Spatial Patterns of Plastic Debris along Estuarine Shorelines

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The human population generates vast quantities of waste material. Macro (>1 mm) and microscopic (<1 mm) fragments of plastic debris represent a substantial contamination problem. Here, we test hypotheses about the influence of wind and depositional regime on spatial patterns of micro- and macro-plastic debris within the Tamar Estuary, UK. Debris was identified to the type of polymer using Fourier-transform infrared spectroscopy (FT-IR) and categorized according to density. In terms of abundance, microplastic accounted for 65% of debris recorded and mainly comprised polyvinylchloride, polyester, and polyamide. Generally, there were greater quantities of plastic at downwind sites. For macroplastic, there were clear patterns of distribution for less dense items, while for microplastic debris, clear patterns were for denser material. Small particles of sediment and plastic are both likely to settle slowly from the water-column and are likely to be transported by the flow of water and be deposited in areas where the movements of water are slower. There was, however, no relationship between the abundance of microplastic and the proportion of clay in sediments from the strandline. These results illustrate how FT-IR spectroscopy can be used to identify the different types of plastic and in this case was used to indicate spatial patterns, demonstrating habitats that are downwind acting as potential sinks for the accumulation of debris.

Introduction

Plastics have brought many societal benefits over the last 50 years and global production is now greater than 260 million tonnes per year (*1*). The scale of production coupled with the great durability of plastic and poor rates of recycling has resulted in the accumulation of plastic debris in the environment from the poles to the equator, and from the depths of the oceans to the tops of mountains (*2*). Plastic debris enters marine habitats from a variety of sources, including littering, discarded fishing gear, illegal-dumping, and discharges from rivers, storm drains, and sewage outfalls (*3*). Particles of macroplastic (>1 mm) can be transported thousands of kilometres and can contaminate relatively remote locations (*4*). The large-scale dispersal of marine debris provides a vector for the transport of sorbed chemicals that are potentially toxic (*5*), nonindigenous organisms (*6*), and potentially harmful micro-organisms that colonise the surface of the plastic (*7*). Plastic debris has a range of shapes and sizes from tiny fragments, micrometres in length (*8*), to larger items, including hulls of boats and fishing nets many meters long. When this material fragments into smaller pieces, the potential for ingestion and accumulation within the tissues of animals increases (*9*). Yet a detailed quantitative description of the relative abundance of microplastic debris (<1 mm) compared to larger items remains to be achieved. Particles of plastic, <2.8 mm in diameter, reportedly make up the majority of the debris in the Pacific Ocean (*10, 11*). Certain types of plastic are expected to occur in greater abundances than others due to the relative proportions that are manufactured, used, and discarded. For instance, half of all the plastics that are produced annually are polyolefins (i.e., polyethylene, polypropylene) (*1*) which are principally used to make packaging that is used once and then discarded. It is, however, not known whether polyolefins occur in greater abundance as items of debris compared to other polymers.

Plastic debris is known to accumulate along strandlines (*12*), in the open ocean (*13*), and on the seafloor (*14, 15*). Individually, many studies have suggested that physical factors such as the wind (*16*–*18*), wave-action (*12*), and the density of plastic (*19, 20*) have a role in influencing spatial patterns of accumulation. Research has also suggested that the accumulation of plastic debris is affected by climatic forcing, geostrophic winds caused by gradients of atmospheric pressure and solar radiation, stratospheric temperature, and the Coriolis effect (*21, 22*). While wind-driven water currents are important in the transport of macroplastic debris (*23*). In coastal areas, movements of water caused by the wind (*24*), waves, and tides transport particles of sediment according to their size, shape, and density (*25*). It is, however, unknown whether these properties also influence the transportation and spatial patterns of plastic debris.

The transport of particles, whether sand or plastic, is a function of their size, shape and density, and these properties largely determine the minimum velocity of water that is required for their transport. The density of plastic can vary considerably depending upon the type of polymer and the manufacturing process. For instance, the density of polyethylene ranges from 0.90 to 0.99 g cm^{-3}, whereas polystyrene can vary from around 1.00 g cm^{-3} ("high-impact polystyrene") to <0.05 g cm^{-3} in expanded-polystyrene. The size and density of plastic debris is likely to determine its vertical position within the water-column, and also influence the extent to which it is transported. Spatial patterns of plastic debris are thought to vary at very small spatial scales (i.e., meters), making experimental design critical (*26*). Furthermore, no study has collectively examined the relative importance of wind, depositional environment, density, and size of debris on spatial patterns of distribution in the environment.

Estuaries provide a tractable model system to study the influence of wind and depositional regime on the distribution of plastic debris. In relatively linear estuaries, there is typically a prevailing direction of the wind and a relatively predictable longitudinal gradient in the depositional regime associated

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with gradients of shear stress caused by the waves, and the tide. These factors structure the distribution of sediments of estuaries by suspending, transporting, and depositing particles according to their size and density. Small particles of sediment settle slowly from the water-column and as such are readily transported by the flow of water, and are deposited in areas where the movements of water is slower (*27*). Consequently, the proportion of fine particulates such as clay within sediments can be used as an index of the depositional environment.

We tested hypotheses relating to the influence of wind and depositional regime on the spatial patterns of micro (<1 mm) and macroplastic (>1 mm) debris in the Tamar Estuary (UK). Transmittance Fourier transform infrared spectroscopy (FT-IR) was used to confirm the identity of the polymers that were present as debris. Specific hypotheses tested were that (1) microplastic debris will be found in greater numerical abundance than macroplastic debris, (2) polyethylene and polypropylene will be more abundant than other polymers (due to differences in levels of production), (3) downwind sites that receive the onshore wind will have greater abundance of plastic debris in their strandlines compared to sites in up-wind areas and this pattern will be stronger for plastic that is less dense (i.e., most likely to be transported at the surface of the water), and (4) the abundance of microplastic debris will be positively correlated with the proportion of clay within sediments.

Materials and Methods

Experimental Design and Sampling. The Tamar Estuary is a partially mixed and moderately sheltered macrotidal estuary in the NE Atlantic. The role of waves in the estuary is small due to a 1.5 km long breakwater in the outer reaches of Plymouth Sound. The estuary is approximately 32 km long and has a mean tidal range of 3.5 m, with gradients relating to wind (www.metoffice.gov.uk) and deposition of smaller particles of sediment on its gently sloping beaches (Supporting Information (SI) Figure S1-Sn b, c). Along the shoreline are accumulations of plastic debris from diffuse sources (inputs from rivers and the sea) and direct sources (littering, runoff from roads, stormwater drains, and a sewage outfall) into the estuary. The City of Plymouth (population 240 K) is located near the mouth of the estuary. To examine spatial patterns of plastic debris, three locations were examined. These represented a gradient of deposition of fine sedimentary material northwards from the mouth of the estuary, with sites on east (E1−3) and west (W1−3) banks in relation to the prevailing southwesterly wind (SI Figure S1-Sn a, b). At each location, there were two replicate up-wind sites (on the bank from which the wind is blowing) located on the west bank of the estuary, and two replicated downwind sites that receive the prevailing wind on the east bank. Sites were selected at locations that had the same slope of beach, but varied in their depositional regimes, indicated by the amount of clay within sediments on the strandline (SI Figure S1-Sn). The strandlines sampled were the most recent line of debris, visible macroplastic debris was collected and the underlying 3 cm of sediment (500 mL) was placed into foil containers (pebbles were not removed) so smaller plastic debris could be extracted and the size of particles of sediment could be determined. Strandlines were mostly composed of pebbles (74.5 ± 18.0%), sand (24.0 ± 17.9%), and varied according their amounts of silt (1.2 ± 0.9%) and clay (0.4 ± 0.6%). Although a sewage outfall, near E2, and the City of Plymouth near E3 are likely to be sources of some plastic debris, our design was sufficiently replicated within the estuary to identify any consistent spatial patterns for debris associated with the prevailing wind and/or depositional regime. Macroplastic debris is known to vary at very small spatial scales (*26*), so at each location there were two replicate sites separated by 60 m. Each site consisted of a 50 m stretch of linear shoreline. Each accumulation of plastic debris were numbered and a list of random numbers used to place a five 0.25 m^2 quadrats to collect replicate samples of plastic debris from within each quadrat. It was important to avoid differences in the spatial patterns of plastic debris being confounded by the rate of settlement of different sizes/densities of plastic debris brought about by sampling at different tidal heights. Consequently, the height at which samples were taken across sites was standardized by collecting material deposited by the last high-tide (5.30 m, 10.23 a.m., 26 July 2005). Before analyzing the particle-size of sediment, microplastic debris was extracted from a 50 mL subsample of sedimentary material collected from each quadrat using the method of Thompson et al. (*9*). This method uses a filtered saturated solution of NaCl to separate less dense particles of microplastic from sediments. Micro- and macroplastic debris were identified using Transmittance FT-IR and a spectral database of synthetic polymers (Brüker I26933 Synthetic fibres ATR-library). For items that were conclusively identified as plastic, the abundance, mass, and maximum diameter was recorded (see SI for further information). To test whether factors that influence the transport of sediment could be used to explain the transport of plastic debris, the relationship between the abundance of plastic and the clay content within the strandline was examined (see SI for further information).

Statistical Analyses. The composition of plastic debris was examined in terms of size, source, and type of polymers. To examine the frequency of different sizes of plastic we standardized the number of microplastic and macroplastic particles per 500 mL of sediment. This was achieved by multiplying the number of particles of microplastic found in each 50 mL replicate sample by 10 and comparing this to the number of particles of macroplastic found in 500 mL of sediment. To determine the relative influence of size, density, wind, and depositional regime on spatial patterns of plastic accumulation within the estuary, a three-factor ANOVA was used to separately analyze micro and macroplastic debris. For microplastic debris, the results are expressed as number of particles <1 mm per 50 mL sediment, whereas for macroplastic the results are expressed as the number of particles >1 mm per 500 mL sediment. It was not necessary to standardize this data to a common volume because this would have affected variance and was not necessary to test hypothesis 3. The factor "wind" had two levels (up-wind and downwind), "depositional regime" had 3 levels (great, moderate, and small) and "site" had two levels. The factors "wind" and "depositional regime" were each treated as fixed and orthogonal, and the third factor "site" was treated as a random factor nested in "wind" and "depositional regime". Plastics identified by FT-IR were put into four groups according to their density. Expanded plastics were types of polymers <0.05 g cm^{-3} (e.g., expanded polystyrene), less dense plastic ranged from 0.90−0.99 g cm^{-3} (e.g., polythene and polypropylene), dense ranged from 1.00−1.20 g cm^{-3} (e.g., polyamide, unexpanded polystyrene) and very dense plastics were greater than 1.30 g cm^{-3} (e.g., polyterephthalate, polybutlyphthalate, polyvinylchloride). Spearman Rank correlation was used to examine the relationship between the amount of clay within sediments and the abundance of different densities of microplastic debris. A conservative value of alpha ($P < 0.01$) was chosen to adjust for multiple comparisons. Statistical analyses were carried out using GMAV (EICC, University of Sydney) and Microsoft Excel (see SI).

Results

Relative Abundance of Different Sizes and Types of Polymers. A total of 952 items of debris were recorded from the

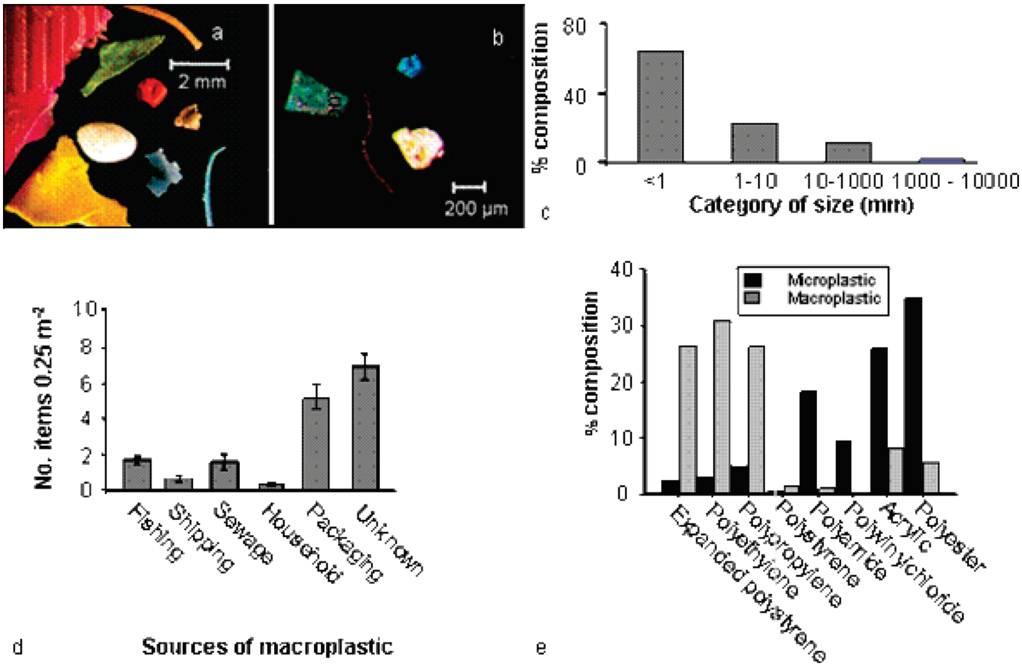

FIGURE 1. Identity and composition of plastic debris collected from the strandline of the Tamar Estuary (U.K.) according to the form and shape (a, b), size (c), source of macroplastic using standardized data see methods for details, (d) and type of polymers (e). Data for (d) is presented as mean ± SE.

30 samples of sediment and as predicted there was greater abundance of microplastic compared to macroplastic debris (Hypothesis 1) (Figure 1c). This was reflected in the frequency-distribution of different sizes of debris, which were skewed toward smaller debris. In terms of numerical abundance, microplastic accounted for 65% of the total amount found. Of the recognizable larger items, the main types of plastic, in terms of their use, were packaging (polystyrene foam 32% and wrappers 18%), others relating to sewage (the handles from buds of cotton wool 4% and sanitary towels 15%), fishing (line 6%), and shipping (rope 7%) (Figure 1d). Over 50% of the polymers found were polyolefins, and over 25% was expanded polystyrene. For particles of macroplastic, there were greater quantities of less dense plastics such as polytheylene (32%) and polypropylene (28%) compared to other types of plastic (Hypotheses 2), however polystyrene (23%) was also abundant (Figure 1e). In contrast to macroplastic debris, assemblages of smaller microplastic fragments were mainly composed of denser plastics such as polyvinylchloride (26%), polyester (35%), and polyamide (18%).

Influence of Wind and Depositional Regime on Spatial Patterns of Different Sizes and Densities of Plastic Debris. There was generally a greater abundance of particles of plastic in sites that were downwind (Hypotheses 3); but this pattern varied with size, density, depositional regime, and at the scale of "site". For microplastic debris, there was significantly more dense, very dense, and total microplastic debris at sites that were downwind; however, the significance of this pattern varied with sites and the depositional regime (Figure 2 and Table S1-Sn in SI). Particles of expanded microplastic were only present at sites that were downwind (east) and in areas that received greater deposition of fine sediment (i.e E3). Particles of less dense microplastic debris (e.g., polyethylene and polypropylene) were distributed evenly throughout the estuary, but were relatively scarce with no more than 1 particle per 50 mL sediment. The abundance of dense microdebris was over 3 times greater at sites that were downwind. Together, acrylic, polyester and polyvinylchloride made up nearly 80% of microplastics debris found. There was also more very dense and total microplastic debris at sites that were downwind. At these sites, there were up to eight fragments (in total) of microplastic per 50 mL sediment, compared to other sites that had less than three fragments per 50 mL sediment.

There was more macroplastic debris at downwind sites and again the distribution varied with the density of the plastic, depositional regime and at the scale of 'site' within locations (Figure 1 and SI Table S1-Sn). Less dense debris was more abundant at sites that were downwind, with greater deposition of fine sediment. Similarly there were consistently more particles of less dense macroplastic at sites that were downwind. The abundance of dense macroplastic, however, remained constant throughout the estuary, irrespective of the prevailing wind and the depositional regime. In contrast to patterns for microplastic debris, there were considerable quantities of very dense particles of macroplastic at one of the up-wind sites that had greater deposition of fine sediment. Similar patterns of accumulation of total macroplastic debris were observed at sites that were downwind, irrespective of the regime of sediment deposition. The composition of the total macroplastic debris was dominated by less dense (50%) and expanded (23%) items.

There was no relationship between the proportion of clay and the abundance of different densities of microplastic debris in the strandline (Spearman Rank, $P > 0.01$ ns) (Hypothesis 4).

Discussion

Relative Abundance of Different Sizes and Types of Polymers. Analysis by FT-IR revealed important information about the composition of debris, with fragments of microplastic debris accounting for the majority (65%), as predicted by Hypothesis 1. Similar proportions of tiny plastic debris (<2.8 mm) have been reported in the Pacific Ocean where they ranged from 43 to 96% of stranded plastic debris (*10, 11, 28*). Smaller particles of plastic in the range of nanometres (<1 μm) are also likely to be present; however, the smallest particles that can be indentified using FT-IR are around 2 μm (*8*). Nearly half of the particles of macroplastic were polyolefins and a further quarter was expanded polystyrene, while nearly 80% of the smaller microplastic

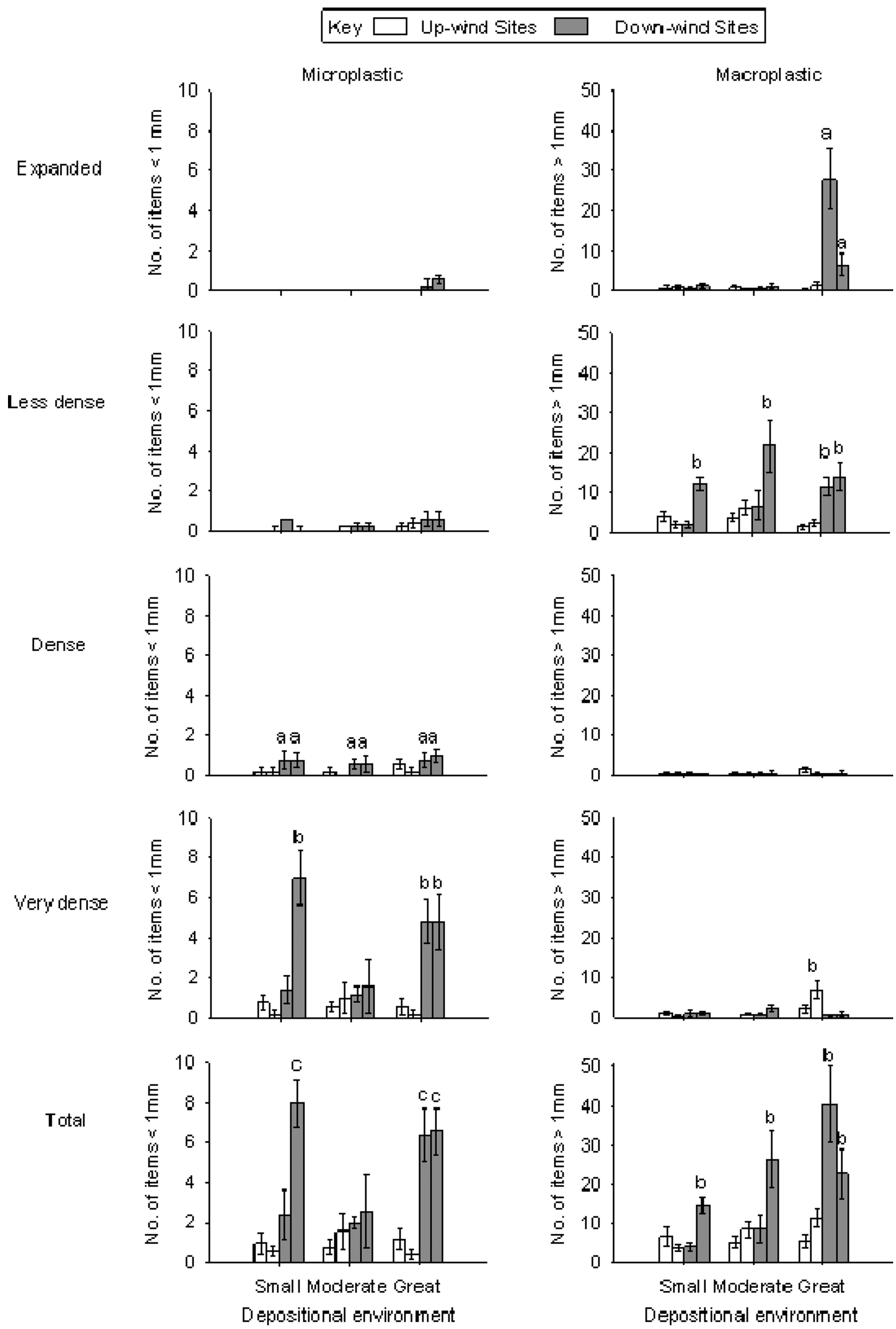

FIGURE 2. Spatial patterns of different densities of microplastic (per 50 mL sediment) and macroplastic debris (per 500 mL sediment) within the Tamar Estuary (U.K.) according to the prevailing wind and the depositional environment. Values expressed as means ± SE. Significantly different groupings from ANOVA and SNK tests are indicated by dissimilar letters ("a", "b", and "c"). Data has not been standardized see methods for details.

fragments were denser plastics. There are four potential explanations for this. First, denser plastics may take longer to degrade compared to less dense plastic. Second, denser plastics are likely to spend more time in contact with abrasive particles of sediment, while the lighter plastics collide less forcefully with particles of sediment and this causes more particles of microplastic compared to lighter plastics. Third, compared to polyolefins and expanded polystyrene, particles of polyester, polyvinylchloride, and polyamide may be more easily distinguished from natural grains of sediment in terms of shape and color than particles of polyolefin and expanded polystyrene. Fourthly, wind may transport less dense plastics higher up the shores and inland, therefore the densities we report in this paper may be conservative values.

It seems likely that a substantial quantity of the microplastic debris present had formed from the breakdown of larger items of clothing, packaging, and rope. Plastics are known to degrade by photolytic, biological, and mechanical processes (*29*−*31*). Consequently, plastic is likely to have greater rates of fragmentation in areas which experience strong wave-action and abrasion by particles of sediment. Although this may explain the greater abundance of microplastic debris at more exposed sites toward the mouth of the estuary it is also likely that less dense particles of microplastic would be expected to be more susceptible to transportation by wind, waves, and currents.

Another possible source of microplastic debris to the estuary is from discharges from the sewage treatment plant. Domestic laundering of clothes may act as a source of fibres of microplastic to treatment plants and more than four microplastic fibres per gram of sediment have been found in samples taken from sewage (*32, 33*).

As predicted by Hypothesis 2 there were greater abundances of polyethylene and polypropylene compared to other types of polymers. This is in agreement with previous studies of macroplastic debris in which packaging for food was the most abundant type of debris found in intertidal habitats (*15, 34*). It is not surprising because 8% of oil produced in the world is used to manufacture plastic and half of this are polyolefins, mainly for single-use packaging that is thrown away within a year (*1, 8, 35*).

Influence of Wind and Depositional Regime on Spatial Patterns of Plastic Debris. Categorizing the plastic debris found using FT-IR revealed that wind, size, and density of plastic debris are important factors influencing the spatial distribution of plastic debris in this estuary. The apparent wind-blown pattern varied with the size and density of the debris. For macroplastic, clear patterns were for sites that were downwind. These contained more expanded and less dense debris, whereas for smaller microplastic debris the clearest patterns were for denser material (e.g., polyvinylchloride).

Expanded particles of macroplastic (polystyrene) debris were found predominately at the first site that was downwind at E3 (SI Figure S1-Sna). Similarly, particles of less dense macroplastic (polyethylene and polypropylene) were most abundant at sites that were downwind, but with greater depositional regimes. Interestingly, denser plastic were less abundant and was distributed evenly throughout the estuary. There are a number of possible explanations for the greater proportion of expanded and less dense fragments of macroplastic on sites near the mouth of the estuary. One is that these sites are closer to densely populated areas where quantities of litter would be expected to be greatest. Also, less dense material floats at the surface of the water where it is transported, by currents created by the wind, from the wider ocean to the estuary. Indeed, similar patterns of transport have been reported in Hawaii and South Australia (*21, 23*). The importance of wind and the density of plastic as factors influencing transport have been suggested previously for debris in North America, with increasing numbers of fragments of expanded polystyrene higher on estuarine shores (*12*). It has been suggested that plastic debris in the open-ocean is vertically distributed within the water-column in relation to density and that the influence of wind on mixing decreases with depth (*36*). Consequently, the greater quantities of less dense pieces of macroplastic on the strandline may have been caused by them accumulating at the top of the water-column and being transported by wind-induced currents. The smaller quantities of denser macroplastic debris is probably because these are more likely to accumulate near the seabed where the speed and movement of water is slower. In the present study, only one estuary was studied and the data may be confounded by inherent differences between the east and west shores of the estuary, other than their orientation in relation to the prevailing wind. To help differentiate between these potential explanations, further research is needed over a greater spatial area, including multiple estuaries, in conjunction with studies that examine the transport of items of plastic between potential sources and sinks using satellite-tracked plastic debris.

Since the 1970s, manufacturers of plastic have been producing increasingly less dense items and in some cases they have reduced their density by a half. This trend has been driven by the need to reduce the consumption of fuel during transport and to increase the quantity of plastic items that can be made from a dwindling and increasingly expensive resource (*1, 35*). This practice may also result in plastics that are less durable and more likely to accumulate at sites that are downwind.

Relationship between the Abundance of Microplastic Debris and Clay within Sediments. The lack of any clear relationship between the proportion of clay within sediments and the abundance of different densities of microplastic is surprising since some of these synthetic particles are similar in size and density to fine particles of sediments. It is unlikely therefore that interactions between the plastic and the sediment particles caused differences in the retention of plastic debris between shores with differing sediment characteristics. Fine-grained sediments are, however, cohesive and regularly flocculate with organic material; hence, flocculation may contribute to movements of microplastic in the water-column. The proportions of clay that we report here are very small (<1%) and so other influences on clay content rather than the hydrodynamic regime may be important in the Tamar Estuary. Although the Tamar Estuary is comparable in terms of proportions of sand (6−46%), silt (0.5−2%), and clay (0.1−0.9%) to numerous other estuaries in Britain and France, open coasts are more exposed with a greater proportion of sand (45−100%), smaller proportions of silt (0−0.2%) and virtually no clay (*37*). Because the average clay content of the sediments within the Tamar is smaller, the chance of finding a significant relationship with the abundance of microplastic is small. Consequently, it is important to test the relationship between the abundance of microplastic debris and the size of particles of sediment from strandlines of beaches from around the world. This could indicate the extent to which more populated regions have more microplastic debris. Previous research has shown that over 90% of the variation in the abundance of plastic debris can be explained by the population density of the country where the samples were taken (*38*).

In this study, microplastic debris also varied in shape, from irregular fractual (e.g., rough, fragmented, or geometric), to spherical, to long-thin fibres (Figure 1d,e). A range of indices (e.g., disk-rod, rod, flatness, oblate-prolate, and sphercity) have been used to quantify the shape of particles of sediment (*25*), and these indices could also be used to quantify the shape of plastic debris. Laboratory trials have shown that fractual shaped aggregates of polystyrene (100−1000 μm) settle more than 8 times faster than equivalent-sized spherical particles (*39*). The majority of fragments of plastic were fibres, it is thus important to examine the influence of the diameter, length, and density on the settling-velocity of microplastic fibres in seawater.

A range of organisms with differing feeding strategies, including mussels, barnacles, amphipods, and lugworms are capable of ingesting microplastic debris (*8, 40*). Recent work has shown that ingested particles of microplastic can accumulate in the gut of mussels and can translocate into their circulatory-system where they can persist for over 48 days. This work also showed that smaller particles had a greater potential to accumulate in the tissues (*9*). Considering that fragments of microplastic accounted for the majority of the debris, research is needed to assess the quantity of microplastic in the tissues of wild and farmed marine animals.

Our study suggests that most plastic is fragmented and that the main types of polymers present in macroplastic debris are not representative of the mixture of polymers found as microplastic debris. Our approach provides useful information about the levels of contamination by different plastics and we call for research to unravel the toxicological effects, and rates of degradation of the most abundant types of plastic debris.

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Supporting Information Available

Seven additional pages including an additional figure and table. This material is available free of charge via the Internet at http://pubs.acs.org.

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