

Chapter 9

Sources and Pathways of Microplastics to Habitats

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Abstract Identifying and eliminating the sources of microplastic to habitats is crucial to reducing the social, environmental and economic impacts of this form of debris. Although eliminating sources of pollution is a fundamental component of environmental policy in the U.S.A. and Europe, the sources of microplastic and their pathways into habitats remain poorly understood compared to other persistent, bioaccumulative and/or toxic substances (i.e. priority pollutants; EPA in U.S. Environmental Protection Agency 2010–2014 Pollution Prevention (P2) Program Strategic Plan. Washington, USA, pp. 1–34, 2010; EU in Official J Eur Union L334:17–119, 2010). This chapter reviews our understanding of sources and pathways of microplastic, appraises terminology, and outlines future directions for meaningfully integrating research, managerial actions and policy to understand and reduce the infiltration of microplastic to habitats.

Keywords Hypothesis · Micrometre · Emissions · Sewage · Storm-water · Textile · Exfoliants

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9.1 Defining Sources and Pathways of Microplastic

Since the first review of microplastic (Browne et al. 2007), a number of terms have been used to describe and categorize sources of microplastic. Some authors have used the terms “primary” and “secondary” to distinguish between sources of microplastic, in which they borrow terminology from atmospheric sciences (Arthur et al. 2009; Cole et al. 2011). In these cases, “primary sources” are those in which microplastic is intentionally produced through extrusion or grinding, either as precursors to other products (e.g. plastic pellets; Costa et al. 2010) or for direct use (e.g. abrasives in cleaning products or rotomilling), whilst “secondary sources” of microplastics are those formed in the environment from the fragmentation of larger plastic material into ever-smaller pieces (Arthur et al. 2009; Cole et al. 2011). Using similar ideas, Andrady (2011) described runoff as a “direct source” (sewage or storm water) whilst fragmentation of existing plastic debris was described as “indirect source” of microplastic to the environment.

Although using adjectives to categorize sources may be helpful, these terms introduce jargon without clearly identifying the actual sources and conflate sources with the pathway by which microplastic enter habitats, which may in turn confuse scientists, public, industry and government. In this chapter, I argue that these problems can be overcome if we choose alternative terms for *sources* that identify the place, person, company, or product where the microplastic originates and use separate terms to describe the *pathways* of microplastic from its source to a habitat.

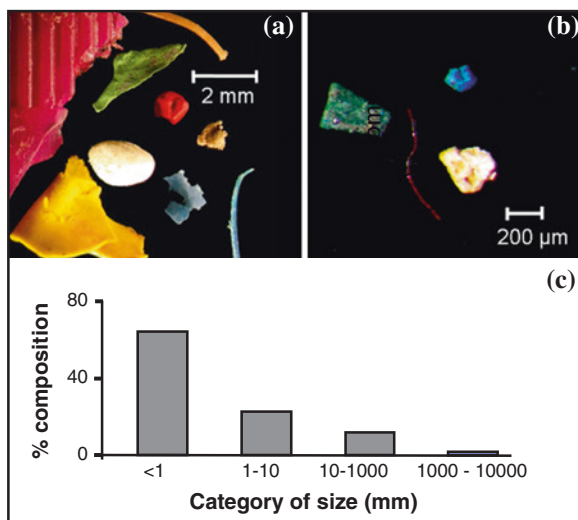
Based upon our current understanding there are four types of sources (i) larger plastic litter, (ii) cleaning products (Zitko and Hanlon 1991; Gregory 1996; Derraik 2002); (iii) medicines; and (iv) textiles. For the latter, I have chosen to use a global case-study to illustrate how one can gain a more meaningful and scientific understanding of the sources and pathways of microplastic through developing better programs of research and monitoring that integrate advances in forensics (e.g. vibrational spectroscopy to identify the shape and type of microplastic), logic and experimental design (i.e. making observations, developing explanatory models, testing explicit hypotheses about composition and spatial patterns; Underwood 1997) and statistics. Throughout the chapter, I have chosen to define microplastic as micrometre-sized particles of plastic because this is consistent with previous work on this topic (Browne et al. 2007, 2008, 2010, 2011, 2013; Costa et al. 2009; Claessens et al. 2011; Rochman et al. 2013; Van Cauwenberghe 2013) and the globally ratified use of the prefix “micro” for measures of length under the International System of Units. Other authors have chosen to ignore the International System of Units definition of “micro” and have instead chosen to use <5 mm to define microplastic, a philosophical discussion about which people should use is beyond the scope of this review.

9.2 Larger Plastic Litter

Sources. Larger plastic debris originates from maritime activities including shipping, fishing (e.g. Merrell Jr 1980, Ramirez-Llodra et al. 2013; Galgani et al. 2015), recreation and offshore industries (e.g. oil, gas). These sources are, however, likely to be much smaller than terrestrial sources. Whatever the source, larger plastic litter (including millimetre-sized pre-production pellets) is likely to be an important source of microplastic. Irregularly shaped fragments are abundant in intertidal and oceanic habitats with the size-frequency of plastic debris skewed towards smaller debris. This suggests that microplastic can originate from the fragmentation of larger objects that causes ever smaller pieces of plastic to be present in the environment (Browne et al. 2007, 2010; Morét-Ferguson et al. 2010; Collignon et al. 2012; Fig. 9.1a–c).

Pathways. Fragmentation is the pathway, by which plastic debris breaks into smaller pieces, which is distinct from the more subtle processes of degradation that reduces the molecular mass of plastic debris (Andrady 2011, 2015). These processes occur through the action of light (photolysis), heat and oxygen (thermal-oxidation), water (hydrolysis), organisms (see review by Andrady (2011) for more details about weathering experiments with pieces of plastic >1 mm) and physical abrasion by particles of sediment. Laboratory and field experiments are required to determine the relative importance of these processes in generating the sizes and shapes of micrometre-sized plastic that we find in habitats. One such laboratory experiment by Davidson (2012) showed that each time a single isopod (*Sphaeroma quoianum*) burrows into a floating dock made of expanded polystyrene they can produce and release between 4900 and 6300 micrometre-sized fragments of polystyrene. There is also a modest literature on plastic degradation in marine habitats (Andrady and Pegram 1989; Andrady 2003; Gregory and Andrady 2003; Corcoran et al. 2009; Cooper and Corcoran 2010; O’Brine and Thompson 2010). But the rigour of these

Fig. 9.1 Fragments (a–b) and sizes (c) of plastic debris found in the Tamar Estuary. Reprinted adapted with permission from Browne et al. (2010). © 2010 American Chemical Society



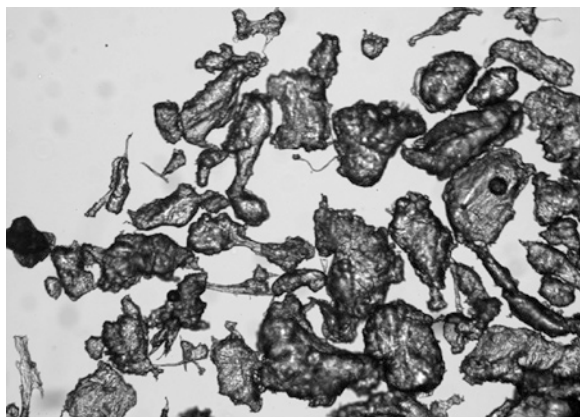
studies varies with some lacking many of the ecological developments in designing, implementing and analysing manipulative field experiments (Underwood 1997). So far, no one has experimentally deployed micrometre-sized plastic (or millimetre-sized pellets) in habitats to determine the rates at which it fragments into smaller pieces. Thus, the role of pellets as major sources of microplastic is unclear.

9.3 Cleaning Products

Sources. Another source of microplastic is from industrial and domestic cleaning products that use microplastic as an abrasive scrubber (Browne et al. 2007). For instance, surfaces of buildings, machinery and boats can be cleaned and prepared (e.g. smoothed, roughened, shaped) using ‘media blasting’, where small plastics (e.g. polystyrene, acrylic, polyester, poly-allyl-diglycol-carbonate, urea-, melamine- and phenol-formaldehyde; 0.25–1.7 mm; DOD 1992) and other types of granules (e.g. sand) are propelled onto a surface using a centrifugal wheel or pressurized fluid/gas (Wolbach and McDonald 1987; Abbott 1992; Gregory 1996; Neulicht and Shular 1997; Anonymous 1998). Although ‘media blasting’ has been suspected of being a source of microplastic to habitats there has been no scientific work to (i) characterize the number of industries using this technique, (ii) the size, shape and amount of microplastic used in the process of cleaning and (iii) the quantity of particles emitted into, or found within, the environment through this source.

More work has been done for microplastics used as physical abrasives in domestic products. Fendall and Sewell (2009) qualitatively showed that the size and shape of microplastic in such products varies (Fig. 9.2). By examining four different facial cleansers with labels that indicated they contained particles of polyethylene, they found that the size of the particles ranged from 4.1 to 1240 μm in diameter, and consisted of uniform spheres, ellipses, rods, fibres and granules (Fig. 9.2). For granules this presents a problem because it will be very difficult to differentiate whether they come from cleaning products or from the fragmentation of larger

Fig. 9.2 Microplastic (polyethylene) fragments found in facial cleansers (Photo: M. Sewell, University of Auckland)



articles of plastic debris. Using vibrational spectrometry Zitko and Hanlon (1991) found that 47 % of the mass of the contents of a single bottle of skin cleanser was made up of irregular fragments of polystyrene (100–200 μm). A separate study that used vibrational spectrometry showed replicate formulations of hand cleansers between 0.2 and 4 % of their mass made up of polyethylene, whilst for facial cleansers it was 2–3 % (Gregory 1996), though it is important to note that this study did not report particle numbers <63 μm in size, which may account for the smaller amounts recorded. Gouin et al. (2011) estimated the emission of microplastic from cleaning products in the U.S. by combining estimates of sales figures and assuming proportions of polyethylene were 10 % by volume. From this, the authors calculated that each year the U.S. could be emitting 263 t of micrometre-sized fragments of polyethylene from domestic cleaning products. Given that the type of polymer (e.g. polyethylene, polystyrene) and proportions of microplastic can vary from 0.2 to 47 %, it seems that more work is needed to test individual products and different batches so that we can provide precise, accurate and ground-truthed estimates of microplastic emissions from cleaning products.

Pathways. Microplastics used in cleaning products are thought to transfer to habitats through sewage and storm water (Fig. 9.3). The quantities of microplastic, however, in water or sediment from habitats, sewage or storm water are unknown because they are interspersed with large concentrations of organic matter, and because it is difficult to distinguish uniform spheres, ellipses and granules with a biofilm from natural particles. Some of these problems may be overcome with the application of chemical techniques to remove organic matter (Claessens et al. 2013) and vibrational spectroscopes that can map microplastic in environmental samples (Harrison et al. 2012).

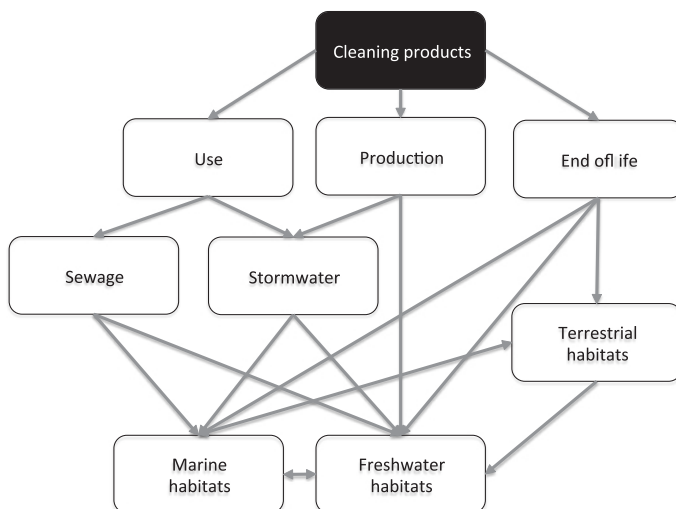


Fig. 9.3 Sources and pathways of microplastic from cleaning products into habitats. *Gray arrows* indicate hypothesized pathways. There are no black arrows because there is currently no research showing evidence of these pathways

9.4 Medicines

Sources. Ingestible and inhalable medicines containing microplastic are used to deliver drugs to the organs of humans and farmed animals (terrestrial and aquatic) because the microplastic can translocate from their lungs or guts into their circulatory system (Thanoo et al. 1993; Dalmon et al. 1995; Curley et al. 1996; Hussain et al. 2001; Matsusaki et al. 2001; Wen et al. 2003; Kockisch et al. 2003; Corbanie et al. 2006). There has, however, been no work to synthesize information about the different types of polymers and sizes of microplastic used in medicines because this information does not seem to be readily available.

Pathways. Like microplastics in cleaning products, microplastics from medicines are likely to transfer to habitats through sewage and storm water, or more directly through treating diseased animals in aquaculture and farming. There is, however, no quantitative work evaluating how much plastic is taken up by animals and excreted compared to those retained in tissues. As such, the quantities of medical microplastic in water or sediment sampled from habitats, sewage or storm water are unknown. Research is needed to synthesize a complete inventory of these polymers so that samples from humans, sewage, storm water, wildlife and habitats can be tested for the presence of these polymers. Some of the polymers used are thought to be biodegradable (Matsusaki et al. 2001), whilst others can be composed of more durable polymers such as polycarbonate and polystyrene (Thanoo et al. 1993; Dunn et al. 1994). The rates and mechanisms of degradation inside human tissues may not be the same as in wildlife or habitats. Moreover, just because a polymer degrades, does not necessarily mean that the resulting metabolites are not toxic themselves: So, research is needed to determine how safe these particles are in humans, wildlife and habitats.

9.5 Textiles

Many people have attempted to examine the sources of microplastic but a lack of a hypothesis-driven framework has meant that sources and pathways are poorly understood. More useful understanding about the sources and pathways of microplastic to habitats comes from work done on fibres that originate from textiles and clothing (Browne et al. 2011). The following case study is provided to illustrate how one can understand better the sources and pathways of microplastic (Browne et al. 2011). The work was done in four phases by examining microplastic in (i) sediment from sandy shores worldwide; (ii) sediment from replicated sub-tidal areas where sewage sludge had, and had not, been discharged; (iii) effluent from replicate treatment plants; and finally; (iv) effluent from manipulative experiments involving washing machines.

- (i) *A global program to sample sediment from sandy shores.* Between 2004 and 2007 samples of sediment were collected from sandy beaches in Australia, Oman, United Arab Emirates, Chile, Philippines, Azores, USA, South Africa,

Mozambique and the U.K. During collection (and in all work), cotton clothing was worn rather than synthetic items to prevent samples being contaminated by plastic fibres. Samples were collected by working down-wind to the particular part of the highest strandline deposited by the previous tide. Using established techniques, sediment was sampled to a depth of 1 cm and microplastic and sediment was quantified using established techniques (Browne et al. 2010, 2011). Two explanatory models for pathways of microplastic in habitats were put forward to explain spatial patterns of microplastic.

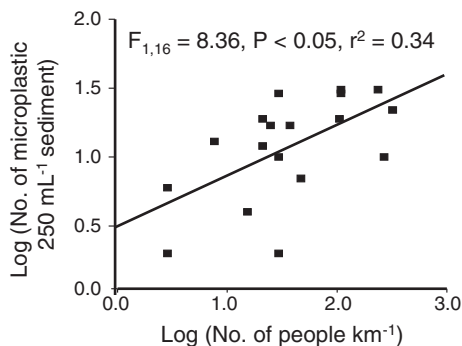
If spatial patterns of microplastic result from the transportation of natural particulates by currents of water (*Model 1*), we expected shores that accumulate smaller-sized particles of sediment would accumulate more microplastic (*Hypothesis 1*). Alternatively, spatial patterns may be influenced by sources of microplastic (*Model 2*). Over the last 50 years the global population density of humans had increased by 250 % from 19 to 48 individuals km^{-2} (UN 2008), and during this time the abundance of microplastic had increased in pelagic habitats (Thompson et al. 2004). Previous observations had suggested that there was a greater abundance of larger items of debris along shorelines adjacent to densely populated areas (Barnes 2005). This led to the prediction that there would be more microplastic along shorelines adjacent to densely populated areas (*Hypothesis 2*). The work showed that microplastic contaminated all 18 shores examined (Fig. 9.4) with more microplastic in sediments collected from densely populated areas (linear regression, $F_{1,16} = 8.36$, $P < 0.05$, $n = 18$, $r^2 = 0.34$; Fig. 9.5), but there was no relationship with the quantity of smaller-sized particles of sediment. Thus, there was evidence to support *Model 2* but not *Model 1*.

To examine the pathway of this microplastic onto shorelines, forensic analysis was used to gather crucial observations about the shapes and types of polymers that made up the microplastic. This showed that the microplastic was mostly made up of synthetic fibres that consisted of polyester (56 %), acrylic (23 %),



Fig. 9.4 Spatial extent of microplastic in sediments from 18 sandy shores. The size of filled-circles represents number of microplastic particles found. Reprinted adapted with permission from Browne et al. (2011). © 2011 American Chemical Society

Fig. 9.5 Relationship between population-density and number of microplastic particles in sediment from sandy beaches. Reprinted adapted with permission from Browne et al. (2011). © 2011 American Chemical Society



polypropylene (7 %), polyethylene (6 %) and polyamide (3 %). Previous observations had shown that coastal habitats receive millions of tonnes of sewage each year (CEFAS 1997) and that sewage can contain microplastic fibres (Habib et al. 1996; Zubris and Richards 2005) because although larger debris is removed during the treatment of sewage, filters are not specifically designed to retain microplastic. *Model 2* was therefore refined to include sewage as the pathway of synthetic fibres to marine habitats causing greater quantities of microplastic fibres in areas adjacent to densely populated areas (*Model 2.1*). The next step was then to test this model to determine whether there was evidence to support the model that the discharge of sewage was an important pathway of microplastic fibres into marine habitats.

- (ii) *Comparison of sub-tidal areas where sewage sludge had been discharged with reference areas.* Previous observations from coastal habitats of the U.K. suggested that each year treatment plants discharge >11 km³ of sewage effluent into coastal habitats (CEFAS 1997) and for nearly 30 years, a quarter of U.K. sewage sludge was dumped at 13 designated sub-tidal disposal-sites around the coast, until this stopped in 1998 through implementation of The Urban Waste Water Treatment Regulations 1994 (Fig. 9.6; CEFAS 1997; British Government 1994). Using replicate disposal-sites and reference-sites allowed us to test *Hypothesis 2.1* that sediments from disposal-sites would contain larger quantities of fibres in their sediments and that the shape and types of polymers that make up the microplastic would resemble those found on shores. For this, van Veen grabs deployed from boats collected replicate samples of sediment from two reference-sites and two disposal-sites in the English Channel and the North Sea (U.K.). Despite sewage not being added for more than a decade, disposal-sites still contained >250 % more fibres than reference sites (Fig. 9.7; ANOVA, $F_{1,16} = 4.50$, $P < 0.05$). Again the types of fibres were dominated by polyester (78 %) and acrylic (22 %).

During discussions with the sewage treatment authorities they explained that filters are not specifically designed to retain microplastic, which suggests that discharges of sewage effluent could also be a pathway of fibres from treatment plants

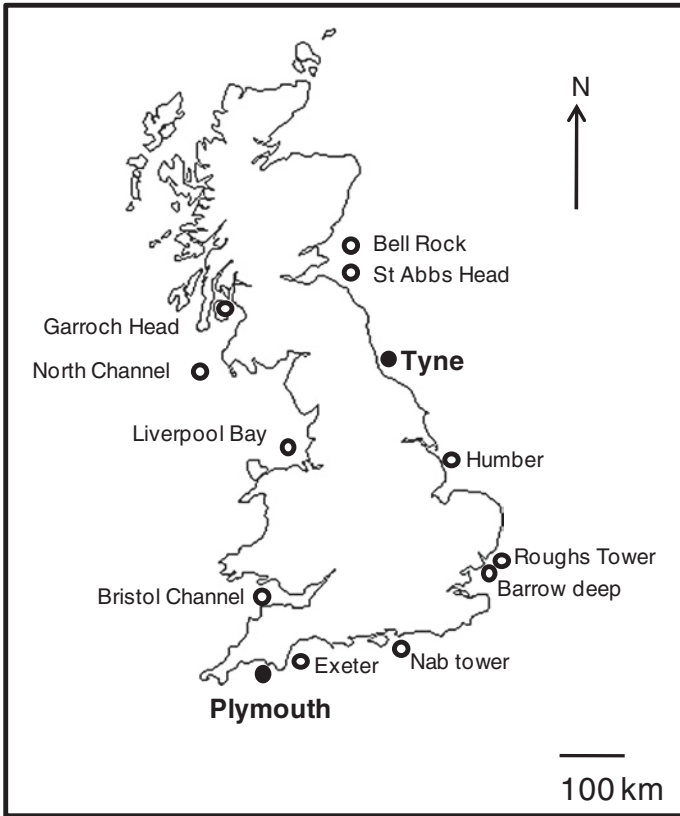


Fig. 9.6 Locations of U.K. sewage-sludge disposal sites (1970–1998) (CEFAS 1997). Plymouth (English Channel) and Tyne (North Sea) disposal sites presented as *filled black circles*, whereas the other 11 sites are with *open circles*

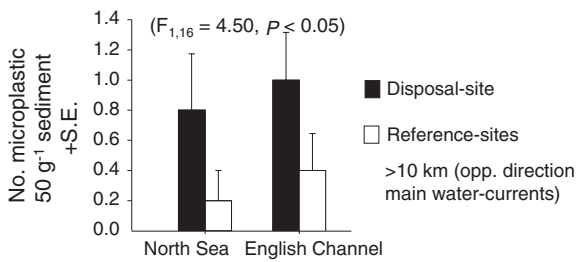


Fig. 9.7 Abundance of microplastic in sediments from disposal sites for sewage and reference sites at two locations in the U.K. (Tyne and Plymouth). Values expressed as mean ± S.E. Reprinted adapted with permission from Browne et al. (2011). © 2011 American Chemical Society

Fig. 9.8 Abundance of microplastic in effluent discharged from two separate tertiary-level treatment plants (West Hornsby and Hornsby Heights, NSW, Australia). Values expressed as mean \pm S.E.

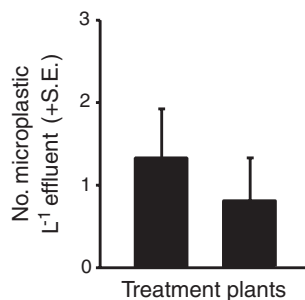
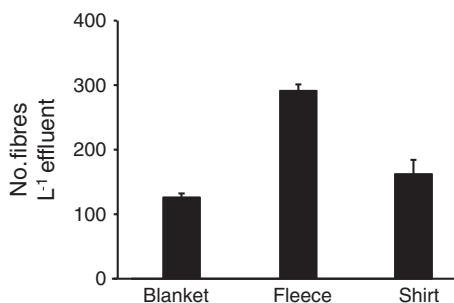


Fig. 9.9 Number of polyester fibres discharged into wastewater from using washing machines with blankets, fleeces, and shirts (all polyester). Reprinted adapted with permission from Browne et al. (2011). © 2011 American Chemical Society



to habitats (*Model 2.2*). To test this model we examined the hypothesis that discharges of sewage effluent would contain similar proportions of polyester and acrylic fibres. As expected, polyester (67 %) and acrylic (17 %) fibres polyamide (16 %) dominated. These proportions of polyester and acrylic fibres resembled those contaminating intertidal and subtidal habitats (Fig. 9.8) suggesting that these microplastic fibres were mainly derived from sewage *via* washing-clothes rather than fragmentation or cleaning products. In recent years, the clothing industry has used textiles that contain >170 % more synthetics than natural fibres (e.g. cotton, wool, silk) and because proportions of fibres found in marine habitats and sewage resembled those used for textiles (78 % polyester, 9 % polyamide, 7 % polypropylene, 5 % acrylic; Oerlikon 2009) we counted the number of fibres discharged into wastewater from using clothes and garments.

(iii) *Experiments with washing machines.* Here, experimental work counted the number of fibres discharged into waste water from domestic washing machines used to launder clothing. To estimate the number of fibres entering wastewater from washing clothes and garments, three replicate washing machines were used with and without cloth (polyester blankets, fleeces, shirts). Effluent was filtered and microplastic counted. The experiments showed all garments released >100 fibres per litre of effluent, with >180 % more from fleeces (>1900 fibres per wash; Fig. 9.9), demonstrating that a large proportion of microplastic fibres found in marine habitats may be derived from sewage as a consequence of washing of clothes.

9.6 Outlook and Conclusion

In the future, contamination by microplastic is likely to continue to increase. Populations of humans are predicted to double in the next 40 years (UN 2008) and further concentrate in large coastal cities that will discharge larger volumes of sewage into marine habitats. The last case study provides a useful approach for identifying and quantifying sources and pathways of microplastic that should be extended to other sources, including medical and cleaning products, by screening sewage, storm water, habitats, wildlife and humans for the types of microplastic found in these products.

In parallel to this, I believe work is needed to reduce and eliminate sources and pathways of microplastic through (i) establishing and controlling inventories of materials; (ii) modifying the process of production by redesigning products so that they contain less hazardous substances; and (iii) using novel equipment and technology. This section now explores some of current opportunities for the public, scientists, engineers, industry and government to reduce sources and pathways of microplastic.

- (i) *Establishing and controlling inventories that detail the use and emissions of microplastics in products.* Inventories are frequently used by European (EA 2012) and U.S. government agencies (EPA 2010, 2012) to control emissions of pollutants. An open-access online inventory is urgently required for textiles, medicines and cleaning products containing microplastic so that we have accurate information about emissions of microplastic during their production, use and disposal. This should include information about the use and emissions of microplastic, in terms of dimensions of size (i.e. minimum, maximum, median, mode and mean) shape, numbers, mass, types of polymers and sales figures. Because industry has, on occasion, been unwilling to provide this information when requested (Rosner 2008), this will probably require a change in policy and specific funding for representative sampling so that measures are accurate and precise (Figs. 9.10 and 9.11).
- (ii) *Modifying the process of production and redesigning products so that they contain less hazardous substances.* Currently there are no published data on the effectiveness of modifying the process of production of products to reduce emissions of microplastic, since microplastic is not currently considered hazardous by policy-makers (Rochman et al. 2013). In response to advocacy from scientists and activists, several companies who make domestic cleaning products (e.g. Unilever, Johnson & Johnson) have agreed to replace microplastic with non-plastic particles. It is, however, unclear what alternatives they will use (Alumina; pumice; seeds of strawberries, blueberries, cranberry, evening primrose, grapes, kiwi or raspberry; stones of apricots, avocados, olives or peaches; peel of oranges or mandarin; castor or jojoba beads, shells of cocoa, coconuts, almonds or walnuts; coir; corn cob; salt; sugar; luffa, rice; macadamia nuts) or whether they will be more or less toxic to humans and wildlife, so scientific research is needed to find the most cost-effective alternative. Similar research is needed within the textile and clothing industry so that they produce cost-effective clothing that sheds fewer and less toxic fibres.

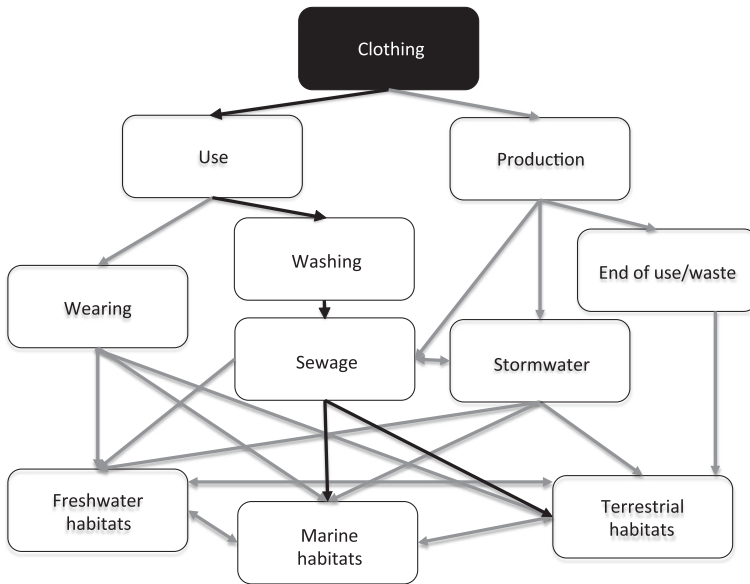


Fig. 9.10 Sources and pathways of fibres from textiles into habitats. *Gray arrows* indicate hypothesized pathways, *black arrows* indicate research that has been showing evidence of these pathways

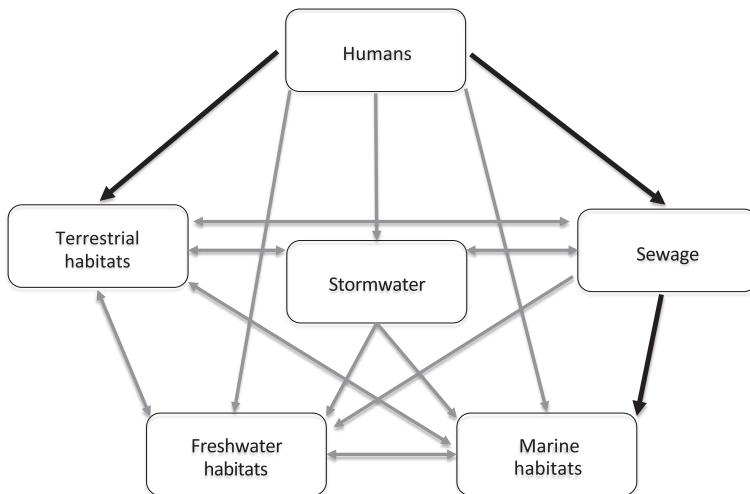


Fig. 9.11 Overview of sources and pathways of microplastic to habitats. *Gray arrows* indicate hypothesized pathways, *black arrows* indicate research that has been showing evidence of these pathways

These types of research are important to ensure decisions by policy-makers are based on robust scientific information as opposed to untested perceptions of the hazards of synthetic materials over those made from natural products.

- (iii) *Novel equipment and technology to reduce pathways of microplastic.* Filters for washing machines are a promising prospect for reducing emissions of fibres to sewage (www.environmentalenhancements.com); however, their effectiveness in reducing emissions has not been tested as yet. Work is also needed to determine how effective different types of sewage treatment are at removing the different sizes, shapes and types of polymers that represent the microplastic found in sewage. However, unless the microplastic can be isolated from the sludge or effluent there are still likely to be problems because sewage is added to soil as a fertilizer. To identify the place, company or product where the microplastic originates requires government, industry and scientists to work together and share information. For this to happen there needs to be policies that (i) provide funded frameworks to measure (and if necessary manage) sources and pathways of microplastic into the environment; (ii) balance the needs of industries, society and the environment. In the U.S., 16 persistent, bioaccumulative and toxic chemicals (i.e. aldrin, benzoperylene, chlordane, heptachlor, hexachlorobenzene, isodrin, lead, mercury, methoxychlor, octachlorostyrene, pendimethalin, pentachlorobenzene, polychlorinated biphenyl, tetrabromobisphenol A, toxaphene and trifluralin) are controlled in this way using the Emergency Planning and Community Right-to-Know Act 1986. The Community Right-to-Know provisions help to increase public's knowledge and access to information on chemicals at individual facilities, their uses, and releases into the environment. Through this facilities are obliged to share information with government, scientists and public to improve chemical safety and protect public health and the environment. Similar requirements are required in Europe under article 5 of the Directive 2008/105/EC where member states are obliged to establish an inventory of emissions, discharges and losses of priority pollutants listed in part A of Annex I (EU 2012). For microplastic, several other solutions could also help. For instance, polymers could be designed with unique chemical fingerprints (that identify particular sites of production or use), which remain even after the polymer has been physically or chemically degraded during its time in the environment. Alternatively, prior to a product being licensed for sale on the market place, information on the composition of polymers (and additives) in commercial applications could be made available to environmental scientists so that the environmental sources and pathways of materials can be quantified and managed if needed. Whatever developments take place, hypothesis-driven frameworks are required to identify and falsify sources and pathways.

In conclusion, if we are to use terms to describe the sources of microplastic they should aim to identify the origin (e.g. larger plastic litter, cleaning products, medicines, textiles, etc.) and separate terms should be used for the pathways (e.g. storm water, sewage). As researchers use more integrated hypothesis-driven frameworks and the chemical methods and inventories improve, it may be possible to be more specific (e.g. the place, person or company).

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