electron microscopy image (SEM) of a typical fibre from each fabric (the scale bar is consistent for all images - 2500× magnification). Key details are included below about the mean dimensions of fibres released during laundering, and estimated quantity released from the fabric during each wash (assuming a typical washing load of 6 kg).
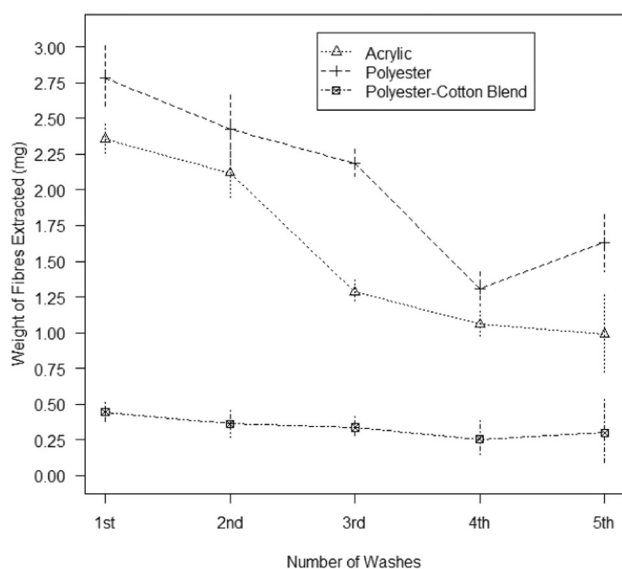
Fig. 3. Fibre loss from three fabrics (acrylic, polyester & polyester-cotton blend), over the first 5 washes. Data from the 5th wash was used in the analysis (n = 4, ±SD).

the differing treatments. This trend was consistent for all 12 relevant interactive effects, and was significantly so for 9 out of these 12 interactions (Table 2a). However, the significance of this effect varied according to the treatment used, creating different interactions. There were some effects of temperature; for example, polyester was often found to release more fibres than acrylic at 40 °C, when compared against 30 °C (Table 2c).

There were also some significant effects of conditioner usage, where polyester-cotton blend consistently shed more fibres when conditioner was used. It was also shown that more fibres tended to be released with the addition of bio-detergent and conditioner. Detergent showed the least clear pattern; however, in some treatment combinations, having no detergent or using bio-detergent resulted in lower quantities of fibres being released. Polyester-cotton blend was also found to shed the least fibres when detergent was absent, and the most when non-bio detergent was used. Hence while there was a clear and fairly consistent trend between fabric types, the effects of temperature, detergent and conditioner were less consistent with some significant effects depending on the specific combinations of factors used.

The extracted fibres were visualised by scanning electron microscopy to examine the differing shapes and surface topography. Polyester-

Table 1
Analysis of variance (ANOVA) for factors affecting release of fibres as a consequence of various laundering treatments (n = 4; **bold** = p ≤0.05). Key: Temp (temperature), Deter (detergent), Cond (conditioner).

Source	Df	MS	F	P
Fabric	2	5.36	83.18	**0.00**
Temp	1	0.10	1.54	0.22
Cond	1	0.37	5.67	**0.02**
Deter	2	0.52	8.07	**0.00**
Fabric × Temp	2	0.02	0.33	0.72
Fabric × Cond	2	0.12	1.88	0.16
Fabric × Deter	4	0.20	3.13	**0.02**
Temp × Cond	1	0.15	2.28	0.13
Temp × Deter	2	0.13	2.09	0.13
Cond × Deter	2	0.58	9.00	**0.00**
Fabric × Temp × Cond	2	0.06	0.86	0.43
Fabric × Temp × Deter	4	0.06	1.00	0.41
Fabric × Cond × Deter	4	0.33	5.05	**0.00**
Temp × Cond × Deter	2	0.64	9.91	**0.00**
Fabric × Temp × Cond × Deter	4	0.38	5.95	**0.00**
Residual	108	0.06		
Total	143			

cotton blend fibres had a rough texture, and were regularly observed as a fusion of 2 smaller fibres. Similarly, acrylic fibres had an extremely coarse surface. Polyester fibres were smooth, without any fracturing (Fig. 2).

Acrylic fibres were on average 14.05 μm in diameter and 5.44 mm in length, giving an average of 763,130 fibres/mg of dry fibres collected from the effluent. Polyester fibres were on average 11.91 μm in diameter, but were longer at 7.79 mm, resulting in around 475,998 fibres/mg of dry fibres collected from the effluent. Polyester-cotton blend fibres were the widest fibres being on average at 17.74 μm, but had the shortest length at 4.99 mm, with an average 334,800 fibres/mg of dry fibres collected from the effluent.

4. Discussion

The environmental consequences of microplastic contamination are not fully understood. The quantity of microplastic in the environment is expected to increase over the next few decades; even if new emissions of plastic debris halted the fragmentation of legacy items that are already in the environment, it would be expected to lead to an increase in abundance (Law and Thompson, 2014). There are concerns about the potential for microplastics to have harmful effects if ingested and some evidence of particle and chemical toxicity have come from relatively high dose laboratory studies. Due to the persistent nature of plastic contamination, there is growing awareness of the need to reduce inputs at source; this includes the direct release of microplastic sized particles including microbeads from cosmetics, and fibres form textiles.

Fibres from fabrics are known to be lost due to pilling. Pilling is defined as the entangling of the fabric surface during wearing or washing, resulting in formation of fibre balls (or pills) that stand proud on the surface of the fabric (Hussain et al., 2008). This occurs as a consequence of two processes: (i) fuzzing; the protrusion of fibres from the fabric surface, and (ii) pill formation; the persistence of formed neps (entangled masses of fibres) at the fabric surface (Naik and Lopez-Amo, 1982). The pill may be worn or pulled away from the fabric, as a consequence of mechanical action during either laundering or wear (Yates, 2002).

Most fabrics pill to some extent and this has always been a concern in the industry as it spoils surface appearance and comfort, reduces the fabric's strength and diminishes its serviceability (Hussain et al., 2008; Chiweshe and Crews, 2000). This problem has become more prominent with the widespread use of synthetic fibres, such as polyester and acrylic, due to their higher tensile strength (Cooke, 1985). These synthetic fibres are widely used because of their low cost and versatile use. Laundry methods have been recognised as being important to minimise the pilling tendency (Cooke, 1985).

The rate or extent to which the pilling stages occur is determined by the physical properties of the fibres which comprise the fabric (Gintis and Mead, 1959). From the fabrics tested here, polyester-cotton blend consistently shed significantly fewer fibres than either of the other fabric types which were entirely synthetic. Polyester is often added to cotton fabric to reduce cost, whilst also increasing tenacity and resilience. This is because cotton fibres have a lower tenacity, and as the pills are formed, the anchor fibres are easily broken; if the tenacity of the fabric is increased with added polyester, the pill break-off rate is lower, resulting in less fibres being released (Mccloskey and Jump, 2005).

Polyester fibres have many desirable properties, including good resistance to strain and deformation (Pastore and Kiekens, 2000). 100% polyester fabrics are renowned for pilling, but because of their high tenacity, the anchor fibres rarely break releasing the pills (Nunn, 1979). Previous research has even reported that as the polyester fibre content in a polyester-cotton blend fabric increases, the pilling gets worse (Gintis and Mead, 1959; Ruppenicker and Kullman, 1981). On the contrary, our research found that polyester fabrics yielded significantly more fibres than polyester-cotton blend. It has previously been suggested that pilling of polyester can be controlled by the modification

Table 2
Outcomes of SNK tests for specific combinations of the factors: a) fabric, b) detergent, c) temperature, d) conditioner. For each combination the relative number of fibres released is indicated by the sequence shown with permutation leading to the greatest release of fibres being shown to the right. Specific variables tested against three different fabric types (acrylic, polyester & polyester-cotton blend), and the subsequent fibre extract from laundering (n = 4; * = p (<0.05)). Key: PE (polyester), Blend (polyester-cotton blend), Acr (acrylic), A (conditioner/detergent absent), C (conditioner present), NB (non-bio detergent), bio (bio detergent).

a) Fabric

Factors			Order
30	C−	No powder	Blend < *Acr < *PE
30	C−	Bio	Blend < *Acr-PE
30	C−	Non-bio	Blend-PE-Acr
30	C+	No powder	Blend < *PE-Acr
30	C+	Bio	Blend < *PE-Acr
30	C+	Non-bio	Blend < *Acr-PE
40	C−	No powder	Blend < *Acr < *PE
40	C−	Bio	Blend < *PE < *Acr
40	C−	Non-bio	Blend-Acr < *PE
40	C+	No powder	Blend < *PE < *Acr
40	C+	Bio	Blend-Acr < *PE
40	C+	Non-bio	Blend < *Acr-PE

b) Detergent

Factors			Order
Acr	30	C−	Bio-NB-A
Acr	30	C+	A-NB-bio
Acr	40	C−	A-NB-bio
Acr	40	C+	Bio-NB < *A
Blend	30	C−	Bio-A-NB
Blend	30	C+	A-bio-NB
Blend	40	C−	A-bio < *NB
Blend	40	C+	A-NB-bio
PE	30	C−	Bio-NB < *A
PE	30	C+	A-bio-NB
PE	40	C−	Bio < *A < *NB
PE	40	C+	A-NB-bio

c) Temperature

Factors			Order
Acr	C−	No powder	40-30
Acr	C−	Bio	30 < *40
Acr	C−	Non-bio	30-40
Acr	C+	No powder	30-40
Acr	C+	Bio	40 < *30
Acr	C+	Non-bio	40-30
Blend	C−	No powder	40-30
Blend	C−	Bio	40-30
Blend	C−	Non-bio	30 < *40
Blend	C+	No powder	30-40
Blend	C+	Bio	30-40
Blend	C+	Non-bio	30-40
PE	C−	No powder	40-30
PE	C−	Bio	40-30
PE	C−	Non-bio	30 < *40
PE	C+	No powder	40-30
PE	C+	Bio	40-30
PE	C+	Non-bio	40-30

d) Conditioner

Factors			Order
Acr	30	No powder	C-A
Acr	30	Bio	A < *C
Acr	30	Non-bio	A-C
Acr	40	No powder	A < *C
Acr	40	Bio	C-A
Acr	40	Non-bio	C-A
Blend	30	No powder	A-C
Blend	30	Bio	A-C
Blend	30	Non-bio	A-C
Blend	40	No powder	A-C
Blend	40	Bio	A < *C
Blend	40	Non-bio	C < *A
PE	30	No powder	C < *A
PE	30	Bio	A-C
PE	30	Non-bio	A < C
PE	40	No powder	C-A
PE	40	Bio	A < *C
PE	40	Non-bio	C < *A

of the polyester properties, where a greater fibre release can improve polyester fabrics surface appearance (Doustaneh et al., 2013). Weakening the fibres (reduced ultimate bending stiffness), leads to more rapid break-off of pills due to fibre fatigue, leading to greater fibre release while at the same time improving the fabrics topography and surface appearance (Doustaneh et al., 2013). Hence from an aesthetic perspective, there may be benefits to the release of pills from garments during washing. However, this can also create a trade-off between garment appearance, and fibre release. More research would be needed to establish how release rates vary over the lifetime of a garment in service in order to fully establish the temporal dynamics of fibre emissions.

During the laundering of clothes, detergent and fabric conditioner are often used in combination. Synthetic detergents remove the oils and waxes that serve as lubricants in natural fibres, making a garment clean but harsh, scratchy, and uncomfortable to wear (Egan, 1978). Fabric softeners are used to counteract these effects. In addition, the use of fabric conditioners can reduce the build-up of static electricity, which can make the fabric objectionable to the wearer. Fabric softeners act as antistatic agents by enabling synthetic fibres to retain sufficient moisture to dissipate static charges (Ward, 1957).

Fabric conditioners may also increase pilling, and this is especially the case for synthetic fibres (Smith and Block, 1982). Work by Chiweshe and Crews (2000), showed that use of fabric conditioner on all cotton-containing fabrics resulted in increased pilling and/or an increase in the size of pills, as well as increased breaking strength losses in polyester woven fabric. Hence, it might be expected that the presence of conditioner could increase the release of fibres. This was observed in some of the treatment combinations here, but there was no clear trend relating to the presence of conditioner.

Detergent use presented the least clear pattern for fibre release when compared against the other factors. However, it was found that having no detergent or bio-detergent in a wash cycle occasionally resulted in the fewer fibres being released. Previous research has also shown that when polyester-cotton blend fabric has been laundered with a bio-detergent, it exhibited less piling than when laundered using a non-bio (Chiweshe and Crews, 2000). Our research produced some similar results, where polyester-cotton blend was also found to shed fewer fibres when detergent was absent, and the most when non-bio detergent was used.

Using the results from this experiment, the number of fibres potentially released into washing machine waste water per wash was estimated. This was achieved by examining the average fibre size, the various Factors tested and assuming a typical washing load of 6 kg. Based on this, a washing load (6 kg) of polyester-cotton blend was estimated to release 137,951 fibres, polyester to potentially release 496,030 and Acrylic 728,789. The large number of fibres released when clothing is laundered is therefore likely to represent a substantial contributor to microplastic contamination in the environment. Our estimates are similar to research by Browne et al. (2011), where it was suggested that a single garment could produce >1900 fibres per wash (Browne et al., 2011).

Wastewater Treatment Plants (WWTPs) play a critical role in the fate and transport of microfibres into the environment. In countries with sewage infrastructure, the effluent from washing machines is discharged into the local sewer system. This is then treated by a WWTP and discharged as treated effluent, which is released into the aquatic environments. Effluent discharge often contains suspended solids, such as microfibres, which are not removed during the treatment

processes. In Amsterdam, Leslie et al. (2013) found concentrations from WWTP effluent ranged from 9 particles/L (min.) to 91 particles/L (max.) with a mean and median of 52 particles/L. A study by Murphy et al. (2016), compared the influent and effluent from a WWTP. The influent contained on average 15.70 (± 5.23) microplastic/L, and was found to be reduced to 0.25 (± 0.04) microplastic/L in the final effluent, a decrease of 98.41%. However, emissions of microplastics may still be substantial. For example, Mintenig et al. (2014) calculate between 8.2 and 93 billion microplastics and synthetic fibres being discharged from wastewater treatment plants in Germany (Essel et al., 2015). Even a small amount of microplastic being released per litre can result in substantial amounts of microplastics entering the environment due to the large volumes being treated. It has been predicted that a WWTP plant in the United Kingdom could release up to 65 million microplastics into the receiving water every day (Murphy et al., 2016).

Even if WWTPs are completely effective in the removal of microfibres, the extracted plastic particles may still enter the environment if the resultant sewage sludge, a by-product of the wastewater treatment process, is returned to the land; for example as a fertilizer (Habib et al., 1998; Zubris and Richards, 2005). Microfibres in sewage sludge may subsequently persist in the terrestrial environment, or be transported to aquatic environments via runoff. The potential for sewage sludge to transfer microplastic into the marine environment was shown in a preliminary study by Habib et al. (1998), where sediments were collected from a bay downstream of a sewage treatment plant. It was found that the sediment contained numerous synthetic fibres, and as distance from the sewage treatment plant increased, the size and number of fibres decreased. This effect was also observed by McCormick et al. (2014), where a higher concentration of microplastic (17.93 m^3) was recorded downstream of a WWTP, compared to upstream (1.91 m^3) (McCormick et al., 2014).

Clothing design, including the type of fabric used, clearly has considerable potential to influence fibre release; for example, our research found that a fabric made from a synthetic-natural combination released around 80% fewer fibres than acrylic. Further work to better understand how fabric design and textile choice influence fibre release should therefore be undertaken. Important directions for future research include comparing release between different types of washing machine, and using a variety of wash durations and spin speeds together with an assessment of the temporal dynamics of fibre release throughout a products life time. The Plastic Soup Foundation and MERMAIDS Life + project are currently promoting development of innovative solutions to minimise the release of plastic fibres from garments. Filters for washing machines are also being developed (Mermaids Organisation, 2015). These are made of a stainless steel mesh, with hole diameters of 0.0625 in. to collect fibres (Environmental Enhancements, 2016). For this measure to be successful, it will be essential to ensure the filters are not subsequently disposed of via household liquid waste. However, from a material usage and efficacy perspective, minimising fibre release at the design stage should be regarded as the most effective priority in a management hierarchy.

From the perspective of sustainability and environmental contamination, criteria that synthetic garment manufactures should consider might therefore include: 1) performance in service, giving a long lasting product that remains attractive during usage; 2) minimal release of non-degradable synthetic fibres and 3) a product that is compatible with end of life recycling. Such factors need to be taken into account throughout the design and manufacturing stages; for example, including consideration of fibre properties (composition, length), spinning method and the weaving/knitting process. Inadequate consideration of potential environmental impacts at the product design stage has recently led to considerable negative publicity and restrictive legislation relating to emissions of plastic microbeads from cosmetics (Napper et al., 2015); clearly illustrating the benefit of a precautionary approach. With microbeads in cosmetics, one of the considerations guiding policy intervention was the lack of clear societal benefit from incorporating

microplastic particles into the cosmetics, coupled with concerns about environmental impacts. The societal benefits of textiles are without question, and so any voluntary or policy intervention should be directed toward reducing emissions either via changes in textile design or filtration of effluent, or both. As well as considering direct environmental impacts of manufacture, product use and disposal, there is a growing realisation of the need for a more circular approach to material usage in order to maximise long term resource sustainability and waste minimisation via a circular economy (European Commission, 2012; World Economic Forum, 2016).

In conclusion, this work examined the release of textile fibres from three fabrics that are commonly used to make clothing (polyester, polyester-cotton blend and acrylic). The results show that laundering 6 kg of synthetic materials could release between 137,951–728,789 fibres per wash. Our results indicate significant effects of wash conditions, but no clear picture based on the two detergents and one conditioner used. Hence, further work to examine in more detail differing washing machines and wash treatments, involving wash duration and spin speed as well as temperature, detergent and conditioner may be worthwhile. This could help establish whether specific wash conditions could be used to help minimise fibre release. Temporal dynamics of release over the life time of a product should also be examined, as this could help extend garment life while at the same time reducing fibre emissions.

Acknowledgements

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