1	Characterisation, Quantity and Sorptive Properties of Microplastics Extracted
2	From Cosmetics
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23 Abstract

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 to 327 µm). We estimated that between 4594 – 94500 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 <sup>3</sup>H-phenanthrene and <sup>14</sup>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.

### Keywords:

- Microplastic; Exfolianting Microbeads; Polyethylene; Ocean pollution;
- 40 Contaminant

### 47 <u>1. Introduction</u>

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 <5mm). 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 preproduction 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 (Cole et al 2011; Teuten et al., 2007; Thompson et al., 2009a).

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 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 to D contained 150mL of product, E contained 125mL). The contents were subjected to vacuum filtration to obtain the

plastic particles. The procedure required mixing each product in approximately 1L 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 ATRlibrary).

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  $0.015~\mu m$ , to  $2000~\mu m$ ; 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:

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: equation i) allowed calculation of the total volume of microplastic extracted; equation 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, equation 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, 2014b, 2012).

Adsorption experiments were conducted in an ISO9001 accredited radioisotope facility at the Plymouth University.  $^3\text{H-Phe}$  and  $^{14}\text{C-DDT}$  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  $\mu$ L of  $^{14}\text{C-DDT}$  and 16  $\mu$ L of  $^3\text{H-Phe}$  were added to the walls of the tubes. The solvent was allowed to evaporate and 25 mL of seawater (35 psu, 59.3  $\pm$  0.26 mS) was added and the tubes were equilibrated for 48 hours (Bakir et al., 2014a) in the dark at 18  $^\circ\text{C}$ 

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  $\beta$  decay from the <sup>14</sup>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:

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$$Kd = [q_e]_{solid} / [C_e]_{aa}$$

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

## 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 prerequisites 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.

### 3.0 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 ( $10\mu m$  to >2000  $\mu m$ ), whereas product A was the most homogenous, ranging from  $8\mu m$  to  $56\mu m$ , 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  $288.80~\mu m$ ,  $289.63~\mu m$  and  $293.48~\mu m$  respectively. The particles in product B and C were larger, with mean diameters of  $326.83~\mu m$  and  $317.91~\mu m$ , while product A was much smaller with a mean diameter of  $163.82~\mu m$ . 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, 6.11g in each bottle, but resulted in an estimate of 18906 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 (Figure 3). The 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.
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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.

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### 4.0 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). Effulent 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.

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 5mL, between 4594 – 94500 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 <sup>-1</sup>.d <sup>-1</sup>, amounting to an emission of 263 tonnes yr<sup>-1</sup>. 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 <sup>-1</sup>.d <sup>-1</sup>, or between 16 and 86 tonnes yr<sup>-1</sup> (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 115km 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., 2009; Browne et al., 2008). However, studies that quantify the 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.

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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 94500 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 manufactuers do voluntarily opt to remove microplastics from their products. Such monitoring will help to establish whether there is a need for further legislation.

506 <u>Tables</u>

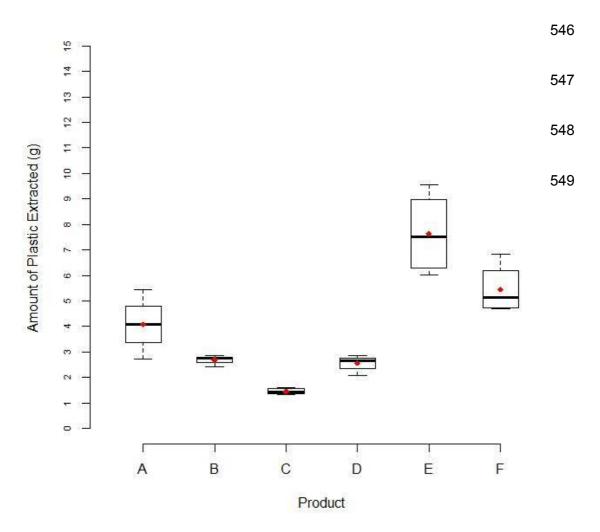
Product	Colour of Microplastic Present
А	White and Pink
В	White
С	White
D	White and Light Blue
Е	White and Dark Blue
F	White and Dark Blue

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

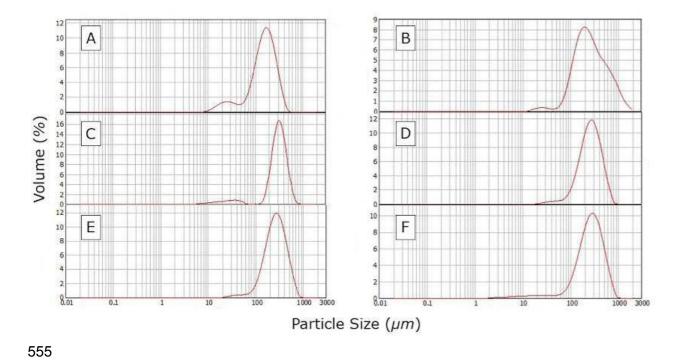
				Solid	Total
Particle type	POP	Aqueous phase	Glass wall	phase	recovery
Product X beads	DDT	12	8	59	78
	Phe	43	1	24	68
Product Y particles	DDT	7	8	91	106
·	Phe	13	3	65	81
Product Z beads	DDT	20	26	33	79
	Phe	64	2	6	73
Product Z particles	DDT	3	8	90	101
·	Phe	11	5	60	75
UHMW PE	DDT	2	6	87	94
	Phe	7	2	80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

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

544 <u>Figures</u>



**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.

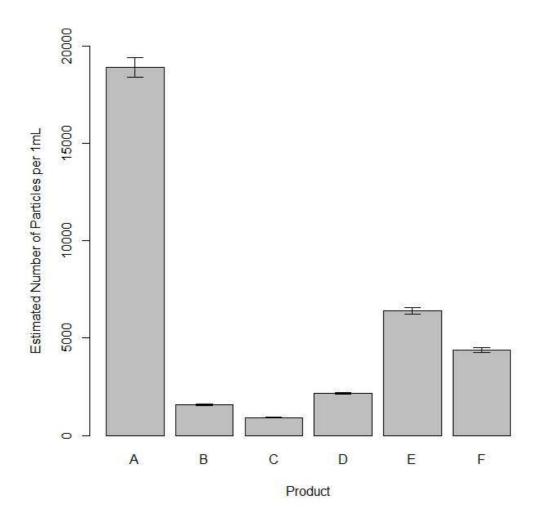
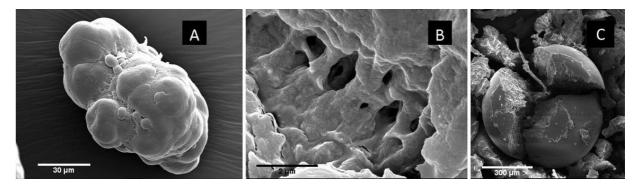
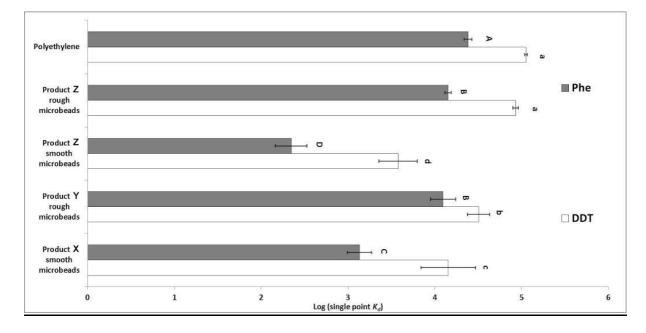


Fig 3. Estimates for the number of PE microbead particles in six brands of facial scrubs per 1mL. Calculated using data from the volume weighted mean (n = 3, ±SD; correlating to the spread of the different amounts of particles calculated for high, medium and low density PE).



**Fig 4.** A-Scanning electron microscopy (SEM) of a typical rough facial scrub plastic microbead particle (9000 magnification). B- SEM of surface microbead topography (16000 magnification). C- SEM of a broken smooth spherical plastic microbead from 'product F' (900 magnification).





**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,  $\pm$  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).

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