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Accumulation of Microplastic on Shorelines Woldwide: 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 miocroplastics and the mean size-distribution of natural particulates. An important source of microplastic appears to be through sewage contaminated by fibers from washing clothes. Forensic evaluation of microplastic from sediments showed that the proportions of polyester and acrylic fibers 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 fibers per wash. This suggests that a large proportion of microplastic fibers 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.

INTRODUCTION

We use >240 million tonnes of plastic each year[1] and discarded 'end-of-life' plastic accumulates, particularly in marine habitats,[1] where contamination stretches from shorelines[2] to the open-ocean[3−5] and deep-sea.[6] Degradation into smaller pieces means particles <1 mm (defined here as microplastic[2,7,8]) are accumulating in habitats,[1] outnumbering larger debris.[7] 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 health.[9−16] Over the last 50 years the global population-density of humans has increased 250% from 19 to 48 individuals per square km,[17] during this time the abundance of micrometer-sized fragments of acrylic, polyethylene, polypropylene, polyamide, and polyester have increased in surface waters of the northeast Atlantic Ocean.[1] This debris now contaminates sandy, estuarine, and subtidal habitats in the United Kingdom,[1,6] Singapore,[18] and India.[19] 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 Commision,[14] European Union,[15] Royal Society,[3] 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 debris[20] and receive millions of tonnes of sewage each year[21] which has also been shown to contain microplastic.[22−26] 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

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microplastic fibers.[27] In the UK alone, over 11 km^3 of water is discharged into inland waters, estuaries, and the sea each year[21] from treatment plants. Certain subtidal marine sites may, however, contain large quanitites 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 polymers[28] 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 irrgeular fragments from the major types of macroplastic (e.g., polyethylene, polystyrene, polypropylene) found in habitats.[7] If, however, scrubbers in cleaning products 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 fibers in sludge and effluent taken from sewage treatment works[26] and soil from terrestrial habitats where sewage sludge had been applied, the source of which is more likely explained by fibers shed from clothes/textiles during washing.[27] 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 with larger population-densities of humans (hypothesis 2). Based on forensic analyses of the material we then tested the hypotheses that sediment collected from sewage-disposal sites contains more microplastic than reference sites (hypothesis 3), that microplastic found on the shoreline will resemble microplastic found in subtidal sewage disposal sites, sewage-effluent discharged from treatment works, and wastewater from washing clothes using washing machines (hypothesis 4).

MATERIALS AND METHODS

Global Sampling of Sediment from Shores. Samples of sediment were collected from sandy beaches in Australia (Port Douglas; 16°29S, 145°28E; Busselton Beach 33°39S, 115°19E), Japan (Kyushu 32°24N, 131°39E), Oman, United Arab Emirates (Dubai 25°17N, 55°18E), Chile (Vina Del Mar 32°56S, 71°32W; Punta Arenas 53°08S, 70°53W), Philippines (Malapascua Island 01°18N, 01°103E), Portugal (Faro 36°59N, 07°57W), Azores (Ponta Delgado 37°44N, 25°34W), USA (Virginia 36°56N, 76°14W; 36°57N, 76°14W; California 35°50N, 118°23W), South Africa (Western Cape 33°06S, 17°57E), Mozambique (Pemba 19°01S, 36°01E), and the United Kingdom (Sennon Cove 50°04N, 05°41W) from 2004 to 2007. During collection (and in subsequent sections), cotton clothing was worn rather than synthetic items (such as fleeces) to avoid contamination by plastic fibers. Samples were collected by working down-wind to the particular part of the highest strandline deposited by the previous tide. Sediment was sampled to a depth of 1 cm deep using established techniques.[7] As the sampling was opportunistic, the sampling design was unable to remove possible confounding due to intrinsic dfferences in the tidal range and position of the strandline that will vary spatially and temporally on the shores. The extraction and identification of microplastic, including the analysis of sediment particle-size, was done using established methods.[1,7] Microplastic debris was extracted from a 50 mL subsample of sedimentary material using a filtered, saturated solution of sodium chloride to separate particles of microplastic from sediments. This involved three sequential extractions using the saline solution and identifying the microplastic using Transmittance FT- IR and a spectral database of synthetic polymers (Bruker I26933 Synthetic fibres ATR-library).

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 precleaned 500 mL aluminum foil containers and microplastic extracted as before. During collection, cotton clothing was worn rather than synthetic items to avoid contamination by plastic fibers.

Sewage Effluent. Microplastic was extracted from effluent discharged (n = 5) by two sewage treatment plants. Precleaned 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 standardized to give the amount of microlastic per liter of effluent.

Washing Machine Effluent. Because the proportions of synthetic fibers found in marine sediments and sewage resembled those used for textiles, we counted the number of fibers discharged into wastewater from using domestic washing machines used to launder clothing. To estimate the number of fibers 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 minimized (<33 fibers) 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 DISCUSSON

Eighteen shores across six continents were contaminated with microplastic (Figure 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.) fibers per 250 mL of sediment (Figure 2A), consisting of

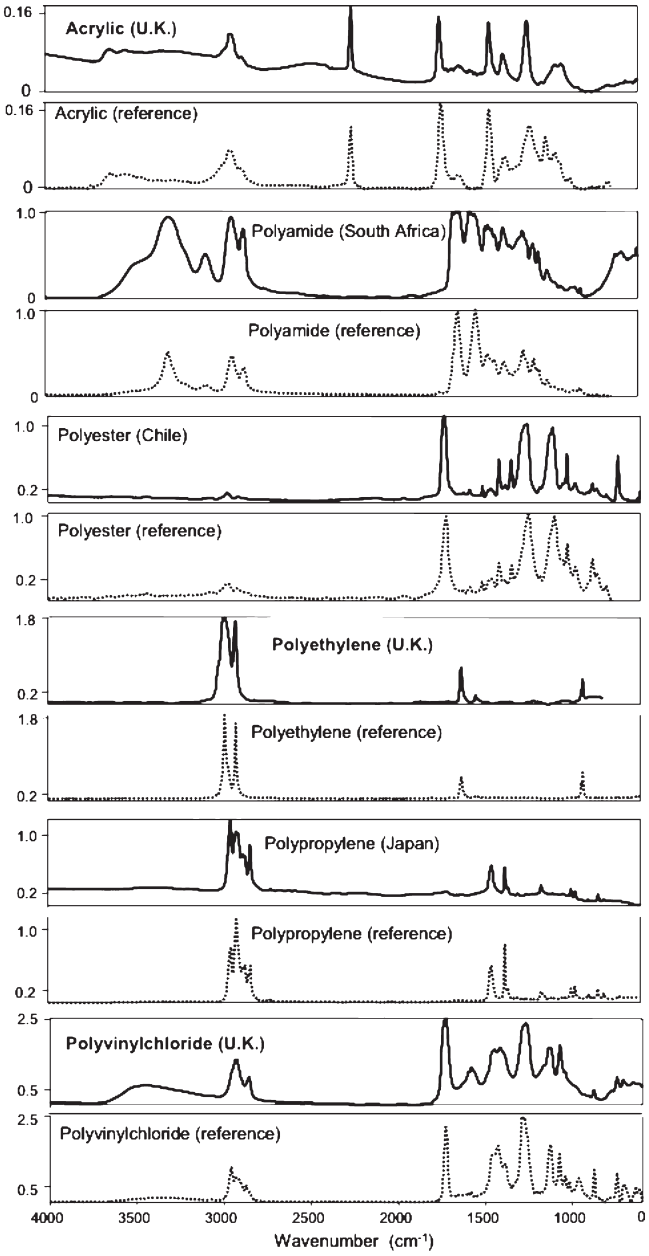

Figure 1. Examples of Fourier transform infrared spectra of microplastic and corresponding reference material from ATR spectral database, vertical axis represents transmission in standard optical density units. (Bruker Optics ATR-Polymer Library - a Collection of Synthetic Fibres, Copyright 2004 Bruker Optic GmbH).

polyester (56%), acrylic (23%), polypropylene (7%), polyethylene (6%), and polyamide fibers (3%). There was more microplastic in densely populated areas[24] with a significant relationship between its abundance and human population-density (Linear Regression, $F_{1,16} = 8.36$, $P < 0.05$, $n = 18$, $r^2 = 0.34$; Figure 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 (Figure 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 fibers 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 liter. As expected, polyester (67%) and acrylic (17%) fibers dominated, including polyamide (16%), showing proportions of polyester and acrylic fibers in sewage-effluent resembled microplastic contaminating sediments from shores and disposal-sites. This suggests these microplastic fibers 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 fibers 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 fibers discharged into wastewater from using washing blankets, fleeces, and shirts (all polyester). Here we show a garment can shed >1900 fibers per wash. All garments released >100 fibers per liter of effluent, with > 180% more from fleeces (Figure 2E), demonstrating that using washing machines may, indirectly, add considerable numbers of microplastic fibers to marine habitats. Because people wear more clothes during the winter than in the summer[30] and washing machine usage in households is 700% greater in the winter,[31] we would expect more fibers to enter sewage treatment during the winter. Research is therefore needed to assess seasonal changes in the abundance of plastic fibers in sewage effluent and sludge. In our study it was not possible to use detergent and conditioners because they blocked the filter-papers and prevented us from fitering the samples of effluent, so work is needed to investigate the effect of detergent and conditioner on the quantities of fibers in effluent.

Our work provides new insights into the sources, sinks, and pathway of microplastic into habitats. We show polyester, acrylic, polypropylene, polyethylene, and polyamide fibers contaminate shores on a global-scale, with more in densely populated areas and habitats that received sewage. Work is now needed to establish the generality of the relationship with population-density at smaller spatial scales, including freshwater and terrestrial habitats where sewage is also discharged. One source of these fibers of microplastic appears to be the disposal of sewage contaminated with fibers from washing clothes because these textiles contain >170% more synthetic than natural fibers[29] (e.g., cotton, wool, silk). The quantity of microplastic in sewage and natural habitats is, however, likely to be much greater. Brightly coloured fibers are easily distinguished from natural particulates, but microplastic from cleaning products and fragmentation will be discoloured by biofilms and resemble natural particulates, so better methods are required. In the future microplastic contamination is likely to increase as populations of humans are predicted to double in the next 40 years and further concentrate in large coastal cities[17] that will discharge larger volumes of sewage into marine habitats. To tackle this problem, designers of clothing and washing machines should consider the need to reduce the release of fibers into wastewater and research is needed to develop methods for removing microplastic from sewage. One means of mitigation may be ultrafiltration because fewer fibers have been found downstream from a sewage treatment plant that use this process as opposed to one that did not.[26] Work is urgently needed to determine if microplastic can transfer from the environment and accumulate in food-webs through ingestion. In humans, inhaled microplastic fibers are taken up by the lung tissues and can become associated with tumors,[32] while

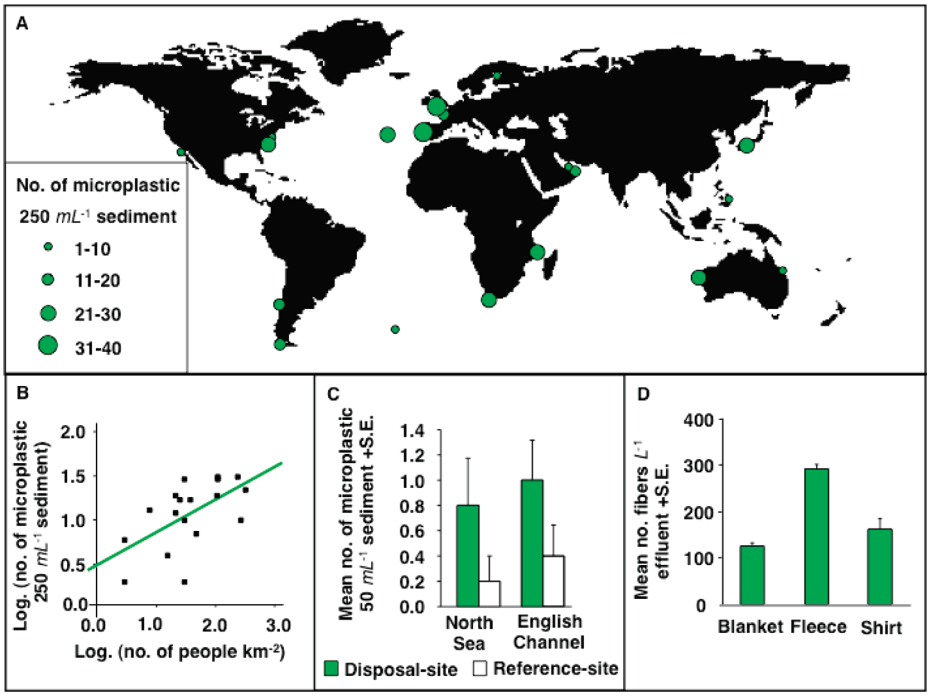

Figure 2. (A) Global extent of microplastic in sediments from 18 sandy shores and identified as plastic by Fourier transform infrared spectrometry. The size of filled-circles represents number of microplastic particles found. (B) Relationship between population-density and number of microplastic particles 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 fibers discharged into wastewater from using washing-machines with blankets, fleeces, and shirts (all polyester).

dispersive dyes from polyester and acrylic fibers have been shown to cause dermatitis.[33] Research is therefore needed to determine if ingested fibers are taken up by the tissues of the gut and release monomers (e.g., ethylene glycol, dimethyl terephthalate, propenenitrile, acrylonitrile, acrylonitrile, vinyl chloride, vinylidene chloride, vinyl bromide), dispersive dyes, mordants (e.g., aluminum, chromium, copper, potassium, tin),[34] plasticisers from manufacture and sorbed contaminants from sewage (e.g., organotin,[35] nonylphenol,[36] and Triclosan.[37] The bioavailability of these chemicals is likely to be greater from fibers of polyester and acrylic, compared to the more hydrophobic microplastics (e.g., polyethylene, polypropylene) that have more heterogenic atoms. In conclusion, our study shows the importance of testing hypothesis to improve our understanding about the sources and sinks of microplastic in habitats. Such experimental approaches are vital if we are to target the pathways of microplastic into habitats with effective mitigation-measures that reduce contamination by microplastic.

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