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Accumulation of microplastic on shorelines worldwide:
sources and sinks

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ABSTRACT

Plastic debris <1 mm (defined here as microplastic) is accumulating in marine habitats. Ingestion of microplastic provides a potential pathway for the transfer of pollutants, monomers and plastic-additives to organisms with uncertain consequences for their health. Here, we show that microplastic contaminates the shorelines at 18 sites worldwide representing six continents from the poles to the equator, with more material in densely populated areas, but no clear relationship between the abundance of microplastics and the mean size-distribution of natural particulates. An important source of microplastic appears to be through sewage contaminated by fibres from washing clothes. Forensic evaluation of microplastic from sediments showed that the proportions of polyester and acrylic fibres used in clothing resembled those found in habitats that receive sewage-discharges and sewage-effluent itself. Experiments sampling wastewater from domestic washing machines demonstrated that a single garment can produce >1900 fibres per wash. This suggests that a large proportion of microplastic fibres found in the marine environment may be derived from sewage as a consequence of washing of clothes. As the human population grows and people use more synthetic textiles, contamination of habitats and animals by microplastic is likely to increase.

KEYWORDS. Clothes, forensics, fragmentation, FT-IR, sewage, synthetic polymers, washing machines, wastewater, textiles.

BRIEFS. Global accumulations of microplastic

INTRODUCTION

We use >240 million tonnes of plastic each year¹ and discarded ‘end-of-life’ plastic accumulates, particularly in marine habitats¹, where contamination stretches from shorelines² to the open-ocean³⁻⁵ and deep-sea⁶. Degradation into smaller pieces means particles <1 mm (defined here as microplastic^{2,7,8}) are accumulating in habitats¹, outnumbering larger debris⁷. Once ingested by animals, there is evidence that microplastic can be taken up and stored by tissues and cells, providing a possible pathway for accumulation of hydrophobic organic contaminants sorbed from seawater, and constituent monomers and plastic-additives, with probable negative consequences for health⁹⁻¹⁶. Over the last 50 years the global population-density of humans has increased 250 % from 19 to 48 individuals per square km¹⁷, during this time the abundance of micrometer-sized fragments of acrylic, polyethylene, polypropylene, polyamide and polyester have increased in surface waters of the north-east Atlantic Ocean¹. This debris now contaminates sandy, estuarine and sub-tidal habitats in the United Kingdom^{1,6}, Singapore¹⁸ and India¹⁹. Despite these isolated reports, the global extent of contamination by microplastic is largely unknown. This has prompted the United Nations, Group of Experts on Scientific Aspects of Marine Environmental Protection, International Oceanographic Commission¹⁴, European Union¹⁵, Royal Society³ and National Oceanic and Atmospheric Administration (USA)¹⁶ to all identify the need to improve our understanding about how widespread microplastic contamination is, where it accumulates and the source of this material. If spatial patterns of microplastic result primarily from the transportation of natural particulates by currents of water, shores that accumulate smaller-sized particles of sediment should accumulate more microplastic. Alternatively, spatial patterns may be influenced by sources of microplastic; with more material along shorelines adjacent to densely-populated areas which already have a greater abundance of larger items of debris²⁰ and receive millions of tonnes of sewage each year²¹ which has also been shown to contain microplastic²²⁻²⁶. Although larger debris is removed in sewage treatment plants, filters are not specifically designed to retain microplastic and terrestrial soils that have received sewage sludge do contain microplastic fibres²⁷. In the UK alone, over 11 km³ of water is discharged into inland waters, estuaries and the sea each year²¹ from treatment plants. Certain

sub-tidal marine sites may, however, contain large quantities of microplastic in their sediments because for nearly 30 years, a quarter of UK sewage sludge was dumped at 13 designated marine disposal-sites around the coast, until this practice was stopped in 1998 through The Urban Waste Water Treatment Regulations 1994^{21,22}. Since substantial quantities of sewage sludge and effluent are discarded to the sea, there is considerable potential for microplastic to accumulate in aquatic habitats, especially in densely populated countries.

To manage the environmental problems of microplastic it is important to understand and target the major pathways of microplastic into habitats with mitigation-measures. While sewage waste provides one potential route for entry of microplastics, others have been identified including; fragmentation of larger items, introduction of small particles that are used as abrasives in cleaning products and spillage of plastic powders and pellets. Forensic techniques that compare the size, shape, and type of polymers²⁸ may, provide useful insights into the sources of the microplastic. For instance, if the material originated from fragmentation, the frequency-distribution of sizes of plastic debris would be skewed to smaller irregular fragments from the major types of macroplastic (e.g. polyethylene, polystyrene, polypropylene) found in habitats⁷. If, however, scrubbers in cleaning products spheres were more important, we would expect most of the material to consist of fragments and spheres of polyethylene. These sources do not, however, account for the occurrence of microplastic fibres in sludge and effluent taken from sewage treatment works²⁶ and soil from terrestrial habitats where sewage sludge had been applied, the source of which are more likely explained by fibres shed from clothes/textiles during washing²⁷. Work is therefore needed to gather forensic information about the number, type of polymer and shape, to assess the likelihood of microplastic entering marine habitats through this possible pathway.

Here, we investigate the spatial extent of microplastic across the shores of six continents to examine whether spatial patterns relate to its sources or sinks. We test the following hypotheses that there will be more microplastic in habitats that accumulate smaller particles of sediment (hypothesis 1) and in areas

1 with larger population-densities of humans (hypothesis 2). Based on forensic analyses of the material
2 we then tested the hypotheses that sediment collected from sewage-disposal sites contains more
3 microplastic than reference sites (hypothesis 3), that microplastic found on the shoreline will resemble
4 microplastic found in sub-tidal sewage disposal sites, sewage-effluent discharged from treatment works
5 and wastewater from washing clothes using washing machines (hypothesis 4).
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13 MATERIALS AND METHODS

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15 **Global sampling of sediment from shores.** Samples of sediment were collected from sandy beaches in
16 Australia (Port Douglas; 16°29S, 145°28E; Busselton Beach 33°39S, 115°19E), Japan (Kyushu 32°24N,
17 131°39E), Oman, United Arab Emirates (Dubai 25°17N, 55°18E), Chile (Vina Del Mar 32°56S,
18 71°32W; Punta Arenas 53°08S, 70°53W), Philippines (Malapascua Island 01°18N, 01°103E), Portugal
19 (Faro 36°59N, 07°57W), Azores (Ponta Delgado 37°44N, 25°34W), USA (Virginia 36°56N, 76°14W;
20 36°57N, 76°14W; California 35°50N, 118°23W), South Africa (Western Cape 33°06S, 17°57E),
21 Mozambique (Pemba 19°01S, 36°01E) and the United Kingdom (Sennon Cove 50°04N, 05°41W) from
22 2004-2007. During collection (and in subsequent sections), cotton clothing was worn rather than
23 synthetic items (such as fleeces) to avoid contamination by plastic fibres. Samples were collected by
24 working down-wind to the particular part of the highest strandline deposited by the previous tide.
25 Sediment was sampled to a depth of 1 cm deep using established techniques⁷. As the sampling was
26 opportunistic, the sampling design was unable to remove possible confounding due to intrinsic
27 differences in the tidal range and position of the strandline that will vary spatially and temporally on the
28 shores. The extraction and identification of microplastic, including the analysis of sediment particle-
29 size was done using established methods^{1,7}. Microplastic debris was extracted from a 50 mL subsample
30 of sedimentary material using a filtered, saturated solution of sodium chloride to separate particles of
31 microplastic from sediments. This involved three sequential extractions using the saline solution and
32 identifying the microplastic using Transmittance FT- IR and a spectral database of synthetic polymers
33 (Brucker I26933 Synthetic fibres ATR-library).
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Marine sewage disposal and reference sites. In 2008 and 2009, samples of sediment ($n = 5$) were haphazardly collected from each reference (Plymouth 50°14N, 04°10W and Tyne 55°06N, 01°18W) and sewage-sludge disposal site (Plymouth 50°14N, 04°18W; Tyne 55°03N, 01°17W) using van Veen grabs deployed from a boat. The surface 5-10 cm of sediment of each sample was placed into pre-cleaned 500 mL aluminium foil containers and microplastic extracted as before. During collection, cotton clothing was worn rather than synthetic items to avoid contamination by plastic fibres.

Sewage effluent. Microplastic was extracted from effluent discharged ($n = 5$) by two sewage treatment plants. Pre-cleaned glass bottles (750 mL) with metal caps were used to collect effluent from discharges from Tertiary-level Sewage Treatment Plants at West Hornsby and Hornsby Heights (NSW, Australia) in 2010. Effluent was filtered and microplastic counted as before but without additional saline water and standardised to give the amount of microplastic per litre of effluent.

Washing machine effluent. Because the proportions of synthetic fibres found in marine sediments and sewage resembled those used for textiles, we counted the number of fibres discharged into wastewater from using domestic washing machines used to launder clothing. To estimate the number of fibres entering wastewater from washing clothes, 3 different front-loading washing machines (Bosch WAE24468GB, John Lewis JLWM1203 and Siemens Extra Lasse XL 1000) were used (40 °C, 600 R.P.M.) with and without cloth (polyester blankets, fleeces, shirts). Detergent and conditioner were not used because these blocked the filter-papers. Cross-contamination was minimised (<33 fibres) at the start of the experiment and in between washes, by running washing-machines at 90 °C, 600 R.P.M for 3 cycles without clothes. Effluent was filtered and microplastic counted^{1,7}.

RESULTS AND DISCUSSION

Eighteen shores across six continents were contaminated with microplastic (Fig. 1) and so we investigated whether spatial patterns relate to its sources or sinks. The abundance of microplastic per sample ranged from 2 (Australia) to 31 (Portugal, U.K.) fibres per 250 mL of sediment (Fig. 2A), consisting of polyester (56 %), acrylic (23 %), polypropylene (7 %), polyethylene (6 %) and polyamide fibres (3 %). There was more microplastic in densely-populated areas²⁴ with a significant relationship between its abundance and human population-density (Linear Regression, $F_{1,16} = 8.36$, $P < 0.05$, $n = 18$, $r^2 = 0.34$; Fig. 2B), but no clear relationship with the mean-size of natural particulates (Spearman Rank $\rho = 0.39$, $n = 18$, $P > 0.05$). As a consequence we explored the importance of sewage-disposal as a source of microplastic to marine habitats (Fig. 2C). Despite sewage not being added for more than a decade, disposal-sites still contained >250 % more microplastic than reference sites (2 Factor ANOVA, $F_{1,16} = 4.50$, $n = 5$, $P < 0.05$), mainly fibres of polyester (78 %) and acrylic (22 %). To further examine the role of sewage as a source, microplastic was extracted from effluent discharged by sewage treatment plants and compared with sediments from disposal-site. Effluents contained, on average, one particle of microplastic per litre. As expected, polyester (67 %) and acrylic (17 %) fibres dominated, including polyamide (16 %), showing proportions of polyester and acrylic fibres in sewage-effluent resembled microplastic contaminating sediments from shores and disposal-sites. This suggests these microplastic fibres were mainly derived from sewage via washing-clothes^{26,27}, rather than fragmentation^{1,4,5,7,13-15,18,23} or cleaning-products^{2,7,11,14,16,23-25}. Because proportions of polyester fibres found in marine sediments and sewage resembled those used for textiles (78 % polyester, 9 % polyamide, 7 % polypropylene, 5 % acrylic)²⁹, we counted the number of fibres discharged into wastewater from using washing blankets, fleeces and shirts (all polyester). Here we show a garment can shed >1900 fibres per wash. All garments released >100 fibres per litre of effluent, with >180 % more from fleeces (Fig. 2E), demonstrating that using washing machines may, indirectly, add considerable numbers of microplastic fibres to marine habitats. Because people wear more clothes during the winter than in the summer³⁰

1 and washing machine usage in households is 700 % greater in the winter ³¹, we would expect more
2 fibres to enter sewage treatment during the winter. Research is therefore needed to assess seasonal
3 changes in the abundance of plastic fibres in sewage effluent and sludge. In our study it was not
4 possible to use detergent and conditioners because they blocked the filter-papers and prevented us from
5 filtering the samples of effluent, so work is needed to investigate the effect of detergent and conditioner
6 on the quantities of fibres in effluent.
7

8 Our work provides new insights into the sources, sinks and pathway of microplastic into habitats. We
9 show polyester, acrylic, polypropylene, polyethylene and polyamide fibres contaminate shores on a
10 global-scale, with more in densely populated areas and habitats that received sewage. Work is now
11 needed to establish the generality of the relationship with population-density at smaller spatial scales,
12 including freshwater and terrestrial habitats where sewage is also discharged. One source of these fibres
13 of microplastic appears to be the disposal of sewage contaminated with fibres from washing clothes
14 because these textiles contain >170 % more synthetic than natural fibres²⁹ (e.g. cotton, wool, silk). The
15 quantity of microplastic in sewage and natural habitats is, however, likely to be much greater. Brightly
16 coloured fibres are easily distinguished from natural particulates, but microplastic from cleaning
17 products and fragmentation will be discoloured by biofilms and resemble natural particulates, so better
18 methods are required. In the future microplastic contamination is likely to increase as populations of
19 humans are predicted to double in the next 40 years and further concentrate in large coastal cities¹⁷ that
20 will discharge larger volumes of sewage into marine habitats. To tackle this problem, designers of
21 clothing and washing machines should consider the need to reduce the release of fibres into wastewater
22 and research is needed to develop methods for removing microplastic from sewage. One means of
23 mitigation may be ultrafiltration because fewer fibers have been found downstream from a sewage
24 treatment plant that use this process as opposed to one that did not²⁶. Work is urgently needed to
25 determine if microplastic can transfer from the environment and accumulate in food-webs through
26 ingestion. In humans, inhaled microplastic fibres are taken up by the lung tissues and can become
27

1 associated with tumours³², whilst dispersive dyes from polyester and acrylic fibres have been shown to
2 cause dermatitis³³. Research is therefore needed to determine if ingested fibres are taken up by the
3 tissues of the gut and release monomers (e.g. ethylene glycol, dimethyl terephthalate, propenenitrile,
4 acrylonitrile, acrylonitrile, vinyl chloride, vinylidene chloride, vinyl bromide), dispersive dyes,
5 mordants (e.g. aluminium, chromium, copper, potassium, tin)³⁴, plasticisers from manufacture and
6 sorbed contaminants from sewage (e.g. organotin³⁵, nonylphenol³⁶ and Triclosan³⁷. The bioavailability
7 of these chemicals is likely to be greater from fibres of polyester and acrylic, compared to the more
8 hydrophobic microplastics (e.g. polyethylene, polypropylene) that have more heterogenic atoms. In
9 conclusion, our study shows the importance of testing hypothesis to improve our understanding about
10 the sources and sinks of microplastic in habitats. Such experimental approaches are vital if we are to
11 target the pathways of microplastic into habitats with effective mitigation-measures that reduce
12 contamination by microplastic.
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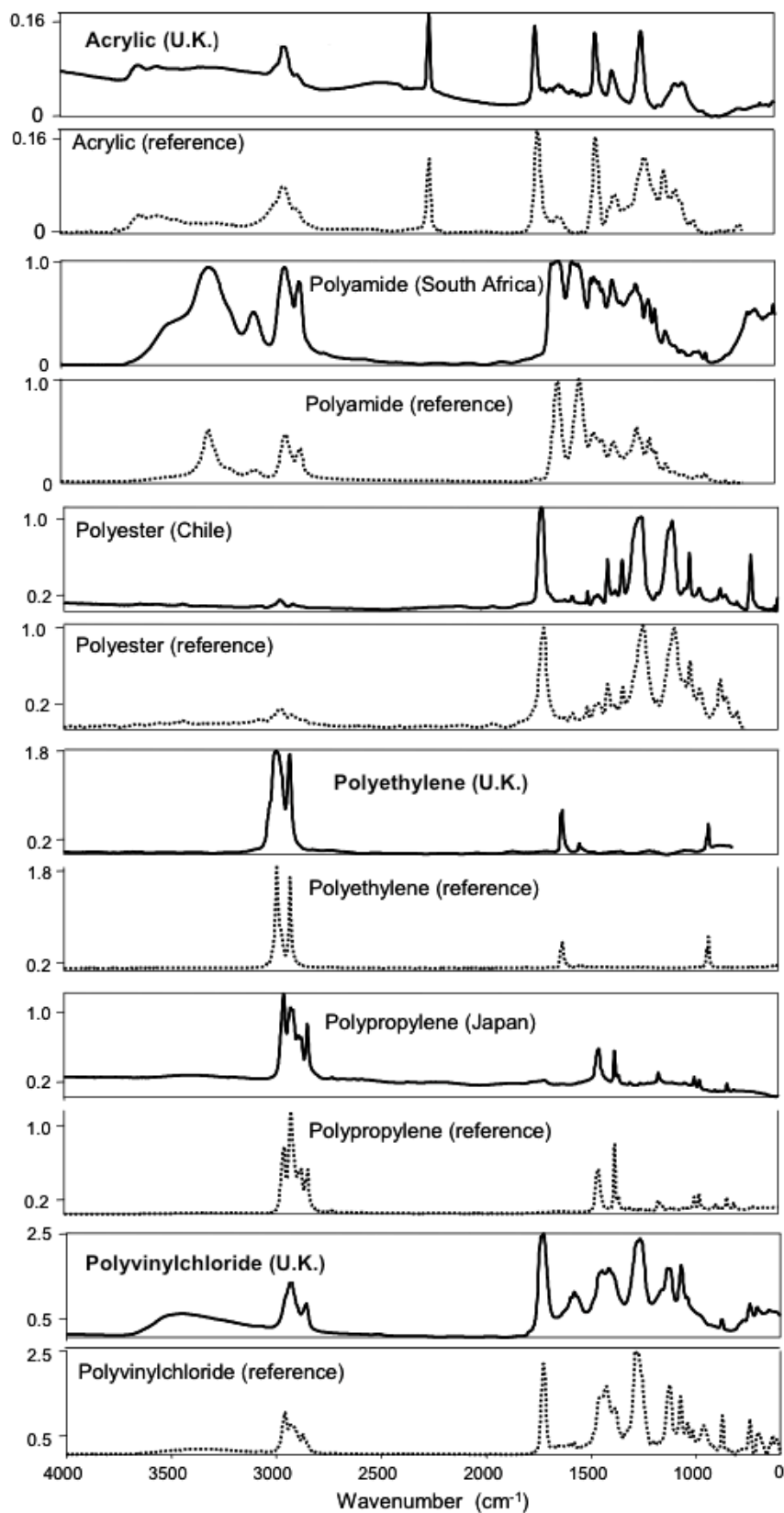


Figure 1. Examples of Fourier Transform Infrared spectra of microplastic and corresponding reference material from ATR spectral database, vertical axis represents transmission in standard optical density units.

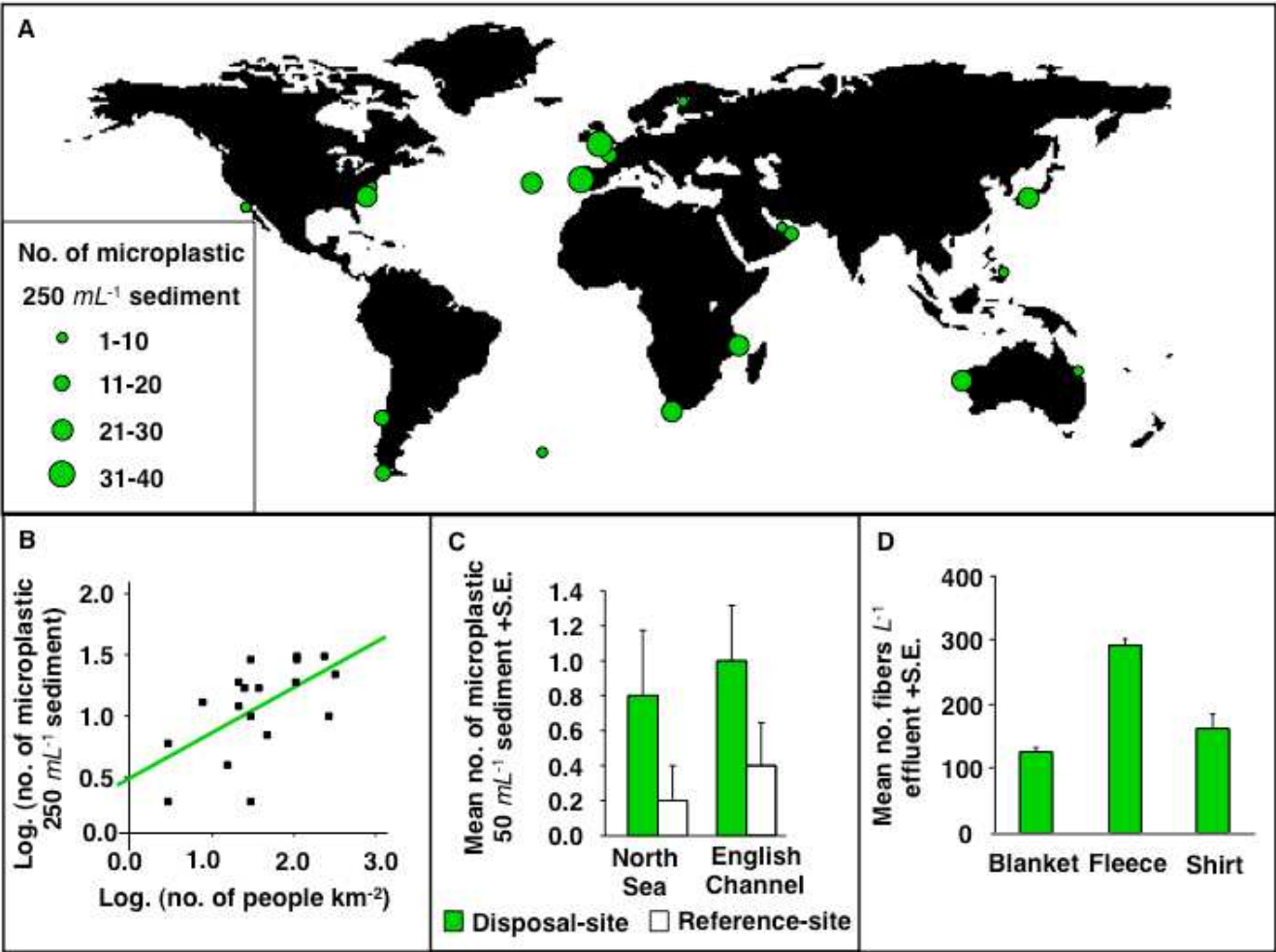


Figure 2. (A) Global extent of microplastic in sediments from 18 sandy shores and identified as plastic by Fourier Transform Infrared Spectrometry. The size of filled-circles represents number of microplastic particles found. (B) Relationship between population-density and number of microplastic particles in sediment from sandy beaches. (C) Number of particles of microplastic in sediments from sewage disposal-sites and reference-sites at two locations in U.K. (D) Number of polystester fibres discharged into wastewater from using washing-machines with blankets, fleeces and shirts (all polyester).

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