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THE SOURCES AND FATE OF PLASTIC ENTERING THE MARINE ENVIRONMENT

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THE SOURCES AND FATE OF PLASTIC ENTERING THE MARINE ENVIRONMENT

by

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in partial fulfilment for the degree of

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AUTHOR'S DECLARATION

At no time during the registration for the degree of Doctor of Philosophy has the author been registered for any other University award without prior agreement of the Doctoral College Quality Sub-Committee.

Work submitted for this research degree at the University of Plymouth has not formed part of any other degree either at the University of Plymouth or at another establishment.

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Abstract

THE SOURCES AND FATE OF PLASTIC ENTERING THE MARINE ENVIRONMENT

Imogen Ellen Napper

Plastics can come in many forms and bring a wide range of societal benefits in healthcare, agriculture, transport, construction and packaging. However, plastic is a large proportion of marine litter and an internationally recognised pollutant. Marine litter results from the indiscriminate disposal of waste items that are either directly or indirectly transferred into aquatic environments. This debris can cause harmful effects for wildlife, commercial fisheries, maritime industries, tourism and human well-being (Chapter 1).

Over the past decade, increased scientific interest has produced an expanding knowledge base for plastic contamination in the environment. However, fundamental questions and issues remain unresolved. This thesis addresses several prominent sources of plastic litter and discusses the routes by which the plastic can enter the environment; these include facial scrubs, laundering clothes, wet wipes and carrier bags. Whilst the emphasis of this thesis is on microplastics (both primary and secondary source), the indiscriminate disposal of macroplastics are also considered, as, with time, they have the potential to degrade into secondary microplastics.

Cosmetic products, such as facial scrubs, were identified as a potentially important primary sources of microplastics to the marine environment. Up to 94500 microplastics were found to release in a single use (Chapter 2). The laundering clothes was also found to be an important source of primary microplastic where over 700,000 fibres were estimated to be released from an average 6 kg wash load (Chapter 3). Wet wipes degradability was tested for both flushable and non-flushable alternatives. There are currently no standardised criteria to measure whether a product can be flushed, and the degradability of these products still remains unclear (Chapter 4). Furthermore, different formulations of carrier bags that stated that they were biodegradable and oxo-biodegradable, were still present in the soil and marine environment after 3 years. This suggests that they have no meaningful advantage over conventional bags in terms of consequences of persistence. (Chapter 5).

The versatility, low cost and often single use application of plastic mean that the material is utilised for a wide variety of products. Therefore, one of the major challenges in addressing the issues surrounding marine plastic debris is the diverse nature of plastic products and the many routes by which they can enter the environment. To help manage and reduce emissions it is essential to better understand the relative importance of these sources, assess regional variation and target specific solution options (Chapter 6).

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Chapter 1

General Introduction; Micro- and Macroplastics in Aquatic Environments

Abstract

Marine litter is a growing environmental problem which can pose threats to the environment, the economy and human wellbeing on a global scale. Over 300 million tons of plastic is produced annually and around 75% of all marine litter is plastic. Plastic litter is widespread in aquatic ecosystems and comes from a variety of sources. Once in the environment, plastic can fragment due to exposure to UV light, heat and abrasion; this can lead to the formation of microplastic (<5mm). Plastic debris can be harmful to wildlife as a consequence of ingestion and entanglement. There are also concerns about the transfer of chemicals by plastic to organisms upon ingestion, but there is currently little evidence to indicate that plastics provide a major vector for chemical transfer in the environment. Plastic debris also has a range of negative effects for commercial fisheries, maritime industries and tourism. There is also emerging evidence of negative consequences for human well-being. This general introduction provides a summary for the current knowledge of plastic as a component of marine litter. It focusses on the different size classes, potential impacts and then links to the different sources the plastic debris could originate from. In the final section it also outlines the objectives of this thesis and indicates the specific aims of each chapter.

1.0 General Introduction; Micro- and macroplastics in aquatic environments

1.1 Introduction

The human race generates a considerable amount of solid waste on a daily basis. Quantities of this waste are increasing, although amounts vary between countries. Plastics are nearly always a major component of this waste, as well as being the dominant category of litter reported in the marine environment (Thompson et al 2004; Jambeck et al., 2015). There has been growing evidence about the effects plastic contamination can have in the environments . Plastic litter now contaminates marine habitats from shallow water to the deep sea (Bergmann and Klages, 2012; Woodall et al., 2014) and has been identified as a major global issue by the United Nations Environment Assembly and in the G7 Leader´s declaration 2015 (GESAMP, 2016; UNEP, 2017; Werner et al., 2016).

Plastics are synthetic or semi-synthetic organic polymers. They are typically lightweight, strong, inexpensive, durable and corrosion-resistant (Derraik, 2002; Thompson et al., 2009b). Most plastic items are composed of hydrocarbons derived from fossil oil or gas feedstocks (Am. Chem. Counc., 2015). During the conversion from resin to product, a wide variety of additives (such as fillers, plasticizers, flame retardants, thermal stabilizers, antimicrobial agents and colouring agents) may be added to enhance performance and appearance of plastic items (Andrady and Neal, 2009). As a consequence, plastic materials can take many forms including rigid items together with more flexible films, adhesives, foams and fibres.

The most commonly used polymers are high-density polyethylene (HDPE), low-density polyethylene (LDPE), polyvinyl chloride (PVC), polystyrene (PS), polypropylene (PP) and polyethylene terephthalate (PET), which cumulatively account for approximately 90% of total plastic production. These plastics bring a wide range of societal benefits in healthcare, agriculture, transport, construction and packaging (PlasticsEurope, 2016). The versatility of plastic materials has resulted in a substantial increase in their use from 5 million tons globally in the 1950s to over 335 million tons today (Andrady and Neal, 2009; PlasticsEurope, 2018).

Despite the durability of plastics, the main uses are in relatively short-lived applications, such as packaging, which accounts for around 40% of all production. While packaging can help protect food, drink and other items (thus reducing damage and wastage of products), it also results in rapid accumulation of persistent plastic waste. This has led to one of the most ubiquitous and long-lasting recent changes to the surface of our planet; the accumulation and fragmentation of plastic debris (Barnes et al., 2009). Plastics represent a substantial fraction of the municipal waste stream and a large proportion of marine litter is plastic (Jambeck et al., 2015). This debris is widely reported in the environment where it has accumulated at the sea surface (Law et al., 2010) , on shorelines of even the most remote islands (Barnes, 2005), in the deep sea (Bergmann and Klages, 2012; Woodall et al., 2014) and in arctic sea ice (Obbard et al., 2014). There is also increasing awareness of the accumulation of plastic litter on land as well as in freshwater habitats (Eerkes-Medrano et al., 2015).

Once in the environment, exposure to ultra-violet radiation, heat and oxygen can cause plastics to become brittle and physical action can then cause them to break down into smaller pieces, including microplastics. The timescale for degradation of discarded plastics is not known with certainty and will depend on the chemical nature of the material, the characteristics of the environment in which they persist and the manner in which degradation is measured (Andrady and Neal, 2009). However, some polymer chemists suggest that all of the conventional plastic that has ever been produced, with the exception of any material that has been incinerated, still persists in the environment in a form too large to be biodegraded (Thompson et al., 2005).

1.2 Definitions of Plastic Litter

Plastic debris can be defined and described in a variety of ways including by shape, colour, polymer type, origin and original usage (e.g. packaging). Plastics enter the aquatic environment in a wide range of sizes (Cole et al., 2011; Hidalgo-Ruz et al., 2012) and have been reported hundreds of meters in length to microns in diameter. There are three categories that are typically used to describe the size of plastic contamination; macroplastic (>20 mm diameter), mesoplastic (5–20 mm) and microplastic (<5 mm) (Barnes et al., 2009; Thompson et al., 2009a). Although there is uncertainty about absolute quantities of plastic in the environment, or the ultimate sinks for this debris, there is evidence of increasing quantities over time (Jambeck et al., 2015; Thompson et al., 2004).

1.2.1 Macro & Mesoplastic

Macroplastic refers to plastic items larger than 20 mm. Due to its high visibility, contamination of the environment by macroplastic may be perceived as one of the most concerning forms of plastic pollution. The accumulation of macroplastic has been reported in a wide range of habitats (Browne, 2015; Ryan et al., 2009). Clean-up campaigns typically focus on these larger items and there is wide geographical variability in abundance, which increases the difficulty of analysing potential trends. However, due to the size of this debris, it is often possible to categorize items according to their original usage; for example, packaging, fishing or sewage related debris. Plastic debris that is larger than 5 mm but smaller than 20 mm is termed mesoplastic.

1.2.2 Microplastic

Microplastic is used as a collective term to describe a heterogeneous range of small plastic particles and fibres. In 2008, the National Oceanographic and Atmospheric Agency (NOAA) of the U.S. hosted the first International Microplastics Workshop and as part of this meeting formulated a working definition to include all plastic particles less than 5 mm in diameter (Arthur et al., 2009). The lower size limit is typically set by the capacity of capture methodology or analytical identification equipment and is currently around 20 microns. However, it is widely believed that plastic debris is present in the environment in the nano-size range (Mattsson et al., 2015). As with macroplastics, microplastics can differ in specific density, chemical composition, and shape (Fig. 1.1b) (Duis and Coors, 2016; Law and Thompson, 2014). Microplastic can then be further divided into categories based on their origin; primary and secondary microplastics.

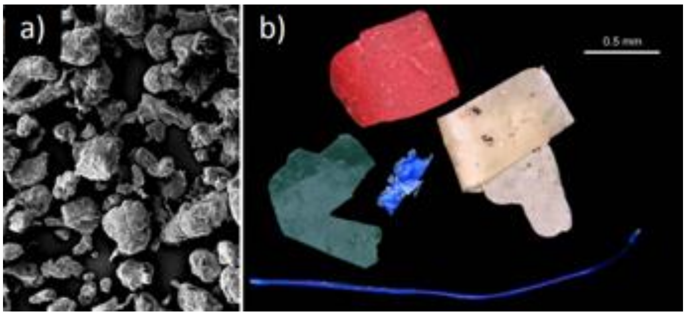

Figure 1.1 *Pictures showing typical microplastic samples: a) Polyethylene particles extracted from a cosmetic product. Source: Napper & Thompson, Plymouth University Electron Microscopy Suite. b) Fragments of microplastic collected from a shoreline near to Plymouth, UK. Source: Thompson, Plymouth University. Note: Scale bar applies to both pictures.*

1.2.3 Primary Microplastic

Particles that directly enter the environment in the microplastic size (<5mm in diameter) are described as primary microplastics. Primary microplastics are produced through extrusion or grinding, either as a feed stock for manufacture of products (Turner and Holmes, 2015) or for direct use (Browne, 2015); for example in cleaning products (Cole et al., 2011; Derraik, 2002), cosmetics (Fig. 1.1a) (Napper et al., 2015; Zitko and Hanlon, 1991) and as air-blasting media (Gregory, 1996).

1.2.4 Secondary Microplastic

Secondary microplastics are those formed in the environment from the fragmentation of larger items of plastic debris (Cole et al., 2011; Law and Thompson, 2014). This degradation occurs as a consequence of ultra-violet (UV) radiation and oxidation, which overtime can reduce the structural integrity of the plastic, resulting in fragmentation. This process is called photo-oxidation and is a result of the absorbance of high-energy wavelengths of the ultraviolet (UV) spectrum by the polymers (Singh and Sharma, 2008). Once degradation is initiated, it can proceed through temperature-dependent thermo-oxidative reactions without further exposure to UV radiation, as long as oxygen is available (Andrady, 2011). This ultimately causes plastics to become brittle and fragment. This can be facilitated by physical forces from abrasion, wave-action and turbulence (Barnes et al., 2009; Browne et al., 2007). The process is ongoing, with fragments becoming smaller and smaller over time (Cole et al., 2011; Galgani et al., 2010).

Even if emissions of larger items of plastic to the environment were to cease with immediate effect, it is likely that there would still be an increase in the quantity of microplastic as a consequence of the fragmentation of larger items that are already in the environment. Secondary microplastic can also be generated as a consequence of wear during the use of a product. For example, fibres generated from the laundering of clothes or from the wear of tyres.

The timescale for degradation of discarded plastics is not known with certainty and will depend on the chemical nature of the material, the characteristics of the environment in which they persist and the manner in which degradation is measured (Andrady and Neal, 2009). A rage of factors influence rates of degradation in the environment; hydrolysis and biodegradation, occur at rates several orders of magnitude slower than the oxidative mechanisms (Andrady, 2011). Plastics typically take much longer to degrade in water than they do on land, mainly owing to the reduced UV exposure and lower temperatures found in aquatic habitats (Gregory and Andrady, 2003). It has been suggested that all of the conventional plastic that has ever been produced, with the exception of any material that has been incinerated, still persists in the environment in a form too large to be biodegraded (Thompson et al., 2005).

1.3 Sources

The majority of plastic in the sea originates from inland sources and is emitted to the oceans from coastlines or by rivers (Jambeck et al., 2015). In addition, quantities are released from ocean-based sources such as shipping and aquaculture (Andrady, 2011; GESAMP, 2015). Smaller particles may also be carried in the air and deposited at sea (Dris et al., 2015). It has been estimated that on a global scale, the input of plastic into the oceans is 4.8 to 12.7 million metric tons per annum (based on 2010 data) (Jambeck et al., 2015). Furthermore, assuming there are no improvements in waste management infrastructure, the cumulative quantity of plastic waste available to enter the marine environment from land could increase by approximately three times over the next decade (Fig. 1.2) (Jambeck et al., 2015). However, a more precise estimate will require direct measurement of the input rates of plastic waste by wind, tidal and ocean wave transport. It will also require a methodical measurement of waste generation, collection rates, classification and waste disposal methods for rural areas and urban centres in countries around the world (Law, 2017). Microplastics have been detected at very high levels globally in rivers and lakes which could further add to this estimation (Auta et al., 2017; Free et al., 2014; McCormick et al., 2016).

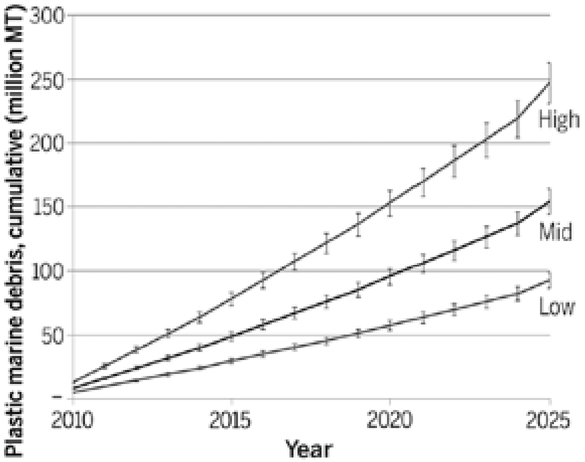

Figure 1.2. *The estimated mass of mismanaged plastic waste (millions of metric tons) input into the ocean by populations living within 50 km of a coast in 192 countries, plotted as a cumulative sum from 2010 to 2025 (Jambeck et al., 2015).*

1.3.1 Sources of Macroplastic

Much of the litter in aquatic environments enters as macroplastic from land-based actions such as general littering, dumping of waste and loss during waste collection as well as that from inappropriately managed landfill sites (Jambeck et al., 2015; Mehlhart and Blepp, 2012). Plastic waste is collected, and then contained in a waste management framework which is designed to help minimize loss to the environment. From these land-based sources, plastic litter then has the potential to end up in municipal wastewater and freshwater systems (Cole et al., 2011; Leslie et al., 2013). This can result from windblown litter escaping into the wider environment (Barnes et al., 2009; Mehlhart and Blepp, 2012; Pruter, 1987). In industrialized countries, waste that is deposited in landfills is usually covered regularly with soil or a synthetic material, and the landfill is cordoned by a fence to prevent any debris accidentally leaving. However, in developing regions this is often not the case (Barnes et al., 2009; Jambeck et al., 2015). The residues from plastic recycling could also unintentionally escape into the environment (Moore, 2008).

Items released at sea are also an important component; analysis of floating macro-debris revealed that 20% by number and 70% by weight was fishing–related, principally floats and buoys (Eriksen et al., 2014). This was based on 4291 visual observations from 891 sampling locations in the North and South Pacific, North and South Atlantic, Indian Ocean, Bay of Bengal, Mediterranean Sea and coastal waters of Australia. Additionally, in 2010, the amount of fishing gear lost to the environment was estimated at around 640,000 tonnes per year (Good et al., 2010). Studies have indicated a significant relationship between the number of ocean-based plastic items found on beaches and the level of commercial fishing activity (Cunningham and Wilson, 2003; Ribic et al., 2010). Furthermore, unintentional loss of in-service macroplastic products can occur when catastrophic events, such as tsunamis, hurricanes, or floods, carry large amounts of material of all kinds into the marine environment (Law, 2017).

Studies have indicated a significant relationship between the number of ocean-based plastic items found on beaches and the level of commercial fishing activity (Cunningham and Wilson, 2003; Ribic et al., 2010). In 2010, the amount of fishing gear lost to the environment was estimated at around 640 000 t yr^{-1} (Good et al., 2010). Discarded fishing items, which include monofilament lines and nylon netting, can float at a variety of depths and result in 'ghost fishing' and entanglement of aquatic organisms; this will be discussed in the 'impact' section (Good et al., 2010).

1.3.2 Sources of Microplastic

Primary microplastic can result from spillage/mishandling of industrial pre-production plastics or from the use of cosmetics (Duis and Coors, 2016; Law, 2017; Napper et al., 2015). Plastic microbeads from facial scrubs are an example of a cosmetic use source. After their intended use, these microbeads are likely to enter household wastewater and some will escape the waste water treatment system into the environment (Murphy et al., 2016). It has been estimated that 94,500 microbeads could be released from an exfoliant in a single use, accumulating to the UK alone to be emitting 16–86 tonnes yr^{-1} (Napper and Thompson, 2016). Other potentially important sources of microplastics are from microplastic used in medicines, drilling fluids for oil/gas exploration and in industrial abrasives (i.e. for air-blasting to remove paint from metal surfaces) (Derraik, 2002; Duis and Coors, 2016; Gregory, 1996; Mintenig et al., 2014; Sundt et al., 2014).

Further sources of microplastic to the marine environment occur as a consequence of the breakdown of larger plastic debris (secondary microplastic). These can then enter the marine environment through two different pathways; a 'direct' (sewage or storm water) or an 'indirect' (fragmentation of existing plastic debris) source. Washing of clothes made from synthetic materials is an example of a direct secondary microplastic source. Again, this microplastic can enter the environment via wastewater after the release of fibres from a washed garment. Some fabrics release fibres more readily than others; research by Napper and Thompson (2016) reported that a wash load of 6 kg of acrylic clothing could release over 700,000 fibres.

For primary and secondary microplastic pieces larger than around 20μm, it is possible to identify what type of plastic polymer a particular piece of marine debris is made out of (Thompson et al., 2004). However, it is extremely difficult to trace back the debris to its origin.

1.3.3 Waste Water Treatment Plants

For any plastic that enters waste water treatment plants, the efficiency of capture (i.e., before the effluent is discharged into the environment) depends on the particular treatment process. However, there is limited information on the efficiency of waste water treatment plants to capture plastic; particularly microplastic. Some studies indicate extremely high capture rates (>95%) of plastic particles (Carr et al., 2016; Murphy et al., 2016). Given the large volume of influent daily, even low loss rates could result in detectable concentrations of these plastic particles in the environment (Browne et al., 2011; Eriksen et al., 2013). Murphy et al. (2016) predicted that waste water treatment plants could release 65 million microplastics every day (Murphy et al., 2016). Wastewater and any plastic debris therein can also bypass treatment as a consequence of sewage overflows. Even if microplastic is intercepted during wastewater treatment, the resultant sewage sludge is often returned to the land as a fertilizer, hence plastic is still released to the environment (Kirchmann et al., 2017).

1.3.4 Source Trends

The sources of marine plastic litter are mostly well known, but there is still a considerable lack of knowledge concerning the relative importance of the different sources and their original usage (Nilsen et al., 2014). Some items can be attributed with a high level of confidence to their origin; such as fishing gear and sewage-related debris (Galgani et al., 2015). However, some types of plastic debris are much harder to trace back; such as microplastic fragments.

Given the large amount of macroplastics entering the environment, it is generally assumed that there will be a rise in microplastics from the fragmentation of larger items (Andrady, 2011). However, the fragmentation rates of macroplastics are largely unknown, and as a result little quantitative information is available on the relative contribution from the fragmentation of larger items to the abundance of microplastics (Koelmans et al., 2014; Law and Thompson, 2014; Sundt et al., 2014).Therefore, it is important to understand sources, pathways and quantities of larger items to the environment as they are the most likely source of microplastic in our oceans (Andrady, 2011; Browne et al., 2007; Hidalgo-Ruz et al., 2012).

In terms of abundance, there are no definitive estimates of the total quantity of plastic in the environment overall. Estimates based on counts at sea suggest between 7000 or >250 000 t of plastic are now present in the open-ocean surface (Cózar et al., 2014; Eriksen et al., 2014). However, estimations can often depend on what size fractions are being referred to. The trends of production, consumer-use and demographics all point to a further increase of plastic use in the future (Auta et al., 2017; Sutherland et al., 2014). Hence there are considerable concerns that the problems of plastic pollution will escalate unless disposal practices change.

1.4 Distribution

Once released into the marine environment, and due to their durability, plastic debris has the potential to become widely dispersed (Fig. 1.3). The plastic debris can be transported by wind and currents (Faure et al., 2015; Ryan et al., 2009). Plastic has been found to accumulate in the oceans (Thompson et al., 2004), estuaries (Browne et al., 2010) and even in remote habitats such as in arctic ice (Obbard et al., 2014). Within these environments, plastic debris of all sizes, has been reported at the sea surface (Cózar et al., 2014), suspended in the water column (Lattin et al., 2004), sediments (including those in the deep sea) (Fischer et al., 2015; Van Cauwenberghe et al., 2013; Woodall et al., 2014) and beaches (Nelms et al., 2016).

Macroplastic has spread to unhabituated areas, such as Antarctica, where research by Barnes et al (2010) found a plastic cup and two fishing buoys in the Durmont D'Urville and Davis seas, as well as two pieces of plastic packaging and a fishing buoy in the Amundsen Sea (Barnes et al., 2010). Plastic debris has also been found in freshwater environments showing that this issue is not limited to the marine environment (Ivleva et al., 2017; Mani et al., 2016; Wagner et al., 2014).

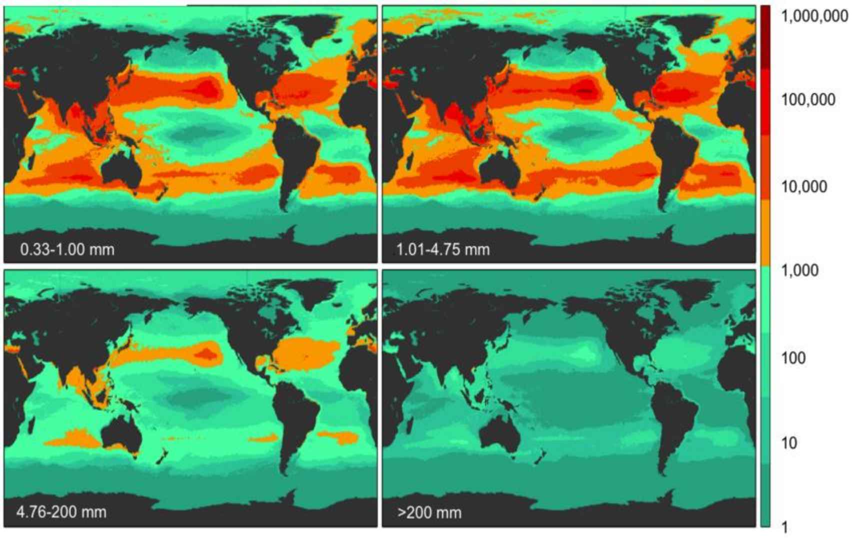

Figure 1.3. *Plastic contamination in the oceans. A model result for a global count density in four size classes of plastic. The combined data from 24 sampling missions with oceanographic computer modelling, the global distribution of plastic particles was predicted in specific size classes. Macroplastic is considered >5 mm, so is displayed by the bottom two tiles. Credit; Eriksen & Lebreton (Eriksen et al., 2014).*

1.4.1 Floating and Suspended Plastic Debris

Rivers can transport considerable quantities of plastic (micro - macro size) to the oceans and some of this debris can travel from locations far inland. In rivers, microplastic concentrations have been found to vary along and across the river, reflecting various sources such as waste water treatment plants, tributaries and weirs (Claessens et al., 2011; Klein et al., 2015; Mani et al., 2016). Once released into the environment, and due to their durability, plastic debris has the potential to become widely dispersed via wind and currents (Faure et al., 2015; Lambert et al., 2014; Ryan et al., 2009).

At the water surface, smaller pieces of plastic present lower rise velocities and are more susceptible to vertical transport (Reisser et al., 2015; Song et al., 2014). Some polymers such as polyvinyl chloride (PVC), and polyethylene terephthalate (PET) are denser than water and are more likely to sink, while polyethylene (PE), polypropylene (PP) and polystyrene (PS) are more likely to float. However, microplastics typically accumulate fouling from micro-organisms as well as sediment particles on their surface. Over time this increases their apparent density causing even some of the less dense polymers to sink (Zettler et al., 2013). Hence, the sea bed could be the most likely long-term place for the accumulation of plastic debris.

Floating debris can be transported by wind and currents at the sea surface but may eventually sink to the seafloor, be deposited on the shore or degrade over time (Andrady, 2015). Hence, the sources of floating marine debris in the oceans can be difficult to identify, given the persistence and potential for long-range transport of lightweight buoyant materials (Ryan et al., 2009).

Floating macrolitter, which includes plastics, is typically monitored by visual observation from ships. Visual sightings of macroplastics from ship-based observers have been reported since the 1970s (Venrick et al., 1973), and have proved to provide useful information about litter densities and how these compare between regions and over time. An example being in 2009; research from Titmus and Hyrenbach (2011) observed the surface water from the American west coast to the North Pacific subtropical gyre and back to the coast (Titmus and Hyrenbach, 2011). This provided data during 74 hours of observation corresponding to a transect length of 1 343 km. A single observer at 10 m above sea level recorded a total of 3868 pieces, of which 90 % were fragments and 96 % of these were plastic. Eighty-one percent of the items had a size of 2–10 cm, 14 % of 10–30 cm and 5 % of >30 cm. Their research found that the abundance of debris increased towards the centre of the gyre.

Visual observations from vessels are constrained by an inability to detect smaller fragments (<20 mm) and to retrieve the observed items for further analysis (Titmus and Hyrenbach, 2011). There has been an effort to standardise the observational methods used in order to reduce potential bias in the data. Factors such as sea state, elevation of the observation position and ship speed can all contribute to variations in the number of items measured (UNEP, 2016). A simple methodology has been proposed that should greatly improve the robustness of observations, allowing a more coherent picture for the distribution of floating plastic objects (Ryan, 2013). This takes into account the minimum size of items counted, the distance of items from the ship, the height of the observer above sea level and the position of the observer relative to the ship's bow wave. This proposed method would be effective as long as there was adequate training for such a method.

Aircraft and satellite observations may also prove useful; for example, in the aftermath of natural disasters, such as the 2011 Tōhoku earthquake tsunami in the North Pacific (NASA, 2012). Looking to the future, automatized approaches using digital imaging and image recognition techniques for the autonomous large-scale monitoring of plastic litter are being researched (Hanke and Piha, 2011).

1.4.2 Plastic Debris on the Seafloor

Plastic items that are a range of sizes are commonly observed, or collected, from the seafloor (Pham et al., 2014). Their ability to sink through the water column to the sea floor is determined by the plastics composition and environmental conditions (Galgani et al., 2015). Polymers denser than seawater (e.g. PVC) will sink, while those with lower density (e.g. PE and PP) will tend to float in water column. Biofouling of organisms on the plastic surface increases the weight of plastic objects, thus increasing the potential for them to sink (Lobelle and Cunliffe, 2011; Ye and Andrady, 1991). Degradation, fragmentation and the leaching of additives can also influence the density of objects and hence their distribution in the water column (Avio et al., 2017). As a result, plastic can dominate macro-debris on the sea floor.

Sea bed surveys of macro-debris, have been conducted with divers (Donohue et al., 2001; Nagelkerken et al., 2001), trawl surveys (Galgani et al., 2000; Moore and Allen, 2000; Tekman et al., 2017) and submersibles/remote-operated vehicles (Loakeimidis et al., 2015). The accumulation of large plastic items has also been found on the sea floor in the deep sea, where research by Galgani et al. (1996) found that plastic bags accounted for more than 70% of total debris off the French Mediterranean coast (Galgani et al., 1996). The amount of plastic litter is so great in some areas that initiatives have been started to clean the seabed with trawls, despite concerns about the ecological impacts of trawling (OSPAR Commission, 2007).

The geographic distribution of debris on the ocean floor is strongly influenced by hydrodynamics, geomorphology and human factors (Galgani et al., 1996; Pham et al., 2014). Moreover, there are notable temporal variations, particularly seasonal, with tendencies for accumulation and concentration of marine litter in particular geographic areas (Galgani et al., 1995). The fragmentation and degradation of plastics at depth is unknown, but accumulation of debris on the seafloor certainly began before scientific investigations started in the 1990s (Galgani et al., 2015).

1.4.3 Plastic Debris on Beaches

The majority of studies on marine plastic debris have focused on its occurrence in coastal waters and open ocean areas. However, plastic debris is now evident on beaches globally (Fig. 1.4). Researching macro-debris on beaches uses different approaches that sometimes lack detail (Hidalgo-Ruz and Thiel, 2015). These studies typically range from a local (Lee et al., 2013) to a regional scale (Bravo et al., 2009) and cover a broad temporal range. Information on sources, composition, amounts, usages, baseline data and environmental significance are often also gathered (Cordeiro and Costa, 2010; Debrot et al., 2013; Nelms et al., 2016).

Figure 1.4. *A beach clean organised by the Marine Conservation Society, UK (Source: Marine Conservation Society)*

The majority of studies investigating the abundance and distribution of plastic debris on beaches have used citizen science-based data (Eastman et al., 2014; Hoellein et al., 2014). An example being research from Nelms et al (2016); this research collected data over a decade (2005–2014 inclusive) using results from Marine Conservation Society (MCS) volunteers during beach litter surveys in the UK. Their results found that plastic was the main constituent of anthropogenic litter and the majority of traceable items originated from land-based sources. However, over the 10 year time there was no statistically significant change in the total quantity of litter detected (Nelms et al., 2016).

Factors influencing the accumulation of this debris in coastal areas include the shape of the beach, location and the nature of debris (Turra et al., 2015). Additionally, the abundance of stranded and drifting plastic debris (both macro and micro particles) along beaches and coastal areas is expected to increase with projected increases in sea level, wind speed, wave height, and altered rainfall conditions (Browne et al., 2015).

1.5 Impacts

There is a reasonably extensive evidence base relating to the harm caused by marine litter. This can have a range of negative impacts on commercial fisheries, maritime industries and infrastructure. It has also been found to affect a wide range of marine organisms; as a consequence of entanglement and ingestion (Gall and Thompson, 2015; Sutherland et al., 2010; Wang et al., 2016).

1.5.1 Impacts on Marine Organisms

Over 700 species of marine organisms have been reported to encounter plastic debris, which can result in severe physical harm and death, or more subtle effects on behaviour and ecological interactions (e.g. the ability to escape from predators or migrate) (Gall and Thompson, 2015). It is likely that there are also a range of sub-lethal effects that have not yet been recognized.

Impacts of plastic vary according to the type and size of the debris, and can occur at different levels of biological organization in a wide range of habitats (Browne, 2015). Encounters between plastic litter and organisms can negatively affect individuals and a substantial proportion of some populations can be contaminated with plastics. For example, over 40% of sperm whales beached on North Sea coasts had marine litter including, ropes, foils and packaging material found in their gastro-intestinal tract (Unger et al., 2016). Also, over 95% of the population of norther fulmars (*Fulmar glacialis*) may contain plastic litter in some European waters (Van Franeker et al., 2011).

Over 330 species (50 marine mammals) have been found to become entangled or ingested plastic debris (Kühn et al., 2015). However, evidence of harm from entanglement is easier to observe and report than ingestion. This is because ingestion typically only becomes apparent when the carcass of an animal opens; either as a result of dissection or decomposition. It has been reported from UK surveys that there is an incidence rate of entanglement between 2-9% for some populations of seabirds and marine mammals (Werner et al., 2016).

The ingestion of meso- or macroplastic litter has been reviewed for numerous marine species; particularly mammals, birds and turtles (Derraik, 2002; Gall and Thompson, 2015; Gregory, 2009; Kühn et al., 2015; Laist, 1997). Studies have also shown that both freshwater invertebrates and fish ingest microplastic (Imhof et al., 2013; Phillips and Bonner, 2015). The potential for ingestion is greater with pieces in the microplastic size range since their small size makes them readily accessible for ingestion by a wide range of organisms (Davison and Asch, 2011) including whales, fishes, mussels, oysters, shrimps, copepods and lugworms, (Cole et al., 2013; Ferreira et al., 2016; Lusher et al., 2015a, 2015b).

However, ingestion also depends on properties other than size including shape, density and colour. For instance, low-density (i.e. buoyant) microplastics are potentially more likely to be ingested by pelagic feeders and high-density microplastics by benthic feeders (Scherer et al., 2017; Wright et al., 2013). With very small particles, including those in the nano size range, there is also the potential for uptake across cell membranes, but little is known about any associated impacts (Koelmans et al. 2014). Organisms at lower trophic levels can ingest and accumulate microplastic particles, and it has been shown that microplastics can transfer between trophic levels in the food-web (Watts et al., 2014). Ingestion has shown to lead to physical effects that include physiological stress responses and even signs of tumour formation (Rochman et al., 2013).

There are also concerns about the potential for ingestion to facilitate the transfer of chemicals to marine life (Bakir et al., 2014). Hydrophobic organic pollutants readily sorb onto plastics, and can accumulate at concentrations several orders of magnitude higher than in seawater (Mato et al., 2001). These chemicals can then be released to organisms upon ingestion. However, modelling estimates indicate the amount of chemical transfer from water to organisms via plastic is probably not a major pathway leading to harm (Bakir et al., 2016).

Additive chemicals incorporated into plastic products at the time of manufacture may also transfer to marine organisms upon ingestion (Tanaka et al., 2013). These chemicals are intentionally added during the manufacture or processing; for example to enhance the plastics durability and corrosion resistance (Andrady, 2016) or act as stabilizers, plasticizers or flame retardants. Some additives, such as plasticizers, are used at high concentrations (10–50%) to ensure the functionality of the product. Degradation of plastic containing these additives may result in the additives leaching out and becoming bioavailable to organisms (Oehlmann et al., 2009; Talsness et al., 2009). Although, there is currently little evidence of harmful effects associated with the release of additives from plastic litter in the environment.

A further source of concern is colonization of organisms on plastic debris; species found on plastic debris can differ from the free-floating microbial communities in the oceans (Zettler et al., 2013). For example, microplastics collected in the surface waters of the North Atlantic were colonized by a variety of organisms including bacteria, cyanobacteria, diatoms, ciliates and radiolaria (Zettler et al., 2013). As plastics have been reported to travel over long distances, they may contribute to the dispersal of alien or invasive species (Barnes, 2002).

1.5.2 Impacts on Maritime Industries, Tourism and Human Health

The presence of litter in the marine environment presents an aesthetic issue with economic repercussions for the tourist industry, a hazard for numerous marine-industries (e.g. shipping, fishing, energy production, aquaculture) and entanglement/damage of maritime equipment (e.g. propellers on vessels) (Barnes et al., 2009a; Derraik, 2002). It is also considered that litter within the marine environment is unsightly and detracts from the inherent value of the world's coastlines, which can further impact human well-being and socio-economic activities (Fig. 1.5) (Axelsson and van Sebille, 2017). As discussed, plastic is the main constituent of marine anthropogenic litter, due to its high persistence within the marine environment (Andrady, 2015; Barnes et al., 2009a; Jambeck et al., 2015; Poeta et al., 2014; Thompson et al., 2009a).

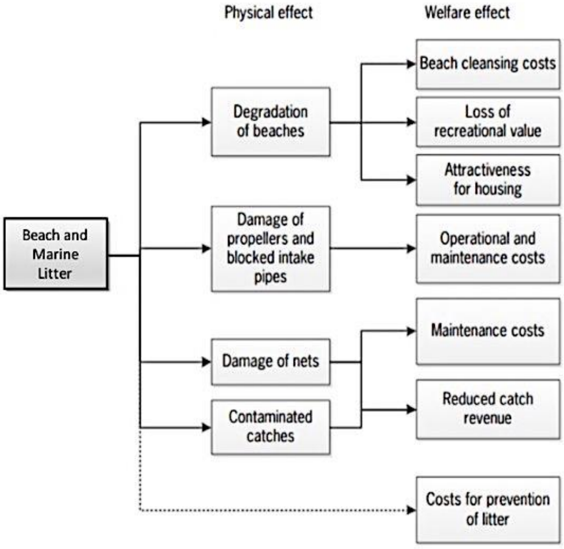

Figure 1.5. *Impacts of beach and marine litter on socio-economic activities; Adapted from Reinhard et al. 2012 (Reinhard et al., 2012).*

It has been reported that beach choice is more strongly determined by clean, litter-free sand and seawater than by safety (Tudor and Williams, 2006). For example, 85% of 1000 residents and tourists said they would not visit a beach with an excess of two litter items per metre (Ballance et al., 2000; Hastings and Potts, 2013). A study in Cape Town estimated that a loss in the standards of cleanliness on the beaches would result in a 97% loss in the value of those beaches (Ballance et al., 2000). It was also found that in Geoje Island, Korea, marine debris cost the island US \$29-37×10^6 in tourism losses following an extreme rainfall event in 2011 (Jang et al., 2014).

There is also emerging evidence that even small quantities of litter on beaches can have a negative effect on human well-being (Wyles et al., 2016). Wyles et al. (2015) found that the restorative psychological benefits ordinarily experienced by people visiting the coast were undermined by the presence of relatively small quantities of litter (Wyles et al., 2015).

Cleanup programs for anthropogenic litter can be ineffective and expensive. The total cost of removing litter of all types from 34 UK harbors was estimated at approximately £236 000. Based on this, it was estimated that marine litter costs the ports and harbour industry in the UK approximately £2.1×10^6 each year (Mouat et al., 2010). Very limited research has therefore been conducted into the costs of marine litter removal and estimates tend to be based mostly on anecdotal evidence. Research in 2000 found that 56 UK local authorities spent a total of £2 197 138 a year on beach cleansing, taking into account the cost of collection, transport, disposal charges, workforce, equipment and administration (Hall and KIMO (Organization), 2000).

Fishing can also be impacted by macroplastic debris (Fig. 1.6). Any subsequent decreases in the abundance of stocks would directly affect the fisheries economy (Axelsson and van Sebille, 2017). However, the most important impact of macroplastic debris on fisheries is from ghost fishing from Abandoned, Lost or Otherwise Discarded Fishing Gear (ALDFG). Ghost fishing occurs when passive gears such as gillnets, trammel nets, wreck nets, pots and traps are lost or discarded. These then continue to catch commercially important species of fish and crustaceans as well as non-commercial species of fish and crustaceans, birds, marine mammals and turtles (Brown and Macfadyen, 2007). It has been estimated that there is an annual loss of 208 t of Antarctic toothfish (*Dissostichus mawsoni*) due to lost longlines (Webber and Parker, 2012). Another study in Australia found that collapsible trawl mesh pots could unintendedly catch 670 866 Blue swimmer crabs (*Portunus pelagicus*) per year.

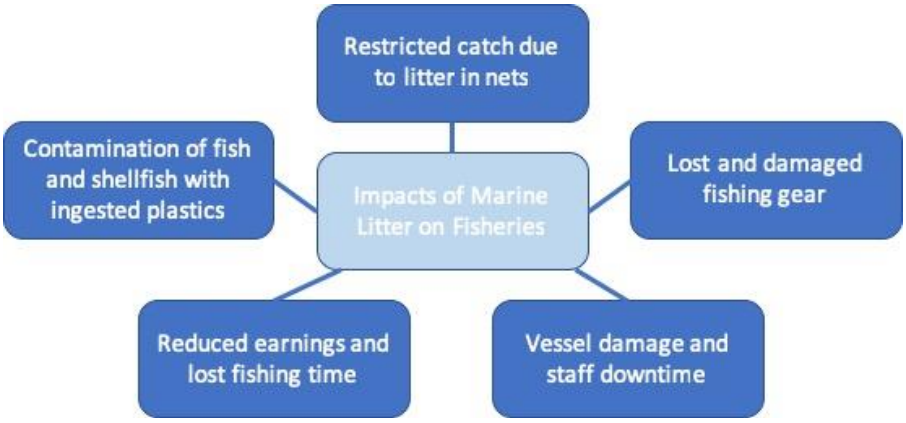

Figure 1.6. *Potential impacts of marine litter on fisheries; Adapted from Mouat et al. (2010) (Mouat et al., 2010).*

Additionally, plastic litter can damage vessels or present navigational hazards. It can lead to injury or death following loss of power, due to entangled propellers or blocked water intakes and collision with floating or semi-submerged objects, including (plastic) insulated shipping containers (Frey and DeVogelaere, 2013). For example, in 2005, the USA coastguard reported that collisions with submerged objects caused 269 boating incidents, resulting in 15 deaths and 116 injuries (U.S. Coast Guard, 2006). The seriousness of the potential consequences was highlighted by the sinking of the Ferry M/V Soe-Hae in 1993 which was, in part, caused by rope around the propellers, and resulted in 292 deaths (Cho, 2005). Clearly, the frequency of these negative impacts will increase in relation to increasing levels of contamination. Furthermore, there is potential of harm to due to entanglement when swimming and diving.

1.6 Conclusion

Once in the ocean, plastic debris has the potential to travel considerable distances from its original location. The debris is persistent, accumulating and can potentially impact marine organisms, maritime industries, tourism and human health. One of the major challenges in addressing the issues surrounding marine plastic debris is the diverse nature of plastic products and the many routes by which they can enter the environment. To help manage and reduce emissions it is essential to better understand the relative importance of these sources and to assess regional variation. We should focus on minimising direct inputs of microplastic and reduce the quantity of larger items of litter entering the environment since they will ultimately fragment into microplastics.

Therefore, understanding the different sources of plastic into the marine environment is critical as human population and usage of plastic continues to rise. The following thesis analyses four common plastic containing products (facial scrubs, clothes, wet wipes and carrier bags) to identify their potential input and risk to the aquatic environment.

1.7 Aim of the Thesis

The overall aim of this thesis was to identify and analyse different sources of plastic entering the marine environment.

The specific objectives were to:

1. Review the current knowledge of plastic marine debris
2. Research four different sources of plastic marine debris and their characteristics
3. Discuss the potential impact that each source could cause
4. Analyse degradation properties

1.8 Thesis overview

This thesis comprises six chapters. Each chapter investigates different plastic products and identifies how they could be an important source of microplastics to aquatic habitats.
A description of each is given below.

Chapter 2 characterises, quantifies and then investigates the sorptive properties of plastic microbeads that are used as exfoliants in cosmetics.

Chapter 3 examined the release of fibres from polyester, polyester-cotton blend and acrylic fabrics when being laundered under various conditions of temperature, detergent and conditioner.

Chapter 4 analyses four different baby-wipe brands labelled as flushable and one baby-wipe labelled as non-flushable. This was to further understand their relative rates of degradation in the marine environment.

Chapter 5 tests the breakdown of carrier bags made out of oxo-/biodegradable, compostable and conventional plastic formulations over three years in three different environments (marine, soil and open-air).

Chapter 6 discusses the solutions to marine litter and the broader implications.

Accumulatively, these chapters develop our understanding of how plastic can enter our marine environment, and in substantial quantities, from sources not often considered.

Chapter 2

Characterisation, Quantity and Sorptive Properties of Microplastics Extracted from Cosmetics

A version of this chapter is available online as:

Napper I.E., Bakir A., Rowland S.J., Thompson R.C. Characterisation, quantity and sorptive properties of microplastics extracted from cosmetics. Mar Pollut Bull 2015;99:178–85.

The Sorptive Properties research was conducted by Bakir, A.

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 understand how microbeads could also 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.

2.0 Characterisation, Quantity and Sorptive Properties of Microplastics Extracted From Cosmetics

2.1 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 <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 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).

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 per day, indicating that the U.S. population may be emitting an estimated 263 tonnes per yr 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).

2.2 Methods

2.2.1 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.

2.2.2 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 µm, to 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:

$$\text{i) } Vt = \frac{Mt}{D} \qquad \text{ii) } V(avg\ particle) = \frac{4}{3}\pi r^3 \qquad \text{iii) } N = \frac{Vt}{V(avg\ particle)}$$

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.

2.2.3 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 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 hours (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:

(iv) $Kd = [q_e]_{solid} / [C_e]_{aq.}$

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}).

2.2.4 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).

2.3 Results

2.3.1 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 2.1); 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).

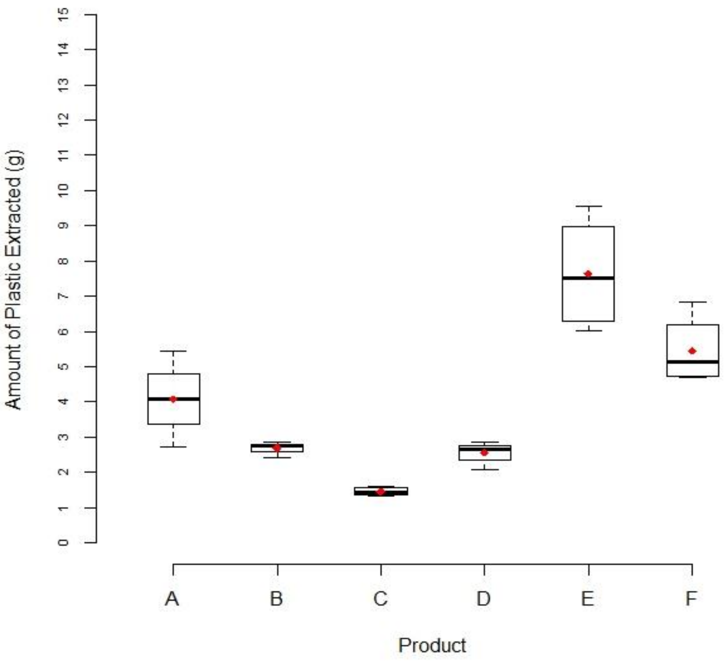

Figure 2.1. *Total mass of plastic microbeads extracted from six facial scrubs (A-F) per 100 mL. Diamond symbol indicates* $\overline{x}$ *(n=4). The tails show both the maximum and minimum mass obtained, and the box represents the upper and lower quartiles. There were significant differences between the amount of microplastic in each of the products* ($p < 0.05$).

2.3.2 Size-Frequency Distributions

Microplastics from the facial scrubs showed polydispersed size ranges, each with logarithmic bimodal distributions (Fig 2.2). Product B had the largest size range (10µm to >2000 µm), whereas product A was the most homogenous, ranging from 8µm to 56µm, 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).

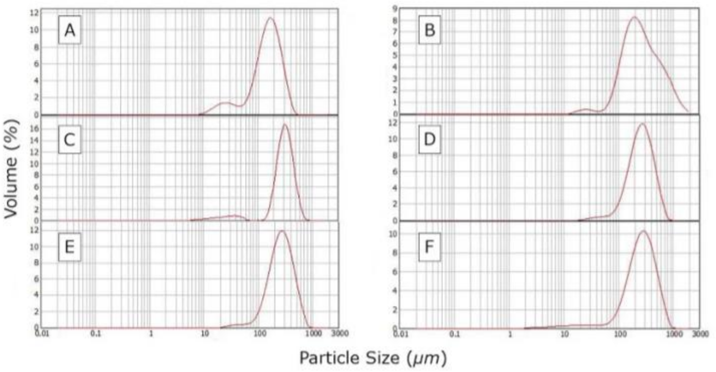

Figure 2.2. *Particle size distribution of PE microbead particles extracted from six facial scrubs (A-F). Determined using a Malvern Mastersizer 2000, laser particle sizer.*

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 (Fig. 2.3). 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.

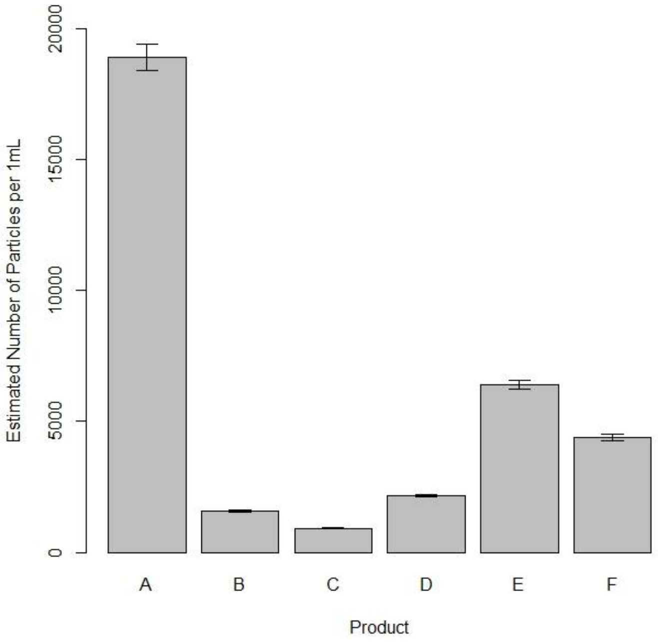

Figure 2.3. *Estimates for the number of PE microbead particles in six brands of facial scrubs per 1ml. Calculated using data from the volume weighted mean (n = 3, ±SD; correlating to the spread of the different amounts of particles calculated for high, medium and low density PE).*

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. 2.4). 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).

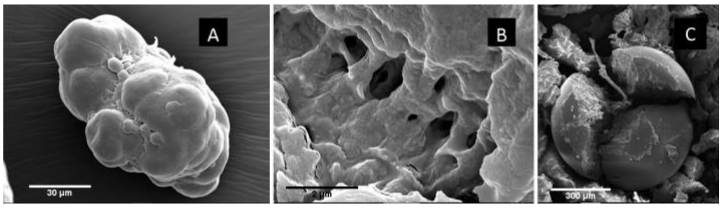

Figure 2.4. *A-Scanning Electron Microscopy (SEM) image of a typical rough facial scrub plastic microbead particle (9000X magnification). B- SEM image of surface microbead topography (16000X magnification). C- SEM image of a broken smooth spherical plastic microbead from 'product F' (900X magnification).*

The colour of microplastics used in the different products also varied (Table 2.1). 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.

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

Table 2.1. *Colour of microplastics found within six facial scrub products.*

2.3.3 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. 2.4). 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. 2.5). Sorption capacity for all plastics was significantly higher for DDT compared to Phe ($p < 0.05$, Table 2.2).

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

Table 2.2. *Recovery (%) of phenanthrene (Phe) and DDT following sorption experiments onto PVC and PE (average values displayed, n = 3)*

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.

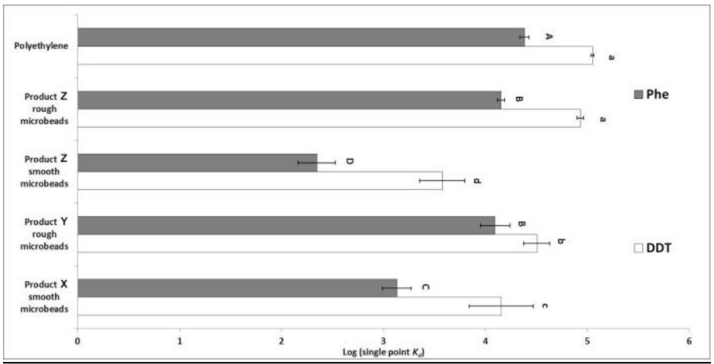

Figure 2.5. *Single point distribution coefficients (K_d) for the sorption of a mixture of phenanthrene (Phe) and DDT onto PE particles and rough and smooth PE-microbeads extracted from cosmetic products (n=3, ± SD). For each contaminant, treatments with the same letters (A-C for Phe and a-d for DDT) were not significantly different ($p < 0.05$).*

2.4 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). 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). This figure is likely to be higher if male useage was also taken into account. 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 $^{-1}$.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 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.

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).

2.5 Conclusion

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.

Chapter 3

Release of Synthetic Microplastic Fibres from Domestic Washing Machines - Effects of Fabric Type and Washing Conditions

A version of this chapter is available online as:

Napper I.E., Thompson R.C. Release of synthetic microplastic plastic fibres from domestic washing machines: Effects of fabric type and washing conditions. Mar Pollut Bull 2016;112:39–45.

Abstract

Washing clothes made from synthetic materials have 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 11.9–17.7 µm in diameter, and 5.0–7.8 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 6kg 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.

3.0 Release of Synthetic Microplastic Fibres from Domestic Washing Machines - Effects of Fabric Type and Washing Conditions

3.1 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 (Eerkes-Medrano et al., 2015; Sanchez et al., 2014). Laboratory studies indicate the potential for physical harm to biota from the result of ingestion (Wright et al., 2013b). 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., 2013b; Koelmans et al., 2014, 2013; Rochman et al., 2013). Encounter rate, as well as polymer type and any associated chemicals (sorbed or additives), will influence the potential for effects in the environment (Bakir et al., 2014a, 2012; Koelmans et al., 2013; Teuten et al., 2007), 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 (Obbard et al., 2014; Woodall et al., 2014). This indicates plastics ubiquity and the need for further understanding about the potential environmental consequences

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 fibres have been used to supplement cotton, wool and linen in textiles for more than 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 (Dris et al., 2015; Leslie et al., 2013). 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.

3.2 Methods

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. 3.3), 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, 20cm X 20cm 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 2L of water and filtered again over Whatman N°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, 20ml bio-detergent present (contains enzymes), 20ml non-bio detergent present); Factor four, conditioner (2 levels; 20ml conditioner absent or present). Factors gave a total of 36 treatments (Fig. 3.1).

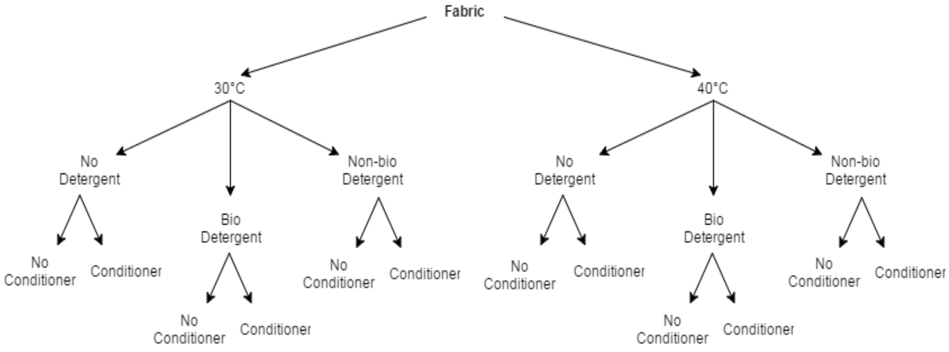

Figure 3.1. *Experimental design showing Factors used for each fabric type (acrylic, polyester, polyester-cotton blend).*

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 hour 15 minutes 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 minutes 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 minimised 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 averge length of a fibre and r is the average radius of a fibre.

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 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.

3.3 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. 3.3). 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. 3.2), 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.

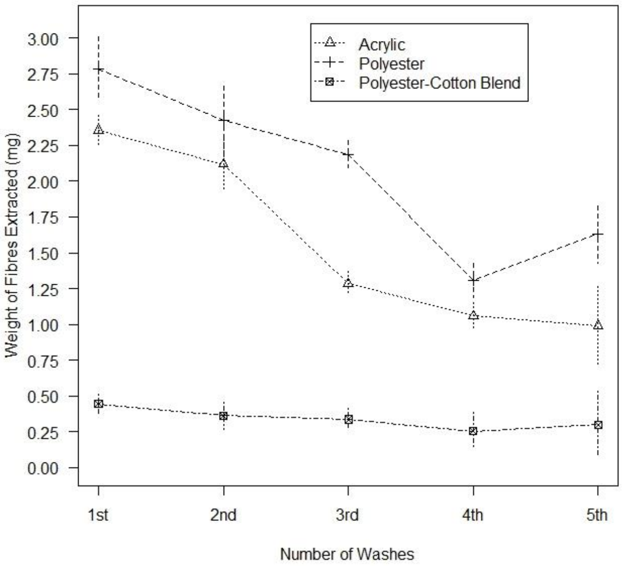

Figure 3.2 *Fibre loss from three fabrics (acrylic, polyester & polyester-cotton blend), over the first 5 washes. Data from the 5th wash was used in the analysis* (n = 4, ±SD).

While there was a consistent trend between fabric types, ANOVA revealed significant complex interactions between the 4 Factors (Table 3.1). Focussing on the type of fabric, polyester-cotton blend was consistently found to shed fewer fibres than both the other fabric types, regardless of the differing treatments. This trend was consistent for all 12 relevant interactive effects, and was found to be significant for 9 out of these 12 interactions (Table 3.2). 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 3.2).

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**
FabricXTemp	2	0.02	0.33	0.72
FabricXCond	2	0.12	1.88	0.16
FabricXDeter	4	0.20	3.13	**0.02**
TempXCond	1	0.15	2.28	0.13
TempXDeter	2	0.13	2.09	0.13
CondXDeter	2	0.58	9.00	**0.00**
FabricXTempXCond	2	0.06	0.86	0.43
FabricXTempXDeter	4	0.06	1.00	0.41
FabricXCondXDeter	4	0.33	5.05	**0.00**
TempXCondXDeter	2	0.64	9.91	**0.00**
FabricXTempXCondXDeter	4	0.38	5.95	**0.00**
Residual	108	0.06		
Total	143			

Table 3.1. *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).*

a) FABRIC				**b) DETERGENT**			
Factors			**Order**	**Factors**			**Order**
30	C-	No Powder	**Blend<*Acr<*PE**	Acr	30	C-	**bio-NB-A**
30	C-	Bio	**Blend<*Acr-PE**	Acr	30	C+	**A-NB-bio**
30	C-	Non Bio	**Blend-PE-Acr**	Acr	40	C-	**A-NB-bio**
30	C+	No Powder	**Blend<*PE-Acr**	Acr	40	C+	**bio-NB<*A**
30	C+	Bio	**Blend<*PE-Acr**	Blend	30	C-	**bio-A-NB**
30	C+	Non Bio	**Blend<*Acr-PE**	Blend	30	C+	**A-bio-NB**
40	C-	No Powder	**Blend<*Acr<*PE**	Blend	40	C-	**A-bio<*NB**
40	C-	Bio	**Blend<*PE<*Acr**	Blend	40	C+	**A-NB-bio**
40	C-	Non Bio	**Blend-Acr<*PE**	PE	30	C-	**bio-NB<*A**
40	C+	No Powder	**Blend<*PE<*Acr**	PE	30	C+	**A-bio-NB**
40	C+	Bio	**Blend-Acr<*PE**	PE	40	C-	**bio<*A<*NB**
40	C+	Non Bio	**Blend<*Acr-PE**	PE	40	C+	**A-NB-bio**

c) TEMPERATURE				**d) CONDITIONER**			
Factors			**Order**	**Factors**			**Order**
Acr	C-	No Powder	**40-30**	Acr	30	No Powder	**C-A**
Acr	C-	Bio	**30<*40**	Acr	30	Bio	**A<*C**
Acr	C-	Non Bio	**30-40**	Acr	30	Non Bio	**A-C**
Acr	C+	No Powder	**30-40**	Acr	40	No Powder	**A<*C**
Acr	C+	Bio	**40<*30**	Acr	40	Bio	**C-A**
Acr	C+	Non Bio	**40-30**	Acr	40	Non Bio	**C-A**
Blend	C-	No Powder	**40-30**	Blend	30	No Powder	**A-C**
Blend	C-	Bio	**40-30**	Blend	30	Bio	**A-C**
Blend	C-	Non Bio	**30<*40**	Blend	30	Non Bio	**A-C**
Blend	C+	No Powder	**30-40**	Blend	40	No Powder	**A-C**
Blend	C+	Bio	**30-40**	Blend	40	Bio	**A<*C**
Blend	C+	Non Bio	**30-40**	Blend	40	Non Bio	**C<*A**
PE	C-	No Powder	**40-30**	PE	30	No Powder	**C<*A**
PE	C-	Bio	**40-30**	PE	30	Bio	**A-C**
PE	C-	Non Bio	**30<*40**	PE	30	Non Bio	**A<C**
PE	C+	No Powder	**40-30**	PE	40	No Powder	**C-A**
PE	C+	Bio	**40-30**	PE	40	Bio	**A<*C**
PE	C+	Non Bio	**40-30**	PE	40	Non Bio	**C<*A**

Table 3.2. *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).*

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-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. 3.3).

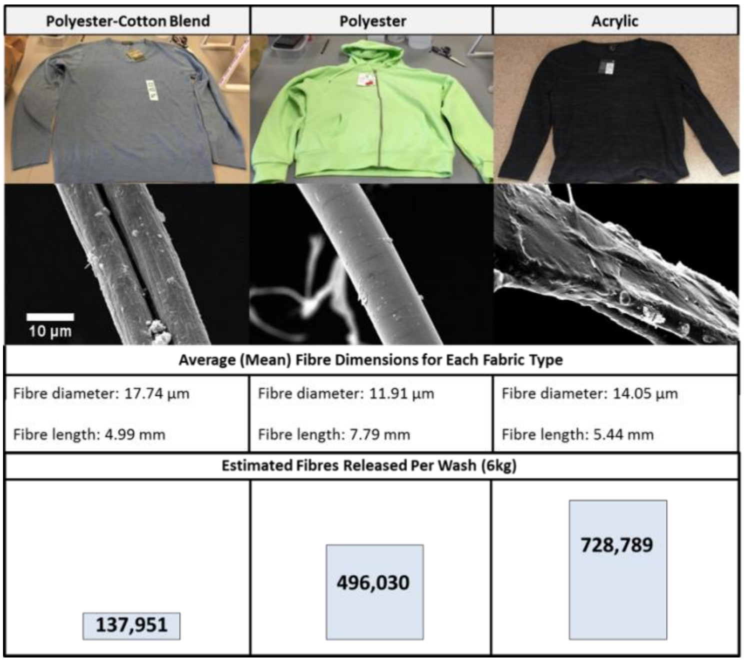

Figure 3.3. *Images to show the original garments (each representing a different fabric), and a scanning electron microscopy image (SEM) of a typical fibre from each fabric (the scale bar is consistent for all images – 2500X magnification). Fibre diameter and length are averages taken from fibres of each garment type. 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 6kg).*

The size parameters of the fibres varied in size, and this affected how many fibres could be found in a certain mass. Focussing on 1mg and dry fibre weight, acrylic fibres were on average 14.05 µm in diameter and 5.44 mm in length, giving an average of 1005 fibres/mg. Polyester fibres were on average 11.91 µm in diameter, but were longer at 7.79 mm, resulting in around 841 fibres/mg. 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 525 fibres/mg

3.4 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 (Chiweshe and Crews, 2000; Hussain et al., 2008). 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 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 increse 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 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., 1998a; 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 can be made of a stainless steel mesh, with various hole diameters, which collect fibres but allow water to pass through (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. Currently, the most common method of removal of fibres collected through these filters will be via household 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)

3.5 Conclusion

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.

Chapter 4

The Properties and Degradability of Single-Use Disposable Wipes (Baby-Wipes)

Abstract

Disposable wipes (wet-wipes) are frequently flushed down the toilet in error often due to unclear labels on packaging and a misconception that they are flushable like toilet paper. Additionally, flushable wipes may still also cause blockages. Subsequently, they enter the sewage system and could potentially enter the marine environment, adding to the accumulation of marine litter.

There are currently no standardised criteria to measure whether a product can be flushed, and the degradability of these products is still unclear. We analysed four different disposable wipe brands labelled as flushable and one labelled as non-flushable. The results help to identify: 1) indicate relative rates of disintegration in the marine environment; 2) how the products disintegrate; 3) how environmental conditions influence their breakdown.

Wipes were tested in both laboratory and environmental conditions over a period of 15 weeks. The flushable wipes were found to contain 2 different fibre structures, and both remained throughout the disintegration process. Over a period of 15 weeks, for samples exposed in lab conditions, the addition of nutrients appeared to accelerate fragmentation for flushable wipes. However, the main wipe structure remained for those exposed to seawater or autoclaved seawater. Within environmental conditions, non-flushable wipes were found to persist in the marine environment after 15 weeks, but flushable wipes were found to disintegrate within 3 weeks, but the degradability of the resultant fragments was not assessed. As a pilot study, our results suggest that wet wipe structure stays intact longer than anticipated.

4.0 The Properties and Degradability of Single-Use Disposable Wipes (Baby-Wipes)

4.1 Introduction

There are numerous innovative brands of toilet paper, biodegradable sanitary protection, disposable nappies and disposable wipes. Some of these have been designed to be disposed of via the toilet to the sewage system, providing a discrete and convenient waste option. The disposal of these domestic items using the toilet as a 'rubbish bin' is habitual in many countries, including the U.K. and U.S.A. (Einsohn, 2000). This stems from the historical link associating health and hygiene risks with human waste. However, there is currently little knowledge of the behaviour of these products after they are flushed.

Disposable wipes' function is typically for cleaning purposes only and they are subjected to light rubbing or friction, in order to remove dirt or liquid from a surface. Consumers want these wipes to absorb, retain or release dust or liquid on demand in order to complete this function. The vast global use of these disposable wipes is due to the many benefits they provide. A disposable wipe is quicker and easier to use than the alternative of dispersing a liquid and then using another cloth/paper towel to clean or remove the liquid. They are also clean, reduce cross contamination and can be pre-cut to convenient sizes and shapes. Pre-cut disposable wipes provide higher levels of convenience and mobility (Rengasamy, 2014; Sahu, 2012).

Disposable wipes are popular and cheap consumer items, which are ready for disposal after a single use,. They can then be further classified into dry wipes and wet wipes (Rengasamy, 2014). These products make a broad market in three main categories: personal care wipes, household cleaning wipes & industrial cleaning wipes (Sahu, 2012). The main category from the above list is personal care wipes which are now widely used in a variety of applications; such as for face and eye cleansing, make-up removal, treatment of oily skin, self-tanning lotions and baby-wipes.

Consumers are likely to consider the following when purchasing disposable wipes: cost, convenience, hygiene, performance, ease of use, time savings, safety/regulation and consumer perceived aesthetics (for example, softness or bulkiness in baby wipes). However, disposability is arguably one of the main factors. Therefore, many brands are now promoting that their wipes are flushable. Disposable flushable wipes have grown in popularity over the last decade and there is currently no legislation enforcing which products can and cannot be labelled as ‘flushable’ or as a ‘tissue’ (Wessex Water, 2016).

The word ‘tissue’ and ‘flushable’ leads the consumer to assume that the product is of natural material and can disintegrate rapidly; therefore, promoting an expectation that the product behaves like toilet paper when flushed. Disposable wipes can be made of different materials such as paper, tissue or nonwoven alternatives, with many wet wipes containing man-made plastic constituent parts (Rengasamy, 2014; Wessex Water, 2016). As consumers are using both flushable and non-flushable wet wipes, there are significant issues with: 1) users disposing non-flushable wipes incorrectly (Kim and Hergeth, 2012) and 2) the lack of clarity on the fate of flushable wipes in waste water treatment and the environment.

Disposable personal wipes, such as baby-wipes, are typically made from material that is nonwoven (Rengasamy, 2014). Nonwoven material is a manufactured structure of directionally or randomly oriented fibres, bonded by friction, and/or cohesion and/or adhesion (ISO 9092, 2011). Formation of a non-woven fabric is produced by suspending fibres in water to disperse them evenly. The suspension is poured onto a moving screen allowing the water to pass through and this leaves a fibre web, which is then dried to form the fabric (Rengasamy, 2014).

All kinds of fibres can be used to produce nonwoven fabrics (Kalabek et al., 2016). The selection of fibres is based on the following features: cost-effectiveness, the ease of manufacture and the desired end-use properties of the webs (Turbak, 1993). The commonly used fibres in non-woven products (such as disposable wipes) include natural fibres (cotton, jute, flax, wool), synthetic fibres (polyester (PE), polypropylene (PP), polyamide, rayon) and special fibres (glass, carbon, nano-fibres, bi-component, superabsorbent fibres) (Hongu et al., 2005; Kalabek et al., 2016; Turbak, 1993).

Two or more types of fibres are typically utilised. The fibres are usually blended or mixed in order to improve performance properties of nonwovens, such as strength. The fibre blend or mix can be natural/natural, synthetic/synthetic, or natural/synthetic (Kalabek et al., 2016; Purdy, 1983). Man-made fibres are the most widely used in the nonwoven industry. Owing to impurities and higher costs, natural fibres are of minor importance for the production of nonwovens (Kalabek et al., 2016).

This means that even when disposable wipes that are flushed eventually break up, the nonwoven fabric that makes the wipe has the potential to end up passing synthetic material (such as plastic) into the environment (Wessex Water, 2016). This can be either onto farmland via removal in sewage sludge or into the water environment via sewage effluent (Murphy et al., 2016).

The time-frame for disposable flushable wipes to breakdown is still not known. The wipes undergo various degradation processes such as physical disintegration by turbulent forces, hydrolysis of soluble material, and biodegradation of dissolved materials. Physical disintegration also depends on solid characteristics such as size, mechanical strength, and density of the solids (Karadagli et al., 2012).

Their presence in the environment has been increasing. During the Marine Conservation Society (MCS) 2016 Great British Beach clean, an average of 14.1 wipes for every 100m of our coastline was found. MCS has reported a 700% increase of disposable wipes found on U.K. beaches over the last decade (MCS, 2016). Furthermore, regardless of whether the wipes are labelled as ‘flushable’ or not, it is predicated that the majority of wipes on the market do not disintegrate quickly enough to avoid clogging the sewage pipes and pumps. This not only reduces the operational safety and stability of the wastewater system, but also puts additional financial cost on the operator (Mitchell et al., 2017; Wessex Water, 2016).

A major UK water company (Wessex Water) cleared more than 13,000 blockages from the public sewer network and a further 5,000 blockages on customers’ privately-owned drains (pipes just serving one property) in 2015. They estimated that within their operating area more than 30,000 blockages are cleared each year. Analysis of the data shows that 80% of all sewer blockages are as a result of customers disposing of inappropriate items down the toilet or sink; often referred to as sewer misuse. Of these blockages, 70% are the result of wet wipes (Wessex Water, 2016). Non-flushable wet wipes may also be mistakenly disposed down the toilet.

For all wipe types, the share of personal care, household and industrial wipe products are about 62%, 23% and 15% respectively. Baby wipes accounted for 63% of personal care wipes in 2010 (Bell, 2011; Rengasamy, 2014). Therefore, our research focusses specifically on baby wipes as they are the most used wipe on the market. Baby wipes are used to cleanse and clean the sensitive skin of infants and adults. These are saturated with solutions ranging form gentle cleansing ingredients to alcohol-based ‘cleaners’. Baby wipes are typically sold in plastic tubs that keep the cloths moist and allow for easy dispensing (Sahu, 2012).

There is currently no peer reviewed research that analyses the breakdown of disposable wipes. Our research analysed the composition of both flushable and non-flushable baby wipes and tested them in environmental and laboratory settings. We investigated the time frame and the process of any disintegration. We also examined different factors which could affect this. From the results, we further discuss the impact disposable wipes have within the marine environment and we also discuss what the current legal and regulatory situation is relating to these products.

4.2 Methods

Four major brands of moistened disposable baby-wipes were chosen based on whether they were stated to be flushable. Three brands were marketed as flushable, and one brand stated to be non-flushable (which was chosen as a comparison). These baby wipes had a high prevalence in major supermarkets in and around Plymouth UK. 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, the flushable baby-wipes were labelled 1F-3F, and the non-flushable wipe 4NF. None of the baby-wipes stated that they were of plastic material from their ingredients list (including the non-flushable wipe).

10 wipes from each brand were dried at 30°C to a constant weight. Once dry, each wipe was weighed by Precisa 2200C weighing scales and the mean dry weight of a wipe recorded for each brand. This data was used for statistical testing and normal distribution was confirmed by using QQ plots. A one-way analysis of variance (ANOVA) was used to compare the dry weight of a wipe for each product type. Post-hoc Tukey tests were used to identify the specific significant effects. All statistical tests were performed in R ver. 3.4.1 (R Core Team 2017).

Fibres taken from each baby-wipe brand had their composition analysed 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 ATR library).

Samples were also taken from each brand, embedded in resin and stained with methylene blue to identify if they contained cellulosic material. Cross sections of the fibres in the resin were then cut off using a microtome. The sections were then exposed to 0.1% aqueous methylene blue for 15-20 minutes and then replaced with deionised water. The samples were then visualised under a LEICA M205C light microscope.

The disintegration of the baby-wipes was then tested in both laboratory and environmental conditions.

4.2.1 Environmental Setup

A whole wipe from each brand was then inputted into a hand-sewn pouch structure made of green high-density polyethylene (HDPE) mesh and nylon fishing wire; both materials were confirmed by FTIR. Each pouch included two HDPE sheets (30cmx30cm) sewn together. The HDPE mesh allowed exposure to external environments due to the continual holes (1 mm X 1 mm) provided by the material. Each compartment was slack to provide any movement from external factors. Pouches were placed in orange HDPE mesh bags that were then sealed (Fig. 4.1). The bags were monitored daily for 10 weeks.

Figure 4.1. ***Left*** *– Whole baby-wipes which have been sewn into mesh pouches and placed in an orange high-density polyethylene bag before deployment into the marine environment (Queen Anne's Battery Marina; Plymouth).* ***Right*** *– Extracted baby-wipes after 1 week exposure in the marine environment. The brand names on the petri dishes have been removed.*

Samples were submerged at Queen Anne's Battery Marina (50°36'48.4"N, -4°12'96.5"W). The samples, connected to permanent panels, were attached to a plastic beam and submerged to a depth of 1m. 3kg weights were connected on each side of the beam to provide stability and assurance of depth.

For each disposable baby-wipe brand, there were 10 planned sample dates (1 every week in a 10-week period) and 4 replicates for each sample date. They were deployed on the 12th June 2017. Collected samples were then dried at 30°C to a constant weight. These samples were taken and visualised by scanning electron microscopy (JEOL, 7001F); images taken were used to visually assess the breakdown of the material by analysing its topography for any cracks or fragmentation.

4.2.2 Laboratory Setup

The laboratory conditions included 3 different variations of media that single wipes would be exposed to; autoclaved seawater, seawater and seawater plus additional nutrients. Seawater was collected from Queen Anne's Battery Marina (50°36'48.4"N, -4°12'96.5"W). The additional nutrients added were an inoculation of nitrogen, phosphorous and potassium at a ratio of 10.6:4.4:1.7 respectively. This ratio was selected to aid microbial growth. 1L of each media type was put into a tank that also had aeration tubes (seawater plus additional nutrients had 1ml of nutrients substituted into media). The aeration tubes in each tank helped to circulate the water (providing movement) and gave a constant oxygen supply.

To stop foreign contaminants entering into the tanks and loss from condensation, they were enclosed with a covering which was further held down with large Pyrex sheeting. These tanks were kept in a 30°C controlled temperature room, which had 12 hours of light per day. The controlled temperature room was cleaned on a daily basis and had limited access to others to avoid potential foreign contamination within the tanks. The pH of the media was analysed using a SevenExcellence pH Meter (METTLER TOLEDO). The salinity measured by a Digital Refractometer for Seawater Analysis (HANNA Instruments).

4 replicates from each baby-wipe brand were tested in the 3 different media types. Additionally, toilet paper and the baby-wipes exposed for a week in the marine environment were tested under the same parameters (seawater media only). Toilet paper and baby-wipes were exposed for one week in the marine environment to assess whether microbial communities within this natural setting aided the disintegration rate.

As a comparison to disposable wipes, 1.25 g of toilet paper was also used. This weight was chosen as it was equal to the mean dry-weight of the disposable baby-wipe brands.

Disposable baby-wipes that had a week exposure in the marine environment prior to lab analysis were also deployed at Queen Anne's Battery Marina. They had the identical set-up as the environmental section of this experiment. Fresh seawater was collected as the media for the exposed baby-wipes at the same time as retrieving them from the marine environment.

The samples were deployed on the 12th June 2017. Their disintegration was monitored, and samples were extracted at 5 and 10 weeks for closer examination. At 5 weeks, a 9 cm^2 sample was taken from the main panel of each wipe. At 10 weeks, 10ml samples were taken from the media itself to collect any released fibres. The media was then filtered, and the fibres collected from a nylon CellMicroSieve™ (Fisher Scientific), with 25 µm pores. Any collected fibres were then dried at 30°C to a constant weight. Similarly, to the environmental deployed baby-wipes, these samples were analysed by scanning electron microscopy (JEOL, 7001F). The wipes were also examined on a daily basis over 15 weeks visually for signs of degradation and any changes recorded. These changes were recorded based on 1) visual deterioration of the wipe (e.g. any holes), 2) reduction in size and 3) the number of fibres clouding the water which had broken off of the main panel of the wipes.

4.3 Results

All of the disposable baby-wipe products provided minimal technical information or production details, but they all promoted that they were hypoallergenic and dermatologically tested. Products 1-3F all stated that they were biodegradable and flushable, and included this function in writing on the main marketing section of the packaging. Product 4NF stated that the product was not to be disposed of by flushing by displaying two small logos; 'do not flush' and 'tidy man' (Fig. 4.2). Products 2F and 3F also displayed Forest Stewardship Council (FSC) 'tick tree' logo, both stating they contained recycled wood. This logo is displayed on a wood or wood-based product for assurance that the product is made with, or contains, wood that comes from FSC certified forests or from post-consumer waste. There are three types of FSC label: 100%, FSC Mix or FSC Recycled (FSC, 2017).

Figure 4.2. *The Forest Stewardship Council (FSC) symbol is displayed on a wood or wood based product for assurance that the product is made with, or contains, wood that comes from FSC certified forests or from post-consumer waste (FSC, 2017). The 'Do Not Flush' (bottom left) and 'Tidy Man' (bottom right) symbols to be used on the packaging of products not designed to be flushed (INDA and EDANA, 2013).*

When comparing the different brands, the mean dry wipe weight was similar for each product type: 1F 0.99g; 2F 1.17g; 3F 1.20g (Fig.4.3). Product 1F had a weight outlier at 0.91g. The non-flushable wipe product, 4NF, was heavier in mean dry weight at 1.66g. It also showed the most variation in weight. The weight of wipes varied significantly between products (one-way ANOVA, *p* = .000). Post-hoc Tukey tests identified that all product combinations were significantly different from each other (*p* = <.000), apart from 3F-2F (*p* = .682).

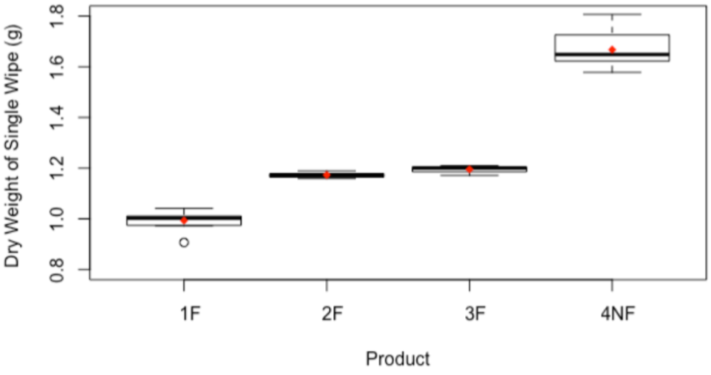

Figure 4.3. *The weight in grams of single baby wipes extracted from four different brands; 1F-3F (flushable) and 4NF (non-flushable). The y axis begins at 0.8 g. The thick black line is the median for each product and the diamond symbol indicates* $\bar{x}$ *(n=4). The tails show both the maximum and 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 < .05).*

The methylene blue staining on the baby wipe samples identified all of the products to contain cellulosic material (Fig.4.4). The flushable wipes (1F-3F) were heavily dispersed with cellulose. However, the non-flushable wet-wipe (4NF) had substantially less staining and therefore less cellulose overall. On closer inspection, the fibre cross sections from 4NF presented cellulose in interspersed small areas, surrounded by undyed non-cellulosic alternatives. This is similar to what the FTIR results reported for product 4NF.

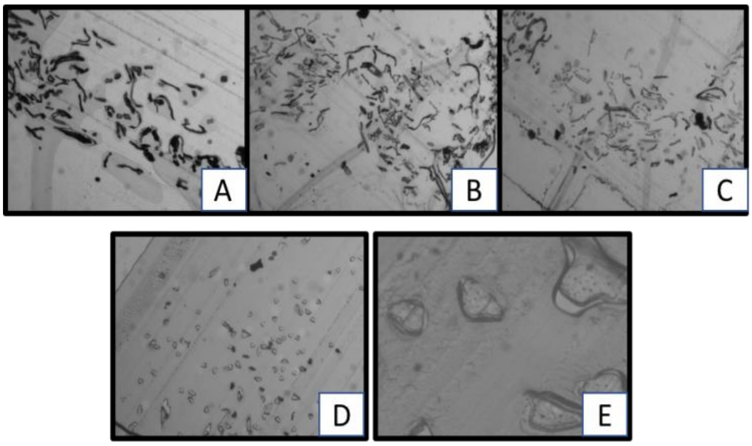

Figure 4.4 *Baby wipe samples embedded in resin and the cross sections then stained with methylene blue to identify if they were made of cellulosic material. The samples were visualised under a LEICA M205C light microscope at a magnification of X100 A-D and X400 E. Cellulosic material was found in all fibres from all four products, identified by the blue staining (shown as black in the diagram): the flushable baby-wipes (A – 1F, B -2F, C - 3F), and the non-flushable wipe (D & E - 4NF). The non-flushable wet-wipe (4NF) had substantially less staining and therefore less cellulose in its material. E shows how the fibre cross sections are interspersed with small segments that have been dyed.*

A spectrum of each of the products was obtained from FTIR. 1F-4NF spectra were compared against library samples; 1-3F matched rayon (a manufactured fibre made from regenerated cellulose) or cellulosic samples and 4NF for polypropylene. Other added ingredients, such as preservatives and antibacterial compounds, were also found to be present on the spectra and were difficult to remove from the wipe itself.

The surface topography and fibre structure of the baby-wipes was visualised by scanning electron microscopy. For brands 2F-3F, 2 different fibre types were apparent; natural and manufactured looking fibres (Fig. 4.5). The manufactured looking fibres looked processed. They were uniformly long and thin with no cracks. In contrast, the cellulosic fibres looked similar to strips of paper. Their shape was erratic and made out of joined segments. Many of these fibres were splitting. 1F was on average 100% manufactured appearing fibres, while 2-3F was on average 40%. Product 4NF, also only appeared to contain manufactured looking fibres (Fig 4.5).

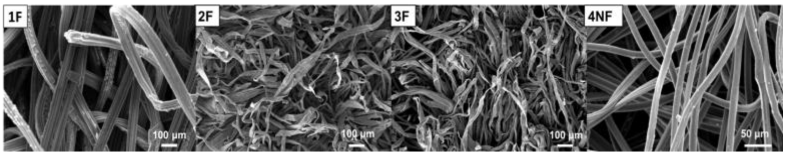

Figure 4.5. *Scanning Electron Microscopy (SEM) showing the topography and fibre composition of baby-wipes before experimentation (time 0). Products (labelled in left hand corner of each row) 1-3F are marketed as flushable/biodegradable baby-wipes. 4NF is marketed as not flushable. Photos taken at X100 (1-3F) & X350 magnification (4NF).*

4.3.1 Laboratory Results

Toilet paper was chosen to be a comparison against disposable wipes in the laboratory testing. This was to compare typical disintegration rates. The toilet paper samples in seawater were the first to show disintegration, which could be visually seen after 24 hours. However, they maintained their core structure for 5 weeks. Fibres from the paper were released making the surrounding saltwater appear hazy.

Focussing on the disposable baby-wipes, 2F-3F showed very similar disintegration rates for all replicate wipes for both seawater and seawater plus additional nutrients within the first 5 weeks. The main structure of the wipe remained, and fibres appeared to break off from the edge. Similarly, to toilet paper, fibre release was apparent after 24 hours. The longer the exposure in the tank, the more cloudy/hazy the surrounding media would become from the released fibres, suggesting disintegration. The disintegration rate was slower for 1F wipes and for wipes exposed in autoclaved seawater.

SEM images after a 5-week exposure in different media types showed any structural changes (Fig. 4.6). Products 1F and 4NF remained unchanged after exposure for all media types. Products 2-3F showed visible deterioration for natural/cellulosic appearing fibres. They were fragmented, with segments breaking off. This was particularly noticeable for 3F wipes in seawater plus additional nutrients (SW + A). The wipe's structure looked pulped, with the manufactured fibres remaining unchanged in the matrix. All samples gained salt deposits on both types of fibre.

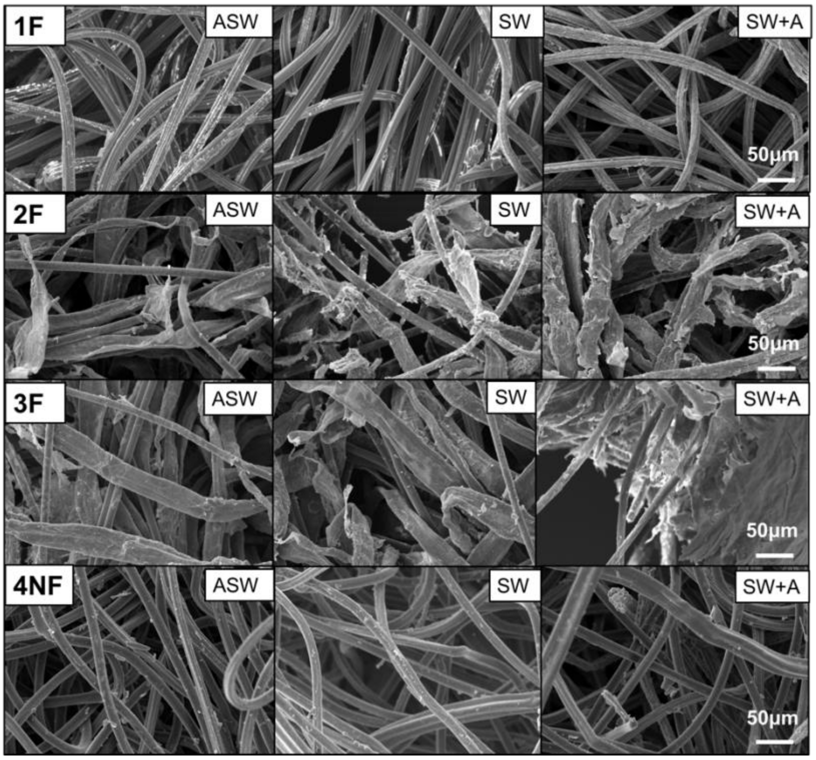

Figure 4.6. *Scanning Electron Microscopy (SEM) of disposable baby-wipes after a 5 week exposure to different media; ASW (Autoclaved Seawater), SW (Seawater) & SW+A (Seawater + Additional Nutrients). Products (labelled in left hand corner of each row) 1-3F are marketed as flushable/biodegradable baby-wipes. 4NF is marketed as not flushable. Photos taken at X350 magnification.*

Excluding the disposable wipes in autoclaved seawater, microbial growth was noticeable after 3 weeks for all flushable wipes. This was particularly prevalent for wipes that had been exposed for a week in the marine environment and in seawater with additional nutrients (SW + A), where the growth covered typically a quarter of each wipe.

For all treatment types, all variants of baby wipes started out being buoyant, allowing a portion of the wipe to be exposed to the surface. The wipe's buoyancy decreased consistently, until it reached stability in the midmost section of the tank. The 4NF wipes remained buoyant throughout the whole exposure period. For all wipe types, the released fibres included a mixture of different buoyancies.

After 10 weeks, samples were taken again. This time, samples were taken from the surrounding media to see what fibres had been released and persisted. SEM images showed that all fibre types were found in the media that corresponded to the main body mesh of the subsequent baby-wipe (Fig. 4.7). This was the same for all treatment types. Fibres from

1F and 4NF replicates showed minimal signs of degradation. 2F-3F looked visibly more aged.

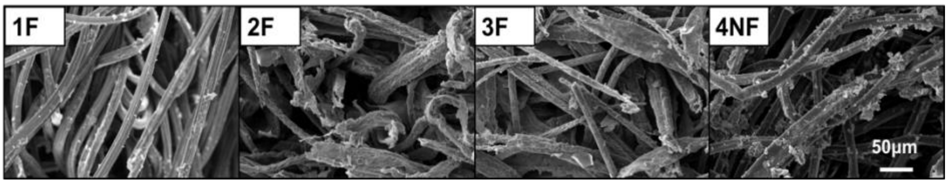

Figure 4.7. *Scanning Electron Microscopy (SEM) of baby-wipes after 10 weeks of exposure to seawater in a laboratory environment. Products (labelled in left hand corner of each picture) 1-3F are marketed as flushable/biodegradable baby-wipes. 4NF is marketed as not flushable. These pictures are representative for each media type. Photos taken at X350 magnification.*

For samples in seawater with additional nutrients (SW + A), visual disintegration after 15 weeks showed that the wipes from products 2-3F had completely broken down, and there was no visible structure of any replicate left in their respective tank. 1F wipes were heavily disintegrated, with approximately 20% of each wipe structure left in each tank. They also were covered in microbial growth. 4NF was also covered in microbial growth but there was no apparent degradation from the non-flushable wipe 4NF.

For the flushable wipes tested in ASW (autoclaved seawater) and SW (Seawater) the main structure of the wipes still remained after 15 weeks. However, they were very sensitive to movement and would subsequently fragment. 4NF had no apparent signs of degradation.

4.3.2 Environmental Results

The flushable wipes deployed in the environment broke down quicker than their counterparts in laboratory conditions. The main structure of the wipes became increasingly smaller throughout the 3-week period. Each flushable wipe appeared to lose mass from the edges, consistently making the surface area smaller until no wipe remained. The last visible sample was taken at 3 weeks; this was recorded for wipes 1-3F.

Analysis of their breakdown every week via SEM produced very similar results to flushable wipes exposed in seawater with additional nutrients (SW + A), in the laboratory experiment at 5 & 10 weeks.

For the non-flushable wipe comparison, 4NF, showed very little signs of disintegration. Similarly, to the laboratory experiment, these wipes were still in full structure after a 15-week period (Fig. 4.8).

Figure 4.8. *A non-flushable wetwipe (baby wipe) after 15 week exposure in the marine environment at Queen Anne's Battery Marina (50°36'48.4"N, -4°12'96.5"W). Minimal degradation was shown.*

4.4 Discussion

The majority of disposable wipe products are made from blends of rayon fibres/wood pulp and plastic fibres (such as polypropylene) (Das and Pourdeyhimi, 2014). For example; baby wipes are typically produced with either 70:30 viscose rayon to polyester, or 50:50 rayon to wood pulp with a basis weight 55 g per m^2 (Das and Pourdeyhimi, 2014).

Similar results were found in this research. FTIR analysis identified the non-flushable wipe (4NF) to be made purely of synthetic material; polypropylene. The material for the flushable wipes (1-3F) showed that the main material was either rayon or cellulosic, with the strongest similarity being rayon. When visualised by SEM, products 2F and 3F also displayed two different types of fibre; manufactured and natural (cellulosic) appearing. The product packaging also contained labelling stating that the wipes contained recycled wood and cellulosic material was confirmed in the samples by a methylene blue test. Therefore, it is expected that these disposable baby wipes are composed of both rayon and wood pulp.

Product 1F is expected to be 100% rayon as only manufactured fibres were shown.

In Europe, fibres and fabrics produced from regenerated cellulose became known as viscose, whereas in the U.S. they are termed rayon (Comnea-Stancu et al., 2017). Viscose/rayon is one of the oldest manufactured fibres. It is a regenerated cellulose fibre with a wide spectrum of properties and commonly used in textiles (Kalabek et al., 2016). The important considerations for rayon are that the essential raw material for its production, namely cellulose, is abundantly available and a renewable source (Kalabek et al., 2016; Sengupta, 1997). Rayon fibres can be produced with a wide range of properties, particularly mechanical properties, making it very desirable (Kalabek et al., 2016; Sengupta, 1997). Due to this, it is common fibre for wipes, especially for baby wipes. (Sahu, 2012). However, it can be argued that rayon fibres behave like a synthetic fibre as rayon is widely reported in the marine environment (Comnea-Stancu et al., 2017). Interestingly, already in 2013, Lusher et al. reported the identification of rayon / viscose fibres in the gastrointestinal tract of fish, and in 2014, Woodall et al. came to the conclusion that rayon fibres are a major source of microplastic debris even in the deep sea. (Comnea-Stancu et al., 2017; Lusher et al., 2013; Woodall et al., 2014).

Both the synthetic and cellulosic fibre types undergo the same manufacturing processes to be made into baby wipes (predominately non-woven material). This typically includes making a pulp which is airlaid (produces a randomly orientated fibrous web structure), hydroentangled (bonding process) and then the surface apertured (creates narrow holes/gaps). The aperturing destabilizes and weakens the structure, especially the thinner fabrics. Additionally, if the hydroentangling is done at low pressures (below 80 bar), then the structure is given either chemical or thermal bonding to stabilize and improve the strength of wipes. The binders usually increase the stiffness of wipes (Das and Pourdeyhimi, 2014).

It has been reported that the bonding process when manufacturing a wipe will determine if a wipe is flushable (Mango, 2008). A flushable wipe is made from a pulp that is bonded by ion sensitive cationic polymer binders or by light hydroentanglement. The process in which it is made means that when the wipe is immersed in water, it should become weak enough to become flushable (Mango, 2008). However, it is also reported that flushable wipes undergo various degradation processes such as physical disintegration caused by turbulent forces, hydrolysis of soluble material, and biodegradation of dissolved materials. Physical disintegration depends on turbulence and solid characteristics such as size, mechanical strength, and density of solids (Karadagli et al., 2012).

Therefore, it is important to understand the definition of a flushable product. The Association of the Nonwoven Fabric Industry collaboratively worked with the Wastewater Industry across the United States and in Europe. This organisation states that for a nonwoven product (such as babywipes) to be deemed flushable there must be evidence indicating that it clears toilets and properly maintained drainage pipe systems when the suppliers recommended usage instructions are correctly followed. It must also pass through wastewater conveyance systems, be compatible with wastewater treatment, reuse and disposal systems without causing system blockage or other operational problems. Additionally, it should also be unrecognisable in effluent leaving onsite and municipal wastewater treatment systems and in digested sludge from wastewater treatment plants that are applied to soil (INDA and EDANA, 2013).

Currently within the UK, there is no way of assessing whether a product can be considered to be suitable for disposal via the sewerage system (Drinkwater and Galletti, 2008). The legal context for the disposal of anything other than faeces, toilet paper and consented trade effluent is not clear. The law anticipates that domestic sewage contains waste from toilets, and water that has been used for cooking and washing, but the relevant law for domestic waste varies between regions of the British Isles. These requirements are ambiguous as there are no compulsory tests that require a product to be tested that could provide evidence. It is also particularly difficult to monitor products, such as baby wipes, which can be made from different materials; often making them non-flushable.

As shown from our research, the quickest possible breakdown for the wipes was 3 weeks when exposed in the marine environment. This is still a slow time frame and will potentially still create blockages within sewage systems. The non-flushable wipe showed no signs of degradation in 15 weeks, and due to its unclear labelling may often still end up in the waste water system.

It has been reported that the most sustainable route for disposal of sanitary products, such as disposable wipes, is via the solid waste disposal route. However, pressure on landfill sites and from local authorities means that many consumers are seeking ways to reduce their solid waste disposal (Ashley et al. 2005). Additionally, customers are wanting their products to be convenient in use and disposal (Sahu, 2012). Flushable products fit these criteria.

Therefore, manufacturers are now making it easier for customers to incorporate these products into their normal bathroom use. Baby-wipes are particularly prevalent due to their frequent and wide variety of uses.

The disintegration of disposable flushable wipes was shown in this study, where it appears that the main structure of flushable wipes is breaking down. Subsequently, this releases fibres which are no longer secured by the structural mesh. The degradability of these fibres is still unknown. However, in laboratory conditions, when analysing the surrounding media for released fibres, all fibre types observed were found even after a 10-week exposure. This could potentially show that the fibres from the wipes are not biodegrading, but the structure is breaking down. To the eye, this can make it appear that the wet wipes are disintegrating as stated on the packaging, but the fibre release may still have a large impact on the environment.

Developing truly disposable flushable wipes that are functional and affordable will undoubtedly pose tremendous challenges to wipe manufacturers in future. Additionally, alongside many disposable wipes there are also a number of other items which are commonly flushed that are not intended by the manufacturer to be introduced into the sewer system. These can include cotton-buds, tampons and disposable razors (Drinkwater and Galletti, 2008). As the number of flushable products increase and consumers become more convinced of their convenience (flushability), the numbers of blockages will increase (Wessex Water, 2016). Water companies are now even urging the disposable wipes industry to stop 'misleading' labelling on disposable wet wipes that are marketed as flushable.

4.5 Conclusion

Our research highlights the persistence of disposable wet wipes and potentially their fibres once released into the marine environment. Our recommendation would be to remove flushable labels from packaging and make it clear the impact that flushing these wipes have on the sewer system and the environment. Furthermore, studies should look into the potential of the persistence of rayon fibres compared to that of synthetic fibres in the marine environment. Although from a natural source, there is potential that these fibres could persist within the marine environment for a prolonged period.

Chapter 5

Environmental Degradation of Biodegradable, Degradable and Conventional Plastic Carrier Bags in the Sea, Soil and Open-Air Over a Three-Year Period

A version of this chapter is currently under review for Environmental Science and Technology

Napper I.E., Thompson R.C., Degradation of Biodegradable, Degradable and Conventional Plastic Carrier Bags in the Environment Over a Three-Year Period.

Abstract

Plastic carrier bags are ubiquitous in modern society but are often single use, disposable items. The majority of bags are made from conventional polyethylene and there is clear evidence of the accumulation of discarded bags in the environment. Partly in response to this environmental concern, various plastic formulations have been developed which state they disintegrate faster and/or have fewer impacts on the environment because their persistence is shorter. This study examined the degradation of five plastic bag materials over a three-year period. These were obtained from U.K. high-street retailers and were either labelled as biodegradable, oxo-biodegradable, compostable or high-density polyethylene (i.e. a conventional plastic carrier bag). These materials were exposed in three natural environments which they could potentially encounter if discarded; open-air, buried in soil and submersed in seawater as well as in controlled conditions in the laboratory. In the marine environment, the compostable bag completely disappeared from the experimental test rig within 3-months. However, the compostable bag type was present in the soil environment after 27 months but was unable to hold any weight without tearing. After 9 months exposure in the open-air, all bag materials had disintegrated into fragments. Our results showed that biodegradable, oxo-biodegradable and conventional plastic formulations persisted and remained functional as carrier bags after being in the soil or the marine environment for over 3 years. Considering the highly variable receiving environments in which plastic litter can accumulate, it is not clear that the oxo-biodegradable or biodegradable formulations tested could be relied upon to offer a sufficiently advanced rate of degradation, compared to conventional bags. Especially if they are to be considered advantageous in reducing the impacts of plastic litter. Plastic films are challenging to recycle even if disposed of properly, but there are additional concerns about the compatibility of modified plastic formulations with available recycling and organic waste streams.

5.0 Environmental Degradation of Biodegradable, Degradable and Conventional Plastic Carrier Bags in the Sea, Soil and Open-Air Over a Three-Year Period

5.1. Introduction

Plastics are lightweight, strong, durable and corrosion-resistant materials which have become an integral part of daily life worldwide (Thompson et al., 2009b). The versatility of plastic, together with its low cost, has resulted in annual worldwide production exceeding 335 million tonnes (PlasticsEurope, 2018).

Approximately 50% of plastics are discarded after a single-use (Hopewell et al., 2009; UNEP, 2018). Therefore, a large quantity of plastic items enter municipal waste streams at the end of their service life. This creates a major waste management problem, with plastics accounting for approximately 8–10% of all the waste generated in the U.K. (Barnes et al., 2009; Hopewell et al., 2009). Considerable quantities of end of life plastics also escape to the environment as litter, and single use items constitute a large proportion of the litter found in marine and terrestrial environments.

Plastics are the dominant category of litter reported in the marine environment and can account for up to 75% of all litter found on shorelines. Plastic debris has also accumulated on the sea bed in shallow water and the deep sea (Bergmann and Klages, 2012; Debrot et al., 2013; Woodall et al., 2014). Plastic litter can harm marine organisms via ingestion or entanglement, and may favour the dispersal of invasive species (Gall and Thompson, 2015; Gregory, 2009; Kühn et al., 2015). Additionally, there are concerns about the release of chemicals from plastic, including additives (components of plastic) and the accumulation of hydrophobic contaminants (adsorbed onto plastic from surrounding sea water) (Teuten et al., 2009), but there is currently little evidence to suggest that plastics will transfer chemicals in sufficient quantities to present toxicological harm in environmentally relevant scenarios (Bakir et al., 2016; Koelmans et al., 2013).

In addition to the negative effects on marine life, there is evidence that plastic debris can harm maritime industries, tourism and human wellbeing (Thompson, 2017; Thompson et al., 2009a; Wyles et al., 2015). In the marine environment the accumulation of plastic debris has been identified as a major global issue by the United Nations Environment Assembly and in the G7 Leader´s declaration 2015 (GESAMP, 2016; UNEP, 2017; Werner et al., 2016).

Plastic debris is also widespread in terrestrial and freshwater environments. However, much of the existing information about the presence of plastics in these environments is focused on sources and transportation pathways to the oceans. Given that the majority of all plastics will be used and disposed of on land, terrestrial environments will themselves be subject to extensive pollution by plastics of all sizes, based on large amounts of anthropogenic litter from both point (e.g. landfill, wastewater treatment discharge, sewage sludge application) and diffuse (e.g. general littering) sources. As such it is highly likely that soils may also act as long-term sinks for plastic debris (Rillig, 2012; Zubris and Richards, 2005).

Since their introduction in the 1970s, plastic carrier bags have become widespread in daily life worldwide (Williams, 2004). They are typically considered as single-use items and are commonly made from polyethylene (European Parliament and Council of the European Union, 1994). These bags can be regarded as an iconic symbol of our ‘throw-away’ society and their waste is often viewed as a very visible nuisance. In 2010, it was estimated that 98.6 billion plastic carrier bags were placed on the European Union (EU) market and about 100 billion plastic bags have been placed additionally every year since (European Commission, 2013).

Plastic carrier bags are often supplied free or for a low charge and used in high volumes. Consumption figures vary greatly between countries, with annual use per capita exceeding 450 bags in some EU countries (European Commission, 2013). Interventions to reduce the use of plastic bags have been varied in range and scope. Governments in many nations have strategies to either ban the sale of lightweight bags, charge customers for their use and/or generate taxes from stores who sell them (Xanthos and Walker, 2017). Several countries have already included bans or taxes, which have resulted in substantial reductions in use (Convery et al., 2007). However, there is no consistency of taxes or regulations between countries.

There are growing concerns that the littering of plastic carrier bags presents a substantial source of contamination in the oceans. They have been found to be one of the most common items in the intertidal (Thiel et al., 2003; Willoughby et al., 1997) and subtidal benthos (Galgani et al., 2000). Even if properly discarded, lightweight bags can unintentionally be transferred away from landfill sites or other areas by wind or heavy rain (Barnes et al., 2009).

The presence of carrier bags in the marine environment can have a number of effects. Research by Bugoni et al. (2001), found that out of 50 dead stranded sea turtles, plastic carrier bags were the main debris ingested. Green et al. (2015), found that within 9 weeks in the marine environment, plastic carrier bags can create anoxic conditions within the sediment, and that their presence can significantly lower abundances of infaunal invertebrates. This indicates carrier bags can rapidly alter marine assemblages and the ecosystem services they provide (Green et al., 2015). Additionally, Hodgson et al. (2018) did laboratory experiments on carrier bags and showed that amphipods can shred plastic carrier bags, generating numerous microplastic fragments (average diameter 488.59 μm). This study was to investigate the shredding and subsequent potential ingestion of plastic by amphipods.

Despite their durability once in the environment, plastics can become oxidized when exposed to ultraviolet B (UVB) radiation from sunlight and the oxidative and hydrolytic properties of the atmosphere and seawater. This can form hydroperoxides which lead to polymer chain scission and fragmentation (Billingham et al., 2000). However, this is a very lengthy process and it is likely that considerable further degradation would be required before the plastic could reach a size sufficiently small that it might biodegrade. The hydrophobicity and long carbon chain molecular structure of polyethylene, which is widely used for plastic bags, makes it resistant to biodegradation under normal conditions and the timeframe for the complete mineralisation is unknown. Therefore, with the large quantities of carrier bags used this presents a major waste management issue.

Awareness of the accumulation of end of life plastic and its impact on the environment has, in part led, to interest in the development of degradable polymers. Biodegradable, oxo-biodegradable and compostable plastics are widely regarded as potential solutions to the accumulation of plastic litter and waste. Some of these products are accompanied by statements indicating they will break down ‘typically within 24 months’ and ‘quicker than conventional petroleum-based plastic'. These materials are widely used for the production of carrier bags and some are also being used to make a variety of other items, including single-use cutlery, water bottles and straws.

Biodegradation takes place through the action of enzymes and/or chemical deterioration associated with living organisms, bacteria, fungi and algae. This occurs in two steps; the first is the fragmentation of the polymers into sections of lower molecular mass by means of either abiotic reactions (i.e. oxidation, photodegradation, hydrolysis), or biotic reactions (i.e. degradation by microorganisms). This is followed by bioassimilation of the polymer fragments by microorganisms and its mineralisation (Lucas et al., 2008). A material may be labelled as 'biodegradable' if it conforms to certain national or regional standards that apply to industrial composters, not to domestic compost heaps or the natural environment (Kershaw, 2015).

Such standards could include: ISO, European Norm – EN and American Society for Testing and Materials (ASTM) International. These standards are appropriate for conditions that occur in an industrial composter, in which temperature are expected to reach 70 °C. For example, the EN standard requires that at least 90% of the organic matter is converted into CO_2 within 6 months, and that no more than 30% of the residue is retained by a 2 mm mesh sieve after 3 months composting (EN 13432:2000, 2000).

Oxo-biodegradable plastics (oxo-plastics) are reported to contain an additive (pro-oxidant) which is intended to break the molecular chain within the polymer which will then lead to its biodegradation (Koutny et al., 2006; Ojeda et al., 2009). However, there is typically no clearly defined timeframe given for the breakdown of oxo-/biodegradable plastics (Association for Organics Recycling, 2011).

'Composting' is enhanced biodegradation under managed conditions, predominantly characterised by forced aeration and natural heat production resulting from the biological activity taking place inside the material. The resulting output material, compost, contains valuable nutrients and may act as a soil improver (European Commission, 2018). Therefore, compostable plastics should biodegrade in a managed composting process through the action of naturally occurring micro-organisms and typically do so in relation to a specified timeframe (Association for Organics Recycling, 2011).

There is a lack of clear evidence that biodegradable, oxo-biodegradable and compostable materials offer an environmental advantage over conventional plastics, and the potential for fragmentation into microplastics causes additional concern (Kershaw, 2015; O'Brine and Thompson, 2010). To date, studies focusing on the breakdown of different types of degradable plastics in the environment give varying results and are shorter in timeframe. For example, O'Brine and Thompson (2010) tested biodegradable plastic within a 40-week timeframe in the marine environment and found that the majority of plastic still remained.

The degree to which synthetic polymers degrade depends on both the properties of the polymer and the environment to which it is exposed (Mohee and Unmar, 2007). Due to lack of consistent evidence about rates of degradation in the environment, the overall environmental benefits, allegedly misleading claims to consumers and risks that labelling products as biodegradable may inadvertently promote littering behaviour, a process is being proposed to restrict the use of oxo-plastics in the EU (European Commission, 2018).

The present study examined the degradation of bags which are stated to have biodegradable, oxo-biodegradable or compostable properties. A conventional polyethylene plastic carrier bag was also examined for comparison. All bags were available at the point of sale in U.K. high-street retailers. These materials were exposed in various environments that discarded carrier bags could encounter; in open-air, buried in soil and submersed in the marine environment. This is the first research where plastic degradation has been examined simultaneously across these three natural environments, together with controlled conditions in the laboratory. Five different plastic carrier bag formulations were considered, and their degradation was evaluated over a 3-year period. Deterioration was considered in terms of visible loss in surface area and disintegration as well as approaches to detect more subtle changes in tensile strength, surface texture and chemical structure.

5.2 Methods

5.2.1 Sample Preparation

Five different types of plastic carrier bags were compared (Table 1): these included two types of oxo-biodegradable bag (labelled here as Oxobio1 and Oxiobio2), one biodegradable bag, one compostable bag, and high-density polyethylene (HDPE) carrier (labelled in this research as a conventional carrier bag), which was not stated to have any particular degradation/compostable properties.

The bags were chosen as they were all opaque and were obtained based on their prevalence in retail stores in and around Plymouth (U.K.). Sixteen samples of each bag were obtained. In order to ensure the samples were as independent from each other as possible, a maximum of two bags were sourced from any one store on any single occasion. Where repeat visits to the same store were necessary to obtain sufficient independent samples, these visits were separated by at least 2 weeks. Hence our experiment was designed to contain a range of products and production batches so as to be as representative as possible. Since the specific retail stores from which the carrier bags were obtained is not of particular relevance, bags will only be described based on their formulation (Table 1).

Designated label for testing	Degradation properties (as stated on bag)	Information stated on websites linked to the product	Disposal/anti-littering information (as stated on bag)
Oxobio1	Degradable Plastics (D_2W trademark, logo)	Oxo-biodegradable (https://www.symphonyenvironmental.com/d2w/)	No information
Oxobio2	Planet safe plastic; incorporating EPI's totally degradable plastic additives (EPI trademark, logo)	Oxo-biodegradable (http://www.epi-global.com/en)	No information
Biodegradable	Biodegradable bag (exo plastics logo, sustainable bioplastic; Biodegradable ISO 14855)	No claims about biodegradability on exo plastics website (https://www.exoplastics.com/) ISO 14855 is an international standard covering aerobic biodegradability of plastic materials under controlled composting conditions	Recyclable (no numerical category for recycling type stated)
Compostable	Completely compostable, recycle me with food	Plant-based compostable foodservice packaging (https://www.vegware.com/about/info_1.html) Compostable packaging is designed to be recycled together with food waste. https://www.vegware.com/close-the-loop/info_50.html EN13432 is the packaging waste directive and standards for compostability http://www.bpf.co.uk/topics/standards_for_compostability.aspx	Reuse me first for shopping, and then as a food waste caddy liner! This completely compostable bag complies with standard EN13432. Suitable for industrial food waste recycling – visit www.foodwastenetwork.org.uk. Recycling category '7 - other'
Conventional	High Density Polyethylene; No degradation properties stated	No manufacturer given	Reuse at home or recycle. Recycling category '2 – HDPE'

Table 5.1. *Information on the tested carrier bags and the properties as stated on the manufacture's website. All bags were opaque and obtained based on their prevalence in retail stores in and around Plymouth, U.K.*

Each carrier bag type was cut into strips; 15 x 25 mm. The strip samples were taken from the main body of the carrier bag (not the handles or the sides), to provide areas of similar structure. A strip of each plastic carrier bag type was then placed into a pouch made of high-density polyethylene (HDPE) mesh and sewn secure using nylon fishing twine. Each pouch structure (150 x 200 mm) was sewn together to provide 5 equally spaced separated compartments. These compartments were then used to house an individual strip of each bag type (Fig.1). The HDPE mesh (1 mm x 1 mm) allowed exposure to external environments and each compartment was sewn so as to allow the bag samples to move relatively freely. Each pouch structure was attached to a permanent panel to aid removal.

These permanent panels were placed in one of four different conditions; buried in soil, exposed outdoors in the air, submerged in the marine environment and placed in a blacked-out box in the laboratory as a control. The buried samples were situated at the University of Plymouth's Skardon Garden (50°22'38.4"N, -4°08'11.9"W) and were buried to a depth of approximately 0.25 m (Fig.1a). The samples that were exposed in open-air were also situated in Skardon Garden and were placed on a South facing wall (Fig.1b). Samples placed in the marine environment were submerged on a beam at Queen Anne's Battery Marina (50°36'48.4"N, -4°12'96.5"W) at a depth of approximately 1 m (Fig.1c). 3 kg weights were connected on each side of the beam to maintain depth. Control samples were placed in a blacked-out box (kept at room temperature) in a laboratory at the University of Plymouth.

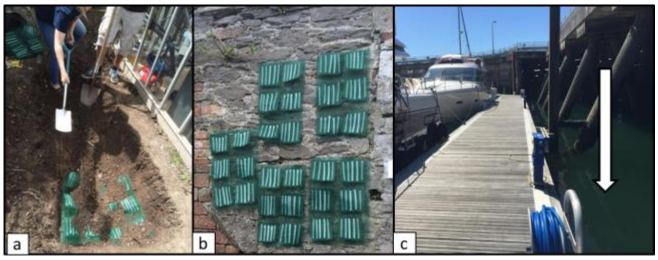

Figure 5.1. *Setup for the degradation experiment testing different types of plastic used for carrier bags. The bags were marketed as being either: oxo-biodegradable, biodegradable or compostable. A bag made from conventional polyethylene was also tested.*

The samples were cut into standardised strips and placed in 4 different environments: A – buried in soil in (Skardon Garden, Plymouth University); B- Strips hung on a south-facing wall, exposed to the outside environment (Skardon Garden, University of Plymouth; C – Strips submerged in a salt-water marine environment (Queen Anne's Battery Marina; arrow denotes where). Control samples (not illustrated) were stored in a blacked-out container at University of Plymouth.

The sea temperature in Plymouth (England) ranges from 8.8 – 18.8 °C (United Kingdom Sea Temperatures, 2018). The air temperature typically ranges between 1 - 21.5 °C, depending on the season (Met Office, 2016).

All samples were deployed on the 10th July 2015. There were 3 subsequent sampling dates; 6th April 2016 (9 months), 6th Jan 2017 (18 months), 6th October 2017 (27 months). Additionally, whole bags of each material were also deployed in polypropylene mesh in each environment at the same time and used for visual inspection over the 3-year period (23rd August 2018).

Before deployment, 4 subsample strips from each carrier bag type were tested to provide a comparison starting point. After deployment, four replicate samples of strips from differing bag replicates were collected from each environment on each sampling date. Samples were removed from the mesh structure, gently cleaned using distilled water, air dried (30°) and tested (see below) within 48 hours.

5.2.2 Visual Inspection

The first step on each sampling date was to visually inspect the samples to check for surface area loss, holes or disintegration. Random samples of each carrier bag type were then also visualised by scanning electron microscopy (JEOL, 7001F) prior to deployment, and then from each environment at 27 months. The images were used to visually assess the deterioration of the material by analysing its surface for cracks or fragmentation.

Measurements of tensile stress and molecular structure using Fourier transform infra-red spectroscopy (FTIR) were made in order to detect any more subtle changes.

5.2.3 Tensile Stress Testing

The thickness of each strip was measured using an electronic micrometer (Sealey; AK9635D). Each strip was measured at 10,50,100 and 140 mm from a central point. This produced 4 reference points for each sample and the mean was then calculated. The maximum load (N) for each strip was then measured using a tensile testing machine at a rate of 100 mm min^{-1} (Instron, system ID 3345 k1669 - USA, force transducer model

2519-104, capacity 500 N). Then, the maximum tensile stress of each strip was calculated using the following equations:

i) $A = bh$ ii) $\sigma = \frac{F}{A}$

where b is the width (25 mm), h is the height (mean thickness) and F (maximum load, N) *is* the force for each extracted strip. For each strip Eq. (i) allowed calculation of the cross-sectional area (A) and Eq. (ii) allowed calculation of the tensile stress (σ, MPa). The maximum tensile stress of a material is also termed as its ultimate strength (and referred to as the rate of disintegration within this research).

Normality of the data was confirmed by using QQ plots to examine distribution. One-way analysis of variance (ANOVA) was used to compare the maximum tensile stress difference between the different bag types before being exposed in any environment. The effects of bag type, environment and time on the maximum tensile stress was then examined. This was compared using the percentage change of tensile stress from 0 to 9 months and 9 to 27 months using a three-way ANOVA. Post-hoc Tukey tests were then used to identify the significant effects. Any samples which were too brittle to test or were no longer visible were omitted from the analysis. All statistical tests were performed in R ver. 3.4.1 (R Core Team 2017).

5.2.4 Molecular Composition Analysis (FTIR) and Image Analysis

In order to assess any subtle deterioration effects on the molecular composition of the materials, Fourier transform infra-red spectroscopy (FTIR) was conducted (Hyperion 1000 microscope coupled to an IFS 66 spectrometer; Bruker). Prior to FTIR, samples were cleaned with absolute ethanol to remove any residues. The spectra obtained were compared against a spectral database of synthetic polymers (Bruker I26933; Synthetic fibres ATRlibrary).

5.3 Results

Prior to exposure in different environments, maximum tensile stress and thickness of the bags was measured. Oxobio2 had the highest tensile stress and thickness (28.82 MPa and 0.04 mm), the compostable bag had the lowest tensile stress (10.47 MPa) and the biodegradable, conventional and Oxobio1 bag had the lowest thickness (0.02 mm) (Table 2).

Bag Type	**Tensile Stress (MPa)**	**Thickness (mm)**
Compostable	10.47 ± 1.23	0.03
Biodegradable	21.36 ± 1.90	0.02
Oxobio1	25.98 ± 8.05	0.02
Oxobio2	28.82 ± 1.55	0.04
Conventional	20.61 ± 1.87	0.02

Table 5.2. *Mean of maximum tensile stress (MPa), standard deviation (SD) and mean thickness of plastic bags before being exposed in different environments (n=4). All bag types had relatively consistent thickness.*

Before commencing the experiment there were significant differences in mean maximum tensile stress [$F_{(4, 15)} = 12.94$, $p = <0.01$)] between the carrier bag materials (Table 3). Post-hoc Tukey's HSD tests showed that the compostable bag had a significantly lower maximum tensile stress when compared against all other bag types. All other comparisons were not significant.

1-way ANOVA					
	Df	Sum Sq	Mean Sq	F value	*P*-value
Bag Type	4	784.5	196.13	12.94	**<0.01**
Residuals	15	227.4	15.16		
Outcome of post-hoc Tukey's HSD tests					
Oxobio2 (28.82 MPa ± 1.55) = Oxobio1 (25.98 MPa ± 8.05) = Biodegradable (21.36 MPa ± 1.90) = Conventional (20.61 MPa ± 1.87) >* Compostable (10.47 MPa ± 1.23)					

Table 5.3. *Outcomes of 1-way ANOVA test with post-hoc Tukey's HSD tests showing the mean +/- S.D. and any significant differences in maximum tensile stress of 5 different carrier bag materials (n=5, >* signifies a significant difference at P 0.05, = denotes no significant difference)*

After the various exposure periods, all pouch structures were successfully recovered from all environments. The strips and whole bags were then analysed visually.

For plastic bag strips in both the control and soil environment, no surface area loss was measurable over the period of 27 months. Within the marine environment, a microbial biofilm was visible on the surface of all bags after 1-month. However, the compostable bag samples (including whole bags) were no longer visible by the 1st sampling date of 9 months.

After 9 months, in the open-air environment all bag types (including conventional polyethylene) were too brittle to test and had or were disintegrating into pieces. Most of the pieces were in the microplastic size range (<5 mm); therefore, they could not be examined for tensile stress. The whole bags were also found to have disintegrated into microplastic pieces. Substantial quantities of the fragments that formed were visible to the naked eye on the ground beneath the test rig and in the pouches. While disintegration into microplastic was apparent it was not clear whether this fragmentation could have altered the potential for the plastic to biodegrade and more work would be needed to establish this together with the associated timescale.

Scanning electron microscope images were obtained before environmental exposure and then again after 27 months. For each material, the topography and roughness were assessed. The main noticeable changes were found with the open-air sample fragments for both conventional and compostable bag types. Cracks and holes were present in the conventional bag material suggesting deterioration (Fig. 5.2.1b). For the compostable material, solid deposits that looked like filamentous bacteria were visible on the surface; however, no cracks or holes were present nearby (Fig. 5.2.2b).

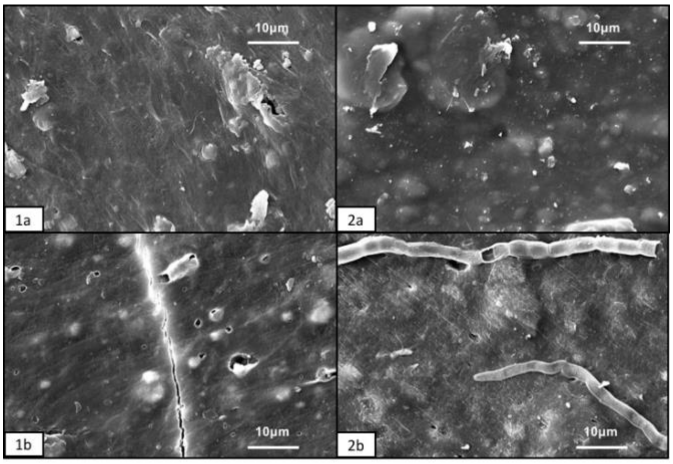

Figure 5.2. *Scanning Electron Microscopy (SEM) imagery showing the typical topography of conventional plastic carrier bag (1a) and a compostable carrier bag material (2a) before testing and then after 27-month exposure outside (1b & 2b, respectively) (2000X magnification) (Images taken at Plymouth Electron Microscopy Centre).*

After 3 years, photographs were taken of the whole bags from both the soil and marine environment (Fig. 3). As a qualitative assessment of functionality these bags were loaded with typical groceries from a local supermarket (weight 2.25 kg). Oxobio1, Oxobio2, biodegradable and conventional were still functional and retained the items with no breakages. However, the compostable bag type (which was only present in the soil environment after 27 months) was unable to hold any weight without tearing.

Figure 5.3. *Oxo-biodegradable bags (oxobio2) which had either been submerged in the marine environment (left) or buried in soil (right) for over three years. Each bag is holding 2.25 kg of typical groceries.*

The maximum tensile stress of all plastic types decreased in all environments over time, but at different rates (Fig. 5.4). A three-way ANOVA was conducted to examine the effect of three factors (Bag type, Environment, Time) on the percentage change in maximum tensile stress of 150 x 25mm strips. The factor Time had two levels (0-9 and 9-27 months); Bag type had 5 levels (Oxobio1, Oxobio2, biodegradable, compostable and conventional) and environment consisted of 4 levels (control, open-air, marine, soil). Overall, this involved destructive sampling of 262 samples, with each measurement taken from a previously untested strip.

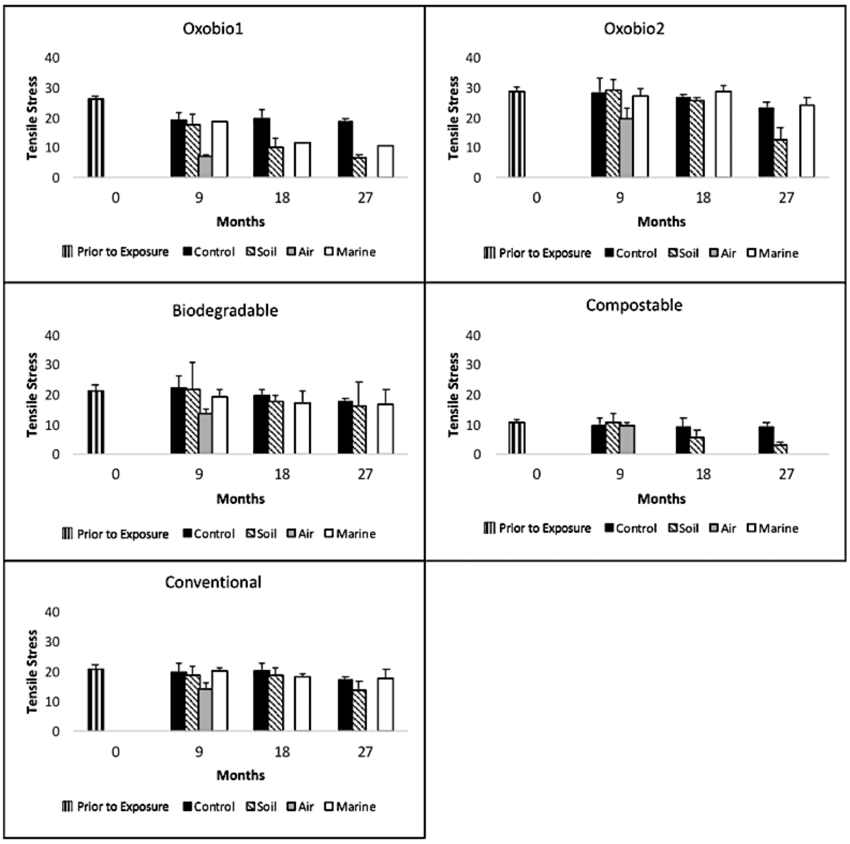

Figure 5.4. *Mean of maximum tensile stress of plastic carrier bag samples, shown as maximum stress before breakage displayed (mean + SE) over a 27-month exposure period in 4 different environments (control, marine, soil, open-air). Open-air is labelled as air in this graph. If bag type is not shown in relation to an environment, it denotes complete disintegration/fragmentation and hence samples were not testable.*

As the compostable bag samples had completely disappeared from the mesh in the marine environment this gave an unbalanced data set, and so this bag type was examined using a separate analysis just considering the remaining environments and sampling dates. Additionally, all bag types after 9 months in the open-air environment could not be tested due to being too brittle; these were subsequently omitted from statistical testing from 9-27 months.

From the perspective of litter and potential interactions with biota most of the bag samples remained intact. However, subtle changes in tensile stress were apparent in all the bag materials indicating some degree of deterioration and the factors Time, Bag type and Environment showed significant differences between 0 - 9 months exposure (Table 4).

Post hoc comparisons found that Oxobio1 lost strength at a significantly faster rate than the other bags between 0 - 9 months (*p* = <0.01). There were also differences between the two Oxo-biodegradable samples; Oxobio1 lost strength significantly faster than Oxobio2 (*p* = 0.01). Additionally, bags exposed in the open-air environment lost strength more rapidly when compared to the other environments: control (*p* = <0.01), marine (*p* = <0.01) and soil (*p* = <0.01).

A second ANOVA was conducted which included the compostable bag type. This bag type needed a separate analysis as all its samples had completely disappeared within the marine environment after 3 months. This ANOVA showed that the compostable bag material had a significant difference in tensile stress to Oxobio1 (*p* = <0.01). The compostable bag material exposed in the open-air environment also lost its strength more rapidly when compared to bags exposed to both control (*p* = <0.01) and soil (*p* = <0.01) environments.

Time: 0 - 9 Months				
Variable	DF	MS	*F*	*P*-value
Time	1	10381	37.16	**<0.01**
Bag	3	1788	6.40	**<0.01**
Environment	3	2244	8.03	**<0.01**
Time:Bag	3	1788	6.40	**<0.01**
Time:Environment	3	2244	8.03	**<0.01**
Bag:Environment	9	69	0.25	0.99
Time:Bag:Environment	9	69	0.25	0.99
Residuals	96	279		
Time: 9 - 27 Months				
Variable	DF	MS	*F*	*P*-value
Time	1	12259	43.69	**<0.01**
Bag	3	4792	17.08	**<0.01**
Environment	2	1722	6.14	**<0.01**
Time:Bag	3	203	0.72	0.54
Time:Environment	2	1476	5.26	**0.01**
Bag:Environment	6	341	1.22	0.31
Time:Bag:Environment	6	325	1.16	0.34
Residuals	72	281		

Table 5.4. *3-Way ANOVA to comparing changes in tensile strength of 5 bag types over 27 months. This focussed on the influence of three independent variables (bag type, environment, time) on the percentage change of maximum tensile stress of 150 x 25mm strips at 0-9 and 9-27 months. Bag type included 4 levels (4 bag types; Oxobio1, Oxobio2, Biodegradable and Conventional), environment consisted of 4 levels (control, open-air, marine, soil). Analysis of 9 - 27 months does not include open-air samples due as they were too brittle to test. The compostable bag was not included in the analysis due to its rapid loss of marine samples.*

Exposure from 9 - 27 months produced similar relative changes in tensile strength patterns as 0 – 9 months (Table 4). However, Oxobio1 and the Conventional bag type were also found to differ in tensile stress (p = <0.01). As samples in the open-air were too brittle to test after 9 months, the only significant difference between environments was between soil and control (p = <0.01). Furthermore, the specific order of tensile stress between the bags was largely unchanged throughout 27 months, whereas environment type seemed to have a greater effect (Table 5).

Environment Type	Sample Extraction Timeframe								
	0 Months								
Control	Oxobio2 (28.82 MPa ± 1.55)	=	Oxobio1 (25.98 MPa ± 8.05)	=	Biodegradable (21.36 MPa ± 1.90)	=	Conventional (20.61 MPa ± 1.87)	>*	Compostable (10.47 MPa ± 1.23)
Soil	-		-		-		-		-
Open-air	-		-		-		-		-
Marine	-		-		-		-		-
	0-9 Months								
Control	Oxobio2 (28.31 Mpa ± 5.12)	=	Biodegradable (22.02 Mpa ± 4.07)	=	Conventional (19.66 Mpa ± 2.86)	=	Oxobio1 (19.07 Mpa ± 0.77)	=	Compostable (9.35 Mpa ± 2.81)
Soil	Oxobio2 (29.29 Mpa ± 3.39)	=	Biodegradable (21.76 Mpa ± 8.76)	=	Conventional (18.67 Mpa ± 3.11)	=	Oxobio1 (17.49 Mpa ± 5.24)	=	Compostable (10.35 Mpa ± 3.48)
Open- air	Oxobio2 (19.74 Mpa ± 3.64)	=	Conventional (14.29 Mpa ± 1.93)	=	Biodegradable (13.71 Mpa ± 1.60)	=	Compostable (9.77 Mpa ± 0.77)	>*	Oxobio1 (6.77 Mpa ± 4.76)
Marine	Oxobio2 (27.09 Mpa ± 2.36)	=	Conventional (20.04 Mpa ± 1.00)	=	Biodegradable (19.05 Mpa ± 2.72)	=	Oxobio1 (18.61 Mpa ± 2.74)	(/)	Compostable
	9-27 Months								
Control	Oxobio2 (23.32 Mpa ± 1.64)	=	Oxobio1 (18.42 Mpa ± 2.05)	=	Biodegradable (17.46 Mpa ± 1.11)	=	Conventional (17.15 Mpa ± 1.00)	=	Compostable (9.26 Mpa ± 1.16)
Soil	Biodegradable (16.01 Mpa ± 7.97)	=	Conventional (13.68 Mpa ± 3.11)	=	Oxobio2 (12.34 Mpa ± 4.49)	=	Oxobio1 (6.38 Mpa ± 2.44)	=	Compostable (3.26 Mpa ± 0.99)
Open- air	/		/		/		/		/
Marine	Oxobio2 (24.05 Mpa ± 2.86)	=	Conventional (17.82 Mpa ± 2.88)	=	Biodegradable (16.43 ± 5.05)	=	Oxobio1 (10.43 ± 2.41)	(/)	Compostable

Table 5.5. *Outcomes of post-hoc Tukey's HSD tests showing the mean +/- S.D. and any significant differences in maximum tensile stress of 5 different carrier bag materials, in four different environments over a period of 27 months. Separate analysis was required for the compostable bag type as all its samples had completely deteriorated within the marine environment in 3 months. (n=4, >* signifies a significant difference at P 0.05, / signifies no available sample for testing and = denotes no significant difference)*

There were also changes in tensile stress between 0 – 27 months; samples exposed in the open-air were found to alter the most and those in the control environment the least (Table 5.6). When comparing bag types (and ignoring any samples that had deteriorated to such an extent tensile strength could not be tested), Oxobio1 had the greatest loss in tensile stress over 27 months for all environments; soil (75% loss), marine (60% loss) and control (29% loss). Conventional plastic had the least reduction in tensile stress for both soil (34% loss) and the marine environment (14% loss). Compostable plastic had the lowest change in tensile stress within the control environment (11% loss), but samples within open-air and marine environment showed total disintegration (Table 5.6).

	% Change in Tensile Stress (MPa)			
Bag Type	Open-Air	Soil	Marine	Control
Compostable	∿	68.86	∿∿	11.48
Biodegradable	∿	25.03	23.10	18.26
Oxobio1	∿	75.46	59.85	29.09
Oxobio2	∿	57.19	16.52	19.07
Conventional	∿	33.60	13.53	16.78

Table 5.6. *The percentage (%) loss change in tensile stress (MPa) of 5 bag types over 27 months from 4 different environment types.* ∿ *denotes that the samples were too brittle to be tested (after 9 months).* ∿∿ *denotes that samples were no longer visible and therefore could not be tested.*

Subtle changes in chemical composition were indicated by FTIR analysis. Some samples developed a small poorly defined carbonyl stretch at a wave number of approximately 1715 cm^{-1}; this is indicative of oxidation which is a sign of deterioration and was more evident for samples exposed in the open-air. However, this varied between materials and environments, with no clear pattern being evident.

5.4 Discussion

Discarded plastics, including carrier bags, are likely to fragment over time in the environment leading to formation of microplastics (Andrady, 2011). Therefore, it is important to understand plastic litter fragmentation processes and mechanisms. However, the degradability of plastics depends on the polymer composition and the environment in which the product is exposed (Rujnić-Sokele and Pilipović, 2017).

Here we report the degradation of several plastic carrier bag materials after exposure in the marine, soil, open-air and control environment over a period of 3 years. All bags were obtained from mainstream retail shops and four of the materials were promoted as having some level of enhanced degradability or composability presumably in relation to conventional polyethylene. Apart from the compostable bag material deployed in the marine environment, fragments or whole samples of each bag material type were present in all environments after 27 months and some of the whole bag samples were still functional as plastic bags after 3 years in the natural environment.

More subtle, but statistically testable and significant, changes in tensile strength were apparent overtime, but the extent of these changes varied among materials and environments. The rates of degradation of plastics in different environments will strongly depend on the local conditions to which they are exposed (Andrady, 2015). Physical and chemical changes in polymers can be caused by environmental factors including light (photo-oxidation), heat (photo-thermal oxidation), mechanical abrasion, moisture, chemical conditions or biological activity (fungi, bacteria, yeasts, algae, and their enzymes) (Charles and Carraher, 2013; Hodgson et al., 2018; Yousif and Haddad, 2013). For example, for the compostable bag samples in the open-air environment, solid deposits that looked like filamentous bacteria were visible on the surface of the material (Fig. 5.2 (2b)).

The tensile strength of bags exposed to sunlight outdoors (labelled as open-air) decreased faster than in the other environments. Between 9 -18 months all of the samples exposed in the open air had fragmented and could no longer hold their original shape because they were too brittle. The faster rate of fragmentation in air may be due to greater levels of ultraviolet (UV) radiation and oxygen, in combination with higher temperatures than in the other environments (Andrady, 2011; Andrady et al., 1993). Ultraviolet (UV) radiation can induce oxidation, which makes plastics brittle and easy to break up due to their decreasing elasticity. This exposure may cause degradation by breaking the polymer chains, producing radicals and reducing the molecular weight, causing deterioration of mechanical properties. This can happen in an unpredictable time frame (Yousif and Haddad, 2013). The amount of exposure to UV would be decreased if plastics are buried in soil, landfill, or if they are submerged in the marine environment and this may explain the slower rates of deterioration observed in these conditions during our study.

Samples buried in soil were found to lose tensile stress significantly faster than samples in the control environment possibly because of increased moisture content in the soil. Understanding the degradation of different plastic types in terrestrial environments is important as substantial quantities of plastic will end up in landfills (Song et al., 2009). Further, in the absence of a specific waste management pathway, for example to a commercial composter, all of these materials will, unless littered be sent as residual waste to landfill or incineration. When plastic accumulates within the soil, it becomes part of a complex mixture of organic matter and mineral substituents. It has been suggested that within this environment microplastics could negatively impact organisms including earthworms (Cao et al., 2017; Huerta Lwanga et al., 2016).

Polyethylene samples in soils have been predicted to take more than 300 years to entirely degrade (Yoshito et al., 1998). However, research by Accinelli et al., (2012) showed that deterioration of bioplastic carrier bags proceeded relatively rapidly in soil incubated under constant moisture and air temperature with a 76% reduction in tensile strength after 3 months.

Our research showed that all carrier bag materials tested appeared intact after they were buried in soil conditions after 27 months. However, more subtle changes were detectable with a 25 – 69% reduction in tensile strength between the different bag types. These results are perhaps more realistic than the previous studies due to being exposed for a longer time period and being exposed to naturally fluctuating soil moisture or air temperature (O'Brine and Thompson, 2010).

Samples exposed in soil and open-air were, overall, found to lose tensile stress significantly quicker than in the marine environment. However, there were no significant differences between the marine environment and control samples (Fig. 5.4), suggesting that degradation in the marine environment was slow. Reduced degradation in seawater has been observed previously. Rutkowska et al., (2002) exposed polyethylene (PE) for 20 months in 2 m water depth in the Baltic sea and reported that there was no biodegradation. Pegram and Andrady (1989) studied the weathering of several plastics typically found in beach debris using floating marine exposure tests over a 6-month period. They measured the rate of deterioration from the changes in tensile elongation at break (and, in some instances, by the force to rupture) and found it to be much slower (2%) for samples exposed in the sea compared to samples exposed in open-air (95%). In the current experiment, after 9 months, conventional polyethylene was found to lose 31% in tensile strength in open-air, but only 2% in the marine environment.

In the marine environment, colonisation by micro- and macro-marine organisms (a process described as fouling), may affect plastic in a variety of ways (Lobelle and Cunliffe, 2011). Firstly, the biofilm may 'shield' the plastic from UV light (O'Brine and Thompson, 2010) thus reducing the rate of photo-degradation. Fouling can also make plastics negatively buoyant causing buoyant items to sink (Fazey and Ryan, 2016); hence further reducing irradiance. In the current experiment, all samples in the marine environment readily acquired a coating of biofilm.

All samples of the compostable bag (Compost), including the whole bag, completely disintegrated within a 3-month period in the marine environment. Similarly, research by O'Brine and Thompson (2010) also found that a compostable bag type had 100% surface area loss between 16 and 24 weeks when deployed in the marine environment. This suggests that degradation of compostable bags can be relatively rapid in seawater. However more work would be needed to establish what the breakdown products or fragments of this deterioration are, and to consider any potential environmental consequences.

From the perspective of the remaining bag types, it might have been expected that the two oxo-biodegradable materials would degrade faster than both the biodegradable and conventional bag types as these bags have pro-oxidants which are incorporated into the polymer chains to accelerate photo- and thermo-oxidation (Koutny et al., 2006). However, throughout the 27 months of this experiment, Oxobio1 was the only bag type to be loose tensile strength significantly faster compared to biodegradable, conventional and Oxobio2 bag types.

Koutny et al., (2006) studied the biodegradability of high-density polyethylene film (HDPE) and low-density polyethylene film (LDPE) containing pro-oxidants and antioxidants. These were tested against microbial strains, including *Rhodococcus rhodochrous* and *Nocardia asteroides which are highly abundant in natural environments including soil (Koutny et al., 2006; Larkin et al., 2005). A*fter an abiotic pre-treatment consisting of photooxidation and unnaturally high thermo-oxidation (60 °C) which was intended to mimic around 3 years of outdoor weathering, the samples were inoculated, incubated up to 200 days (27 °C) and their metabolic activities were followed. An initial phase of *fast* microbial growth was observed, and the authors suggest this was probably caused by utilization of low molecular extractable compounds. This was followed by a long period of stabilized metabolic activity suggesting that the microorganisms continued to gain energy from the substrate but at a much slower rate. Analysis performed at the end of incubation indicated that any biodegradation had probably only affected surface layer of the materials.

Over a 27-month period in the current experiment, little change in the chemical structure of any of the samples was revealed. This outcome is similar to that obtained by Ioakeimidis et al., (2016) who found that according to ATR-FTIR analysis, polyethylene terephthalate (PET) bottles remained robust for at least fifteen years in the marine environment.

The current study showed that, oxo-biodegradable, degradable and conventional carrier bag materials did not degrade quickly in any of the natural environments examined and in some cases, formulations merely disintegrated into small pieces (such as those in the open-air environment). There are considerable concerns about the accumulation of microplastics in the environment and it remains to be established whether fragmentation into microplastics presents greater environmental risks than the original intact items of litter. From the perspective of cleansing, fragments are certainly considerably harder, if not infeasible, to remove from the environment compared to intact items.

It is of importance to understand the actual environmental degradability performance of materials which are claimed to have enhanced degradation properties as these could make consumers more relaxed about discarding, or even littering them, rather than reusing and recycling. Due to the growing interest in products which indicate enhanced environmental outcomes, we should be careful that such products do not inadvertently encourage littering or compromise alternative approaches to waste reduction such as recycling.

Designing products specifically to degrade in the environment is very challenging because of the natural variability between environment types, as illustrated by the present study. In addition, formulations that are designed to be less durable may compromise recyclability since they decrease the durability of the recyclate. It is also important to set the benefits of the various formulations into a wider context since reducing the diversity of polymers that are widely used is likely to facilitate greater recycling. To gain the maximum benefit from materials with enhanced rates of degradability, it is essential to have clear definitions and product labelling to indicate appropriate usage and disposal (Thompson et al., 2009a).

Clearly there may be drivers for the design of products with modified degradability other than deterioration in the natural environment, but in order for any these potential benefits to be realised it is essential that such products have a high probability of actually reaching the appropriate waste stream. This will require availability of a dedicated waste stream, the appropriate infrastructure such as an industrial composting facility and sufficient understanding amongst consumers to correctly separate their waste accordingly. Some nations actively promote the use of carrier bags with biodegradable, degradable or compostable formulations, for example using fiscal measures or other legislation. This includes some nations with relatively poor waste management infrastructure where the likelihood of these products reaching an appropriate waste stream seems low. Given the findings of this study, the benefits of such policy measures are unclear.

5.5 Conclusion

In conclusion, the current experiment has shown that biodegradable, oxo-biodegradable and conventional plastic formulations persist and remain functional in the soil and the marine environment for over 3 years. The compostable bag was the only material that completely disappeared from the experimental test rig within the marine environment and did this within a 3-month period. Hence the current study indicated that over a 3-year period, none of the materials examined could be relied upon to degrade (e.g. reduced effects on aesthetics or biota) in all three environments. Moreover, it was not clear that the materials which claimed to have enhanced degradation consistently degraded faster than conventional polyethylene. Deterioration was influenced by the receiving environment, but this was not consistent among material types. Hence, we suggest that statements about the degradation of products should be made in conjunction with statements on the receiving environment (air, soil, water) and timescale to which the claims relate. Since degradable and compostable materials are not compatible with available recycling, it is also important that the users are informed of the appropriate disposal route which in most circumstances will be disposal to the residual waste stream. It is only by providing accurate, unambiguous and complete guidance to the user regarding disposal that the potential benefits of novel materials can be realised without the negative consequences that could result in inappropriate disposal as well as unintended environmental consequences.

Chapter 6

General Discussion; Solutions and Future work

Napper, I.E., Pahl, S., Thompson, R.C., 2018. Marine Litter: are there solutions to this global environmental problem? Plastic Legacies (in press).

Abstract

Marine litter is a growing environmental problem which can pose threats to the environment, the economy and human wellbeing on a global scale. Over 300 million tons of plastic is produced annually and around 75% of all marine litter is plastic. Plastic litter is widespread in aquatic ecosystems and comes from a variety of sources. Therefore, there are abundant solutions to deter and reduce it from entering different environments. The improvement of waste management frameworks internationally is a critical element in preventing loss into the environment. Education is also crucial for promoting change in reducing waste, limiting indiscriminate disposal and increasing awareness of marine litter. This is particularly effective with citizen focused activities such as beach cleaning. Industry has a key role in helping reduce the potential for end of life plastic to become waste and litter, where appropriate decisions should be meet at the design stage of a product considering the circular economy. Solutions linking to management strategies and policies are also already in place to reduce marine litter. However, further measures and actions are required globally. Additionally, the thesis' contribution to both the issues and solutions to marine debris are discussed, and the limitations and future research directions analysed.

6.0 General Discussion; Solutions and Future work

6.1 Introduction

Modern lifestyles generate considerable quantities of waste on a daily basis. Marine litter (also called marine debris) is solid waste that has been discharged into the marine environment resulting from activities on land or at sea. Plastics represent a substantial fraction of the municipal waste stream as well as marine litter. As a consequence of the durability of plastics, they also have considerable persistence within the environment and landfill. This is a growing issue; just a few decades ago much of our waste was composed of organic, degradable materials yet in the last seven years we have produced more plastic items than in the entire century that preceded (PlasticsEurope, 2016).

There is increasing awareness around the accumulation of litter in marine, freshwater and terrestrial environments, which results from a variety of different sources (Eerkes-Medrano et al., 2015). The importance of various sources varies considerably geographically, but on a global scale it is widely recognised that most litter in the marine environment comes from land-based actions such as general littering, dumping of waste and loss during waste collection as well as from inappropriately managed landfill sites. Litter from shipping and other maritime activities contributes a much smaller proportion (Jambeck et al., 2015; Mehlhart and Blepp, 2012).

It has been suggested that the accumulation of marine litter and its fragmentation has led to one of the most ubiquitous and long-lasting recent changes to the surface of our planet (Barnes et al., 2009). This debris is widely reported in the environment where it has accumulated at the sea surface (Law et al., 2010), on shorelines of even the most remote islands (Barnes, 2005), in the deep sea (Bergmann and Klages, 2012; Woodall et al., 2014) and in arctic sea ice (Obbard et al., 2014).

Globally around 75% of all marine litter is plastic, with other materials such as glass, metal and paper contributing in much smaller relative proportions. Even though the large majority of marine litter is plastic, the timescales for degradation of plastic items is not known with certainty and will depend on the chemical nature of the material, the characteristics of the environment in which they persist and the manner in which degradation is measured (Andrady and Neal, 2009). However, it is clear that from the substantial quantities of marine litter that are entering aquatic habitats daily, this litter presents a range of negative economic and environmental consequences (Jambeck et al., 2015; Werner et al., 2016).

Its accumulation has been identified as a major global conservation issue and a key priority for research (Sutherland et al., 2010). It has also been identified as a major issue by the United Nations Environment Assembly and in the G7 Leader´s declaration 2015 (GESAMP, 2016; UNEP, 2017; Werner et al., 2016). There is broad recognition that marine litter presents a substantial problem therefore the key action must be to reduce the quantity entering the oceans from the land. In addition, we need to further define the scale of the problem in terms of quantities of litter and the types of impact, but in our view, there is sufficient consensus that there is already enough evidence for us to take action to stem the flow of plastics to the environment. This chapter considers the problem and some of the solutions that are currently being implemented or considered.

6.2 Waste Management

The accumulation of plastic litter in the oceans is actually a symptom of a wider, more systemic problem of the linear use of plastic materials and the rapid accumulation of waste. Litter can be defined as something of little or no value and hence the problem may be exacerbated because plastics are inexpensive, which facilitates short-lived applications. The vast variety of different plastic types presents a complication for the viability of recycling and the quantity and diversity of single-use products is putting increasing pressure on waste management infrastructures. Studies have shown that unless waste management improves profoundly in the coming years, by 2025 the amount of plastic waste entering the ocean from land will be three times greater than it was a decade previously (Jambeck et al., 2015). Consequently, effective waste management and recycling is a critical element in preventing loss into the environment.

Waste management frameworks are typically designed to help minimize loss to the environment, but management practices can differ considerably between nations. Incorrectly managed landfills or waste management systems may cause waste to escape into the environment. In industrialized countries, waste that is deposited in landfills is usually covered regularly with soil or a synthetic material, and the landfill is cordoned by a fence to prevent any debris accidentally leaving. However, in developing regions this is often not the case (Barnes et al., 2009; Jambeck et al., 2015). There are also circumstances in which waste management will not suffice in stopping plastic leaking into the ocean. For example, in the immediate aftermath of a tropical storm, resource management is understandably focused on human health, toxic spills and air quality as opposed to waste management (Institute of Medicine, 2007).

There are solid waste management strategies that are used as alternatives of landfill, for example, recycling (Singh et al., 2017), reuse or upcycling (recycling to improve a materials value) (Braungart, 2013). However, the applicability of different approaches depends on the quality of the waste, and a common issue is that the end of life disposal pathway has not been appropriately considered at the design stage. If the quality is insufficient, energy recovery via incineration is also an option. Even in developed countries with robust waste management infrastructure, there are obstacles to recycling, including the lack of availability of collection points, contamination of recycling feedstock, and the limited marketability of some recycled material (Andrady, 2005; Law, 2017). Residues from plastic recycling can also unintentionally escape into the environment (Moore, 2008).

Focusing on the 35 top-ranked countries for mass of mismanaged plastic waste, Jambeck et al. (2015) suggest that to achieve a 75% reduction in the mass of this waste, waste management would have to be improved by 85%. This strategy would require time and substantial infrastructure investment primarily in low- and middle-income countries (Löhr et al., 2017). Within these countries, the main focus is now on improving solid waste collection and management, and some outstanding efforts are being made. Indonesia, for example, set targets at the 2017 World Ocean Summit to reduce plastic waste in 25 coastal cities and reduce marine litter by 70% by 2025 (UN Environment, 2017).

6.3 Education and Behaviour Change

Education is crucial for promoting change in reducing waste, limiting indiscriminate disposal and increasing awareness of marine litter, especially if they include behaviour change principles and go beyond merely teaching facts. Programs to help encourage this are being considered and could be very successful in reducing litter and waste. For example, a study by Hartley et al. (2015) found that school children in the UK significantly improved their understanding of the causes and negative impacts of marine litter, as well as their self-reported behaviour, after an education intervention related to plastic marine debris. Education and behavioural change of children is crucial as they have an important influence on their peers, parents and community (Hartley et al., 2015). Hartley et al., (2018) demonstrated similar results following a European video contest for schools and training specifically tailored to educators. Therefore, making resources available to incorporate marine litter awareness into the school curriculum could prove to be a very successful tool for collective understanding and spreading knowledge of the issues.

Citizen focused activities such as beach cleaning are also well recognized for their educational value as well as in terms of the litter removed (Nelms et al., 2016) and may even have benefits for human wellbeing (Wyles et al., 2016). These can be combined with monitoring exercises and the involvement of local communities. Annual clean-up operations are now organized internationally (Barnes et al., 2009) and are often run by voluntary organizations. Volunteer involvement in two of the largest clean up schemes in the UK (Marine Conservation Society Beach Watch and Keep Scotland Beautiful National Spring Clean) has been estimated to provide a value of approximately £118,500 annually in term of cleaning, which suggests that the total cost of voluntary action to remove marine litter is considerable (Mouat et al., 2010).

Additionally, we need to consider the role of society and the processes of social perception and influence amongst a range of actors (Hartley et al., 2018b). This is critical because unless the efficacy of solutions is properly evidenced and understood there is a significant risk that interventions taken in haste will not be socially acceptable and/or may lead to unintended negative consequences.

In simple terms, it is important to raise awareness about the need to dispose of end of life items properly and not to litter, in addition to raising awareness about often unnecessary plastics use – such as single use carrier bags, cutlery, plates, drinking straws. However, educating the public about the damage alone is unlikely to achieve the substantial change required; we need to harness powerful motivators for managing waste differently, such as the great affinity many children (and adults) feel with the ocean (Pahl et al., 2017). Moreover, systemic change is necessary beyond raising public awareness to reduce the substantial accumulation of end of life plastic waste. This systemic change will require transition within the industry, right from the product design stage in order to ensure maximum value can be recovered at end of life. In the absence of such changes, educating the public alone is to some extent merely educating about a system that is currently broken.

6.4 Industry and the Circular Economy

Industry has a key role in helping reduce the potential for end of life plastic to become waste and litter. Current use of plastic materials is predominantly linear, and this is leading to a rapid accumulation of persistent waste. Long-term sustainable solutions lie in moving from a linear economy towards a more circular economy (Ellen MacArthur Foundation, 2016; European Commission, 2012). This approach involves utilising more sustainable production and consumption patterns, and the circular use of materials that will ultimately lead to waste reduction, for example, designing products for reuse/recycling and also avoiding unnecessary plastics use. Most plastics are inherently recyclable, yet many single-use items are not designed to be widely compatible with recycling. A key challenge therefore is to ensure end-of-life disposal via recycling is appropriately considered at the design stage. For these interventions to be successful, a tax may be required on non-recyclable products or an incentive may motivate the use of recycled content in new products so as to encourage reuse and or design for recyclability.

In addition, we need greater awareness about the applicability of alternative approaches, which from a narrow perspective, may appear to present environmentally friendly alternatives. These need to be considered in terms of the overall environmental footprint and how they interact with existing collection schemes to ensure there are not unintended negative consequences; for example, plastic products that are designed to have greater degradability or are made from renewable rather than fossil carbon sources.

Materials with enhanced degradability may reduce the amount of highly visible macroplastic waste. However, it is challenging to deliver products which are durable while in service, yet can degrade in a meaningful timescale if they become litter in the environment. Some formulations merely fragment compromising the potential for product re-use and accelerating the production of microplastic fragments (Thompson et al., 2009). Even when disposed properly, most degradable formulations are not compatible with recycling and can only be disposed of as residual waste to landfill or incineration. These plastics do have a role but may only present solutions in specific settings where the associated waste collection is specifically managed, provides conditions suitable for degradation and products are labelled accordingly to facilitate appropriate disposal. Similarly, altering the carbon source for plastic by utilizing plant based carbon, rather than fossil carbon from oil and gas, is to some extent a distraction. While this utilizes a renewable and hence a more sustainable carbon source, it will not, *per se,* reduce the generation of waste nor the accumulation of litter and may conflict with other uses of the resource.

In summary, industry has a key role in helping maximise the benefits that plastics products can bring to society, while at the same time helping to minimise emissions of plastic during life in service and at end of life. This requires greater recognition for unintended consequences, via extended producer responsibility. Had this approach been in place when the patent on use of microbeads in cosmetic products was first filed some 50 years ago, much unnecessary contamination and the eventual need for costly legislative measures could have been avoided. Similarly, it is now clear that some types of garment construction release fibres more quickly than others (Napper and Thompson, 2016). This is not in the interest of the consumers, because it means clothing wears out more quickly and it results in more rapid release of fibres to the environment. The key step is to consider this at the design stage to minimise the avoidable emission for synthetic textile fibres. In addition, development of washing machine filters to capture any released synthetic fibres in the washing cycle may be advantageous. The introduction of appropriate labelling on products to indicate the environmental footprint in terms of recycled content, material use and recyclability could be instrumental in guiding product choice along the supply chain. Such information is just as important to major retailers as it is to the consumer since it paves the way to helping ensure sustainability and ethical choices have been made upstream - taking the burden off of the consumer.

6.5 Policy Measures

The United Nations Development Goals request nations to "prevent and significantly reduce marine pollution" by 2025 (United Nations General Assembly, 2015). This may be facilitated by policy measures to help reduce unnecessary plastics usage. However, there are numerous applications where plastic is clearly the best material for a particular task and here policy measures my help nudge behaviours toward more circular material use, for example deposit return schemes. Ultimately these measures need to help us move to more resource efficient circular material use (Lieder and Rashid, 2016). The European Union has set this in motion in their "Action plan for the Circular Economy" implementing a waste hierarchy in which prevention, reuse, recycling and energy recovery are favoured over landfill in this respective order (European Commission, 2015).

Solutions linking to management strategies and policies are also already in place to reduce marine litter (GESAMP, 2015). This can include the use of targets, taxes, education and bans. Banning microbeads in cosmetics is an example of such legislation. However, based on the levels of concern and the scale of the marine litter problem in general it would appear that the current measures used are insufficient. In some cases, there are difficulties associated with enforcement; for example, the regulation of dumping at sea (MARPOL) is extremely difficult to monitor.

Taxes introduced on plastic items have already shown to be instrumental in changing consumer behaviour towards plastic. A fifteen euro cent tax on plastic bags in Ireland led to a 90% reduction of plastic bag usage in the early 2000s (Convery et al., 2007). The tax has successfully removed the widespread use of plastic bags throughout Ireland and has led to similar taxes globally. In San Francisco, a ban on conventional plastic bags has been introduced forcing the use of alternative bags; for example cotton tote bags (Romer, 2010). Unfortunately, these taxes do not always work effectively. South Africa has struggled to achieve similar reduction rates in plastic bag usage through taxes (Dikgang et al., 2012).

Plastic debris does not recognise international boundaries and enforcement of regulations needs to be applied at an international scale. Global commitment and goals provide a good basis for this, but measures and the actions then need to be translated to regional and national levels. There are substantive regional differences in the causes of plastic pollution, both on land and at sea, and solutions will therefore only be effective if they also take into consideration local conditions; for example in terms of waste management infrastructure (Jambeck et al., 2015; Van Franeker and Law, 2015). Hence, design and implementation of effective, efficient and legitimate actions needs to be based on a thorough understanding of the issue as well as the local context.

6.6 Future work

It is important to recognise that the accumulation of plastics in the ocean is largely avoidable. By comparison with many other current environmental challenges, the benefits resulting from the use of plastics are not directly linked to the emission of plastic debris to the environment or to degradation of the environment. Consequently, in theory at least, it is possible for society to retain the benefits of plastic products and at the same time reduce the quantity of plastic litter entering the environment (Thompson, 2015). However, to further quantify and understand the specific impact different sources of plastic debris can have within the environment, each source needs to be analysed. Emphasis should be on further understanding the routes different sources have into various environments. This will then provide guidance to different entities (such as industry or government) to make decision towards sustainable management and reduction. Additionally, the wider implications of its accumulation should continue to be analysed, particularly in terms of human health.

6.7 Conclusion

While the suite of potential solutions is well recognised, there is no one size fits all solution. In the current thirst for action, a major challenge is matching the most appropriate solutions to particular aspects of the problem. To address this type of challenge we think an inter-disciplinary, inter-sector approach will be necessary to reconfigure how modern societies engage with plastics. Profiting from the current groundswell of public opinion, transformative change could be achieved by harnessing the potential of the social and behavioural sciences to understand and influence the decisions and behaviour underlying the plastics challenge. In addition, arts and humanities can be helping to inspire creative change yet be firmly integrated with the natural sciences' evidence base.

Beyond integrating different academic perspectives, such an effort should work with stakeholders, practitioners, policy makers and industry. This approach would be able to capture how plastics are currently viewed and managed in society, truly representing the user perspective. It would also identify and respond to both intrinsic and extrinsic motivations plus constraints along the supply chain. More importantly, the approach can demonstrate how the current situation may change by facilitating evidence-based dialogue with design and waste management, economic and legal studies, arts and other creative disciplines. Looking at the system in such an integrated way has the potential to trigger an irreversible course towards more sustainable design, use and disposal of plastics and could be adapted to other societal challenges.

This thesis addresses different sources to marine litter and shows that they are a symptom of a more systemic issue originating on land that relates to the design, the use and the disposal of waste (particularly single-use plastic items). Solutions to the marine litter problem require coordinated action amongst industry, policy and the general public, on a local to global level. This will involve the interaction of consumers, producers, policy makers, managers, local residents, tourists, industries, and many other factors. Unity, collaboration and ownership of solutions between these groups will provide the greatest potential for success. Currently the scope, timeframe and dynamics of all these initiatives are distinctly different and orchestration at all levels in close collaboration with each other is lacking.

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