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Microplastics in coastal urban sediments: Discrepancies in concentration and character revealed by different approaches to sample processing

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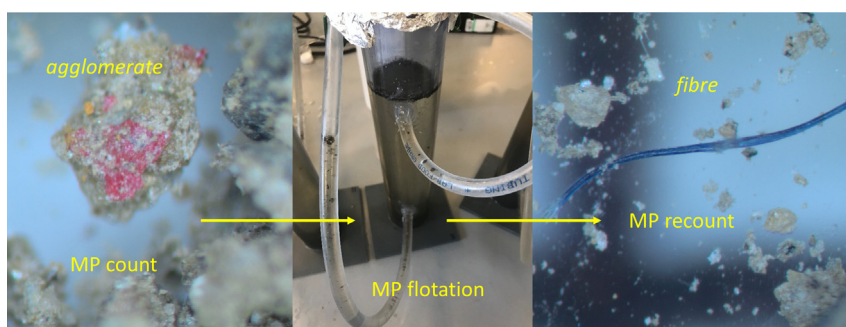
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HIGHLIGHTS

- Coastal, intertidal microplastics (MPs) counted directly and after flotation in ZnCl_2 solution
- MP concentrations greater when analysed directly
- Discrepancies attributed to aggregation-agglomeration and presence of MPs too dense to float
- Concentrations of MPs in coastal sediments may often be underestimated

GRAPHICAL ABSTRACT



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ABSTRACT

Intertidal, silty sediment samples have been collected from three coastal locations with different uses and anthropogenic signatures in the vicinity of Plymouth, southwest England, and analysed for microplastics (MPs) by two independent means. Firstly, MPs were counted and characterised directly on unprocessed dried sediment under a stereo microscope, and secondly MPs were isolated from sediment by flotation in ZnCl_2 solution and filtration before analysis. Direct counting resulted in average (\pm one standard deviation) numbers of MPs per g of dry sediment of 0.77 ± 0.16 at a marina-harbour, 0.58 ± 0.30 under a busy road bridge and 0.79 ± 0.43 adjacent to country parkland. After flotation and filtration, concentrations were reduced to 0.24 ± 0.11 , 0.18 ± 0.06 and $0.48 \pm 0.38 \text{ MP g}^{-1}$, respectively. Observations were attributed to hetero-aggregation of small fibres with settling sediment during flotation, and the presence of MPs (including paints) that were too dense to float or that had aggregated or agglomerated with denser sediment and construction material in situ. The findings have implications for the efficacy of flotation procedures, accurate estimations of MP concentrations in sediment and the representativeness of MPs by type, and inter-site comparisons of MPs that are widely reported in the literature.

1. Introduction

Microplastics (MPs) have received increasing attention as contaminants of aquatic environments over the past two decades (Mato et al., 2001; Rios et al., 2007; Costa et al., 2010; Andrady, 2011; Cole et al., 2015; Lusher et al., 2015; Ross et al., 2021). The precise location, impacts and fate of MPs will depend on factors like the composition, density and size of the

particles and the hydrodynamics and geomorphology of the environment. However, a major receptor and secondary source in most aquatic systems is bed and shore sediment (van Cauwenberghe et al., 2013; Horton et al., 2017; Massos and Turner, 2017; Cincinelli et al., 2021).

Most methods used to retrieve MPs from sediment for subsequent quantification and characterisation have relied on flotation. Here, the sample is mixed with a saline solution of known density, and after a period of settlement buoyant material (including MPs) is removed with the supernatant and isolated by filtration (Claessens et al., 2011; Woodall et al., 2014; Pohl et al., 2020). This approach may also be preceded or succeeded by

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sample digestion (or purification) in, for example, hydrogen peroxide solution, to reduce the amount of natural organic matter present (Peng et al., 2017; Tiwari et al., 2019; Xu et al., 2020).

The efficacy of this method depends on a number of factors, like the ratio of sediment mass to salt solution volume, settling time, particle size, particle-container interactions and settling column shape and internal surface area (Klein et al., 2015; Wang et al., 2018; Yu et al., 2018), but the main driver is the density of the solution relative to the densities of the MPs in the sample. Accordingly, a saturated solution of NaCl of density $\sim 1.2 \text{ g cm}^{-3}$ should, in theory, isolate low-density MPs constructed of polyolefins, polystyrene and nylon, but a solution of greater density, like concentrated or saturated NaI or ZnCl_2 (density > 1.4 to 1.8 g cm^{-3}) would be required to isolate MPs constructed of denser polymers like polyvinyl chloride and polyethylene terephthalate. This is borne out in practice, with solutions of increasing density exhibiting increasing recoveries of distinctive or customised MPs of different compositions added to sediment samples (Klein et al., 2015; Quinn et al., 2017).

The lack of saline solution standardisation hampers comparisons in MP abundance and characteristics between environments, with a reduction in solution density predicted to be associated with an increasing underestimation of MP concentration and a reduction in polymer diversity. More generally, however, there is evidence that flotation in saline solutions can provide a significant underestimate of the MP inventory in sediment relative to direct visual (microscopic) inspection of dried, but otherwise unprocessed sample. For example, Martin et al. (2017) found a recovery rate of MPs from spiked continental shelf sediments of 54 % by flotation in a solution of sodium polytungstate ($\text{Na}_6\text{H}_2\text{W}_{12}\text{O}_{40}$; density = 1.65 g cm^{-3}) compared with 89 % by direct microscopic analysis. Reasons for the discrepancies in MP recovery were not explored but images shown suggest that they could be related to the evasion of flotation by fouling or sediment capture. Horton et al. (2017) adopted three successive phases of large ($> 1 \text{ mm}$) MP identification in four sediments from the River Thames catchment: rapid, visual inspection of whole samples; inspection of material floated out by ZnCl_2 ; inspection of settled residues. In the most polluted sample, 34 % of MPs remained in the post-flotation residual sediment that were relatively dense and complex in nature.

In the present study, MPs are considered in fine, intertidal sediments from three different coastal locations in Plymouth, southwest England, with different uses and anthropogenic signatures. Specifically, MPs are identified and characterised under a microscope by two sequential approaches: firstly, direct analysis of dried and unprocessed sediment, and secondly, analysis of material isolated from sediment after flotation and filtration. The aims are to determine the efficacy of flotation as a means of MP capture more generally, and to investigate the origins and characteristics of MPs that evade flotation.

2. Materials and methods

2.1. Study sites

Three locations around the city of Plymouth were selected for sampling intertidal silt-fine sand: namely, Coxside, Tamar and Saltram (Fig. 1).

Coxside is an urbanised, semi-enclosed marina adjacent to a fishing harbour within 1 km of the city centre. Here, the sediment from a west-facing beach bordered by a slipway and a housing estate was sampled. The Tamar Estuary lies to the west of Plymouth and sampling took place on mudflats on the east bank about 5.5 km to the northwest of the city centre. Specifically, samples were taken at a location immediately to the south of a main road bridge that spans the estuary, with a small boat yard a few hundred m further south and a municipal wastewater treatment plant 2 km to the north. Saltram is a country park on the east bank of the upper, and non-navigable, Plym Estuary, around 4 km to the northeast of the city centre. Sampling here took place on mudflats adjacent to the park boundary and about 1 km downestuary of a municipal wastewater treatment plant.

2.2. Sampling and sample processing

Sampling was undertaken at low tide and during December 2021. At each location, a perpendicular transect was measured from the water line and at distances of 5 m, 7.5 m and 10 m a small trench was dug with a metal spade. Avoiding any large stones, shells, pieces of debris and macroalgae, about 100 g of sediment was retrieved from three depths (0–5 cm, 5–10 cm and 10–15 cm) using a stainless steel spatula. Samples were individually wrapped in aluminium foil and in the laboratory were transferred to a series of aluminium trays, covered with new foil and dried in an oven at $30 \text{ }^\circ\text{C}$ for 24 h. Dried samples were stored in their trays and in a dark, wood-laminate cupboard pending analysis.

2.3. Microplastic identification

In the laboratory, operators wore cotton laboratory coats and nitrile gloves at all times. Sample processing took place in glassware, ceramicware and plastic-ware that had been pre-cleaned with acid and distilled water and that was covered, where possible, with watch glasses or aluminium foil.

Between about 20 and 40 g of each dried sample was accurately weighed into a series of glass petri dishes and any visible aggregates were broken down with the aid of a stainless steel spatula. Microplastics were subsequently identified, sized and counted under a Nikon SMZ800 stereo microscope with a $1\times$ Achrom objective at up to 63 X magnification (and with images captured by an Olympus SC30 digital camera) using the approach and criteria outlined in MERI (2019). Thus, a 90-mm diameter, 1-cm gridded template was constructed and placed under the petri dish to be examined. With the aid of a 2-cm stainless steel probe (with an 8-cm wooden handle), the sample was inspected in each grid from top to bottom in a zig-zag fashion, with material moved diagonally upwards and in the direction opposite to the viewing track in order to avoid double counting. MP identification was based on: the response to the metal probe (plastics should not break under stress but should flex or bend); lack of organic structures evident, except for biofouling; homogeneity of width (for plastic fibres); homogeneity in colour (except for possible partial bleaching of plastic fibres). We also looked for paint particles, herein classified as plastics because of their polymeric composition. Identification criteria for paints included layering, angular shape and brittleness, or adherence to a substrate as a film (Turner, 2021).

As a control, a replicate sediment sample in a petri dish was left open in the laboratory for 24 h before being examined under the microscope as above. A total of 27 excess fibres were observed, suggesting that sample contamination during processing and while uncovered is about 1 fibre h^{-1} , or for a period of 45 min under the microscope, about 0.8 fibres.

2.4. Density separation

After identifying MPs above, samples were subject to separation by density using concentrated solutions of zinc chloride (measured density = 1.60 g cm^{-3}) that had been prepared by the addition of $\geq 98 \%$ anhydrous ZnCl_2 (Fisher Scientific) to distilled water and subsequently filtered through acid-hardened ashless cellulose filters (Whatman grade 541, 22 μm pore size). Thus, each dried sample was washed into a 300 mL (25 cm high, 4 cm internal diameter) PVC column with ZnCl_2 solution before the column was made up to 250 mL. The contents were stirred with a glass rod and allowed to settle for 24 h before the top $\sim 50 \text{ mL}$ of solution was carefully siphoned into a glass beaker through silicone tubing connected to an outlet port located about 10 cm below the column rim. The remaining headspace in the separating column was then rinsed with more ZnCl_2 solution and after about 30 min a further $\sim 50 \text{ mL}$ of was decanted. Rinsing-decanting was repeated before the contents of the beakers were vacuum-filtered through a series of Whatman 541 filters using a ceramic Buchner funnel. Filters were subsequently oven-dried on watch glasses at $35 \text{ }^\circ\text{C}$ for 24 h before MPs were identified, sized and counted on each filter under the microscope as above.



Fig. 1. Sampling locations for intertidal sediments in the vicinity of Plymouth, southwest England.

A series of controls performed without the addition of fractionated sediment revealed a maximum of two MP fibres per sample derived from external sources (e.g., contamination of the ZnCl_2 solution or separating column, or fallout during microscopic analysis).

2.5. Infrared analysis of fibres

A selection of suspected plastic particles ($n = 48$; and including 42 fibres) from different locations and of different colours and sizes (but $>30 \mu\text{m}$) that had been identified before or after density separation were analysed by Fourier transform infrared (FTIR) spectrometry. Here, particles were carefully placed on gridded slides with the aid of stainless steel tweezers or a wetted 000 sable hair paint brush before being transferred to a 2-

mm diameter Specac DC2 diamond compression cell and analysed using a Bruker Vertex70 $\mu\text{-FTIR}$ spectrometer in transmission mode coupled to a Hyperion 1000 microscope. Spectra, acquired with Bruker Opus 7.5 software from 32 scans within the range of 4000 cm^{-1} to 600 cm^{-1} , were compared with various spectral databases of synthetic polymers and fibres, with positive identification corresponding to a hit rate of $>60\%$. Particles analysed before density separation were returned to samples before flotation in ZnCl_2 solution.

2.6. Statistics

Statistical analyses were performed in Minitab v19. Because Anderson-Darling tests revealed a combination of (mainly) normal and (where there

were occasional outliers) non-normally distributed datasets ($n = 3$ or 9), the mean was used as a measure of central tendency and parametric tests (one-way or two-way ANOVA and paired t -tests) were employed for the determination of differences with an α value for significance of 0.05.

3. Results

3.1. Visual characteristics of microplastics

A total of 566 particles suspected of being MPs were identified from direct microscopic analysis of the sediment samples contained in petri dishes. MPs were categorised as fibres ($n = 408$; with a length to diameter ratio > 5), combined ($n = 137$; intertwined or aggregated fibres or plastic, including paint, bonded to an inorganic substrate), and other ($n = 21$; films, fragments, pellets and paint flakes), with examples illustrated in Fig. 2. MPs had a maximum size (length) of about 4 mm, with a detection limit, depending on colour and shape and the amount of sediment and debris in the field of view, ranging from about 25 to 45 μm .

After isolation by suspension in concentrated ZnCl_2 solution, a total of 194 suspected MPs were identified under the microscope, of which 145 were fibres, five were combined and 44 were other types. Here, filter paper backgrounds were much cleaner and MPs were largely free of other debris, making identification more straightforward and less labour-intensive. However, the size range of MPs detected was similar to that reported for particles identified directly.

3.2. Microplastic abundance and distribution

Concentrations of MPs on a number basis and normalised to the dry mass of sediment are summarised in Fig. 3, with data for the transect at each location grouped by depth. Regarding samples analysed directly after drying (Fig. 3a), mean values are greatest at 5–10 cm and lowest at 10–15 cm, but differences within and between locations were not significant according to either one-way ANOVA (assuming that location-depth constitutes a single factor) or two-way ANOVA (assuming that location and depth are different factors) with Tukey's pairwise comparisons as post hoc tests. For samples analysed after flotation in ZnCl_2 solution (Fig. 3b), ANOVA returned no significant differences between mean values

across the different locations and depth categories. In all cases, mean concentrations are lower than corresponding values determined directly, with a series of paired t -tests revealing statistically significant differences between these measures in six cases.

Fig. 4 shows the percentage distribution of MPs by particle type (fibres, combined, other) after pooling all data at each location from the different depths (and because particle type did not exhibit any obvious change with depth). For samples analysed directly (Fig. 4a), the greatest contribution arises from fibres at all locations ($\geq 50\%$), with other types contributing least overall but combined types absent from Saltram. After separation in ZnCl_2 (Fig. 4b), fibres still constitute the dominant type of MP but other types now make a greater contribution at each site than combined forms. That is, there is a clear shift in the distribution of MPs after their isolation using a concentrated saline solution.

3.3. Polymeric makeup of microplastics

FTIR analysis of 48 samples suspected of being MPs returned 41 positive matches with reference spectra, and examples are illustrated in Fig. 5. Specifically, and before density separation ($n = 24$), fibres were constructed of rayon ($n = 9$), polyester ($n = 5$), nylon ($n = 3$) and cellulose acetate ($n = 2$), with polyethylene ($n = 3$), polyvinyl chloride ($n = 1$) and acrylic ($n = 1$) present amongst non-fibrous samples. After density separation, fibres ($n = 17$) were constructed of rayon ($n = 10$), polyester ($n = 4$), polypropylene ($n = 2$) and cellulose acetate ($n = 1$). Overall, there was a roughly equal split between petroleum-based thermoplastics, like polypropylene, nylon and polyester, and semi-synthetic, regenerated cellulosic fibres (mainly rayon). While the latter are not strictly plastics, they are often reported in the MP literature (Woodall et al., 2014; Comnea-Stancu et al., 2017) and are classified as such in the present study.

4. Discussion

The MPs identified in the present sediments (silt and fine sand) likely have a variety of local and more distant sources, having been brought to the intertidal zone via ocean and riverine currents, sewage effluent, road runoff and atmospheric deposition. Qualitatively and quantitatively, the results are broadly similar to those reported previously in coastal

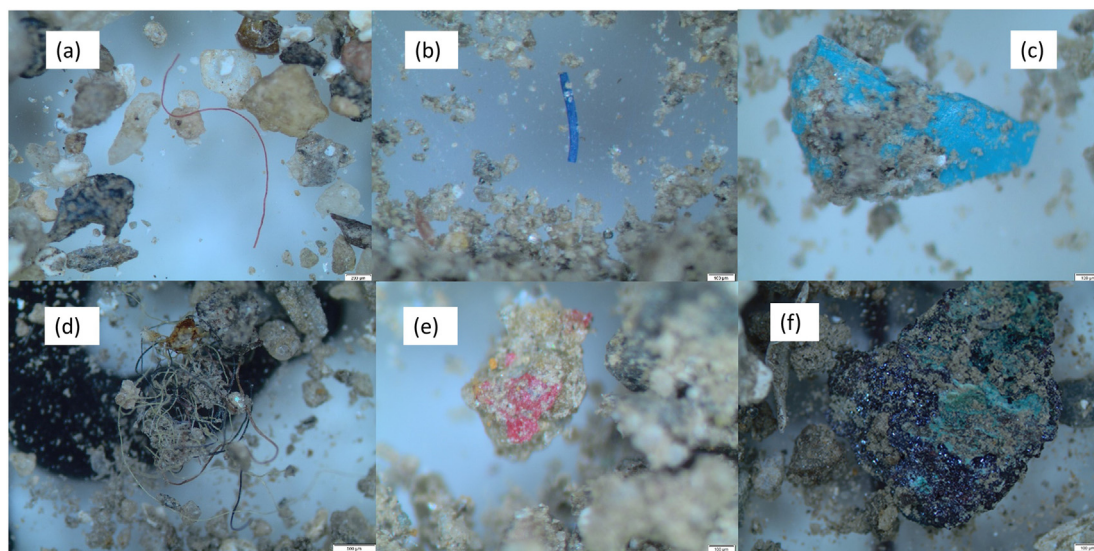


Fig. 2. A selection of MPs in dried sediment samples observed directly under the microscope, with scale bars (where annotated) ranging from 100 to 500 μm . (a) A red fibre of about 2 mm in length and 15 μm in diameter; (b) a blue fibre of about 500 μm in length and 50 μm in diameter; (c) a blue plastic fragment of about 1 mm across its longest dimension and about 50 μm thick and partly and loosely covered with sediment grains; (d) an aggregate of tangled fibres of different colours (and spanning 2–3 mm across) that has entrapped sediment and various other microdebris; (e) a thin layer of red plastic bonded to an inorganic substrate of mm-dimensions, and (f) a thin but irregular layer of green plastic bonded to an inorganic substrate of mm-dimensions.

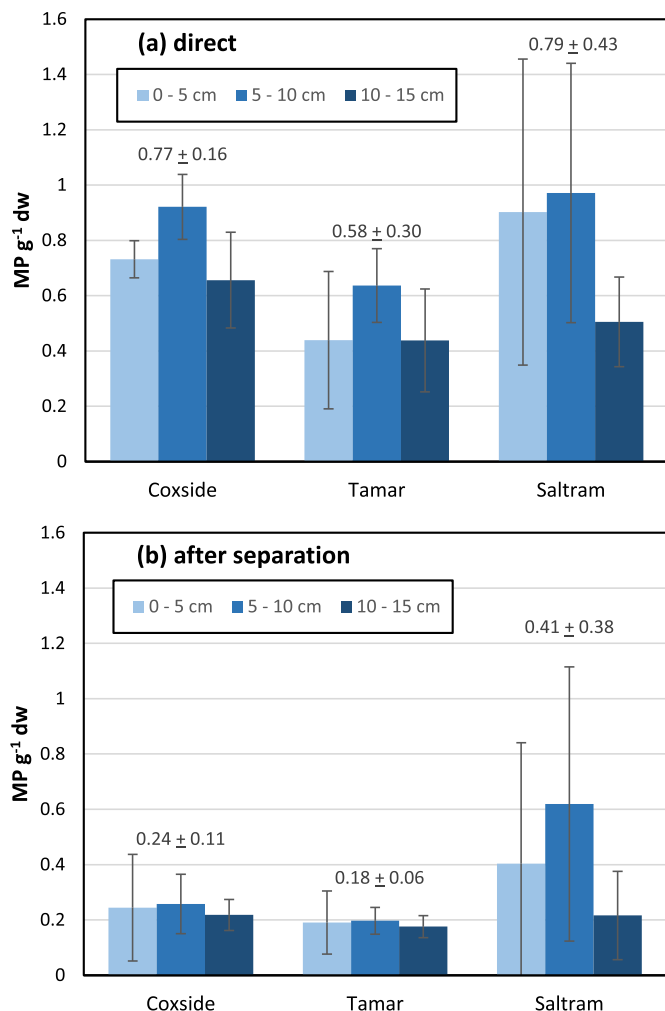


Fig. 3. Mean and one standard deviation ($n = 3$) of MP concentrations in sediment on a dry weight (dw) basis for the different depths sampled at Coxside, Tamar and Saltram (see Fig. 1) and that were analysed (a) directly and (b) after sample separation in $ZnCl_2$. Annotated are overall means and standard deviations for each location ($n = 9$).

environments impacted by various anthropogenic activities (including harbours) and where MPs have been isolated by flotation (and with or without pre-digestion of organic matter in H_2O_2). That is, concentrations are spatially heterogeneous but when normalised to dry mass of sediment dry mass are around 0.1 to 1 MP g^{-1} dw, with fibres a significant or the dominant type of MP (Claessens et al., 2011; Akhbarizadeh et al., 2017; Peng et al., 2017; Tiwari et al., 2019; Firdaus et al., 2020; Xu et al., 2020; Liu et al., 2022; Rasta et al., 2022).

While the present study did not identify any differences in MP concentration or characteristics with sediment depth, perhaps the most significant finding was a persistent reduction in MP abundance and shift in the distribution of MP type after sample settlement-flotation in a solution of concentrated $ZnCl_2$. Specifically, and according to mean values in Fig. 3, flotation returned between 31 % and 52 % of the number of MPs identified visually without separation, with a reduction in the percentage of combined MPs and an increase in the percentage of other types. The latter appeared to be dominated by small, neutral-coloured fragments and transparent, polyethylene films that presumably evaded ready, direct detection amongst sediment grains.

An increase in identification of all plastic shapes following flotation may have been expected as MPs become cleaner and less obscured by sediment and there is added scope (time and processing) for contamination. However, a reduction in overall abundance following flotation has been noted

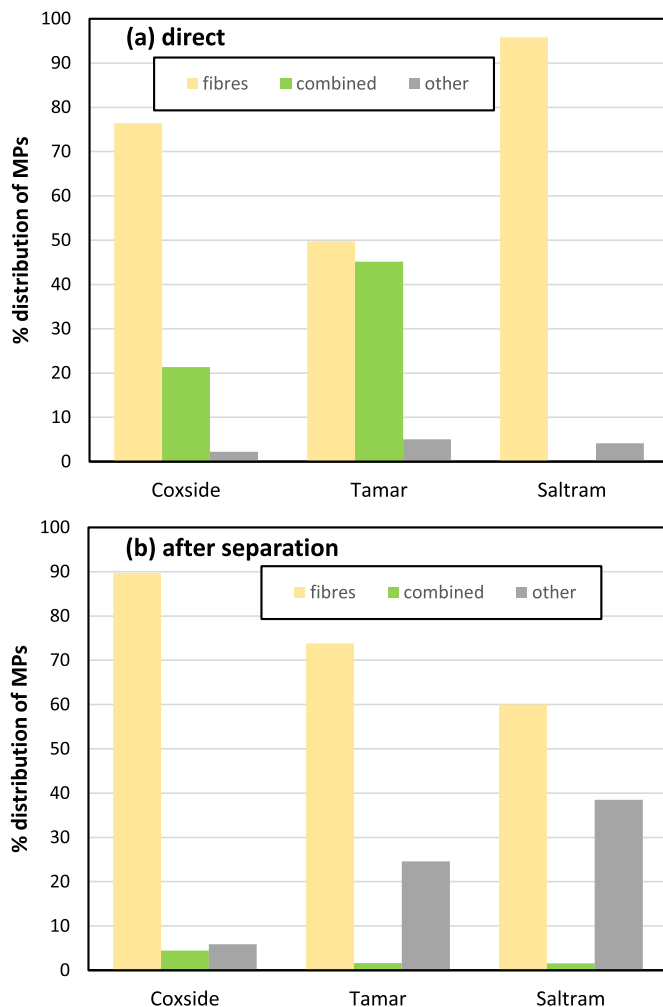


Fig. 4. Percentage MP distribution amongst different particle types (fibres, combined and other) for all sediment samples pooled at Coxside, Tamar and Saltram (see Fig. 1) and that were analysed (a) directly and (b) after sample separation in $ZnCl_2$ ($n = 9$ in each case).

previously in experiments in which continental shelf sediments had been spiked with customised MPs (Martin et al., 2017). Part of the discrepancy may be attributed to the adherence of MPs on the container surfaces (Klein et al., 2015; Coppock et al., 2017), but potentially more important are the presence of MPs, and the pre-existence and formation of plastic-non-plastic associations, whose net densities exceed that of the flotation solution (in the present case about $1.6 g cm^{-3}$) (Horton et al., 2017; Li et al., 2019).

Although, in theory, MPs constructed of the most common polymers should be isolated in $ZnCl_2$ solution, the presence of relatively dense functional additives may impede the flotation of some plastics (Turner and Filella, 2020). Marine and urban paint particles containing a range of fillers and additives may make an important contribution to this category in some environments (Turner, 2021). More generally, however, and evident microscopically as a combined type (see above), MPs may be aggregated or agglomerated with denser extraneous particulate matter, including sediment and construction material. Here, hetero-aggregation between small, buoyant MP fibres and settling sediment may take place during flotation because of the reduction in electrostatic repulsion effected at high ionic strength (Li et al., 2019). However, evidence in Fig. 2 and provided by Horton et al. (2017) suggests that many MPs are associated with denser material in situ. Specifically, aggregates of intertwined fibres and silt that are resistant to disaggregation during sample processing and settlement (Fig. 2d), and plastics that are bonded to or agglomerated with a substrate of, for example,

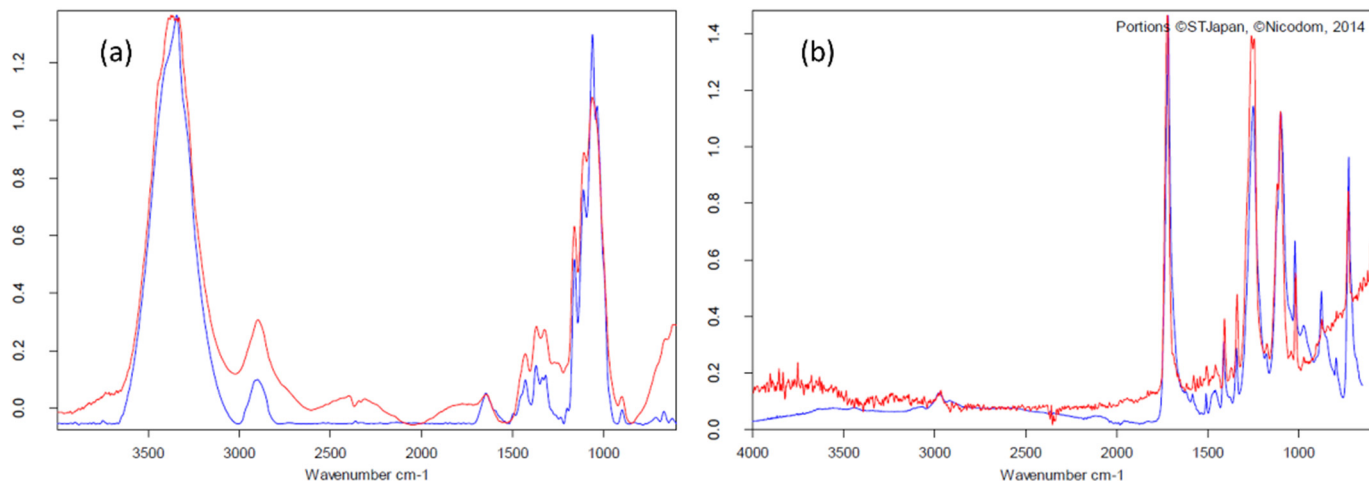


Fig. 5. FTIR spectra for two fibres (in red). Matches were obtained for (a) rayon viscose and (b) polyester, whose reference spectra are shown in blue. Note the peak at 3300 cm^{-1} (O–H stretching) in (a) that is relatively broad (characteristic of man-made fibres) rather than distinctive (characteristic of natural fibres).

bitumen or concrete (Fig. 2f). The latter are effectively microscopic forms of plastiglomerates and plasticrusts – agglutinates of plastic and inorganic material or layers of plastic on a substrate that have been fused together by burning (Corcoran et al., 2014; De la Torre et al., 2022). In many ways, they are also similar to agglomerates of tyre wear particles and road surface particles formed during frictional abrasion at the road surface (Jung and Choi, 2022). In this study, both aggregates and plastiglomerates (or combined MPs) were more important in sediments from Coxside and the Tamar than at Saltram. Consistent with assertions made by Horton et al. (2017) for the River Thames, this suggests that boating, urban or industrial practices (and, potentially, road abrasion) are instrumental to their formation.

The effects of aggregation-agglomeration on net density (ρ) can be demonstrated quantitatively by considering the densities of a range of polymer types (ρ_p) and the density of an aluminosilicate- or concrete-based extraneous phase or substrate (ρ_s) of 2.5 g cm^{-3} :

$$\rho = f_p \rho_p + f_s \rho_s \tag{1}$$

and, after rearranging:

$$\rho = \rho_p + f_s (\rho_s - \rho_p) \tag{2}$$

where f_p and f_s are the fractional contributions by volume from the polymer and extraneous phase or substrate, respectively. Fig. 6 shows ρ calculated for two polymers identified in the present study that have different densities and buoyancies in seawater (polypropylene, $\rho_p = 0.9 \text{ g cm}^{-3}$; polyester, $\rho_p = 1.3 \text{ g cm}^{-3}$) when increasing contributions are made from the extraneous phase or substrate. For polypropylene, a volume contribution of above about 40 % is required for the aggregate or agglomerate to evade capture by concentrated ZnCl_2 solution and for polyester the contribution may be as little as 20 %. For a saturated solution of NaCl, an alternative often used in the marine MP literature (Claessens et al., 2011; Woodall et al., 2014; Yu et al., 2018), polyester itself is not subject to flotation while polypropylene may evade capture with a volume contribution from an extraneous phase-substrate above about 20 %.

The common practice of pre-digesting sediment samples in H_2O_2 in order to destroy organic matter may loosen some aggregates, but this is unlikely to impact on hetero-aggregation or plastics that have pre-bonded to an inorganic phase. There has been increasing documentation of plasticrusts and plastiglomerates in the recent literature (Gestoso et al., 2019; Corcoran and Jazvac, 2020; De la Torre et al., 2022) but, to our knowledge, the present study is only the second (after Horton et al., 2017) to demonstrate this type of association at a microscopic (MP) scale.

With conventional methods, by design, unable to isolate this type of MP, it is not yet possible to comment on its wider abundance, distribution or impacts.

More generally, the findings of the present study have implications for the accurate estimations of MP concentrations and inventories in aquatic, silty sediments, as well as the representativeness of the MP stock in terms of type, density and association. Clearly, quantitative comparisons of MP concentrations reported in the literature must factor in seemingly small but critical differences in methodology.

5. Conclusions

This study has shown that both the abundance and shape distribution of MPs in coastal, silty sediments can be significantly impacted by the means of sample processing. The direct identification of MPs in dried sediments results in a higher concentration of plastics and a higher proportion of relatively dense particles than MP identification after sample flotation in ZnCl_2 solution. These findings have wider implications for the quantification of MPs by number and type in sediments and soils, and suggest that many microscopic paint particles and plastics held in assemblages with mineral material or bound to inorganic (e.g., construction) substrates may evade isolation by conventional methods.

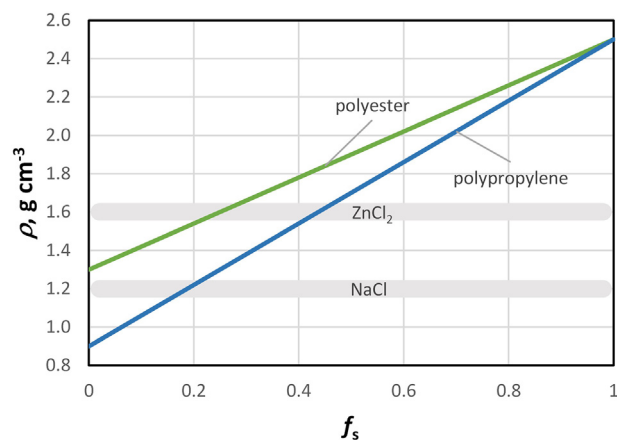


Fig. 6. Calculated net density, ρ , as a function of increasing the fractional volume, f_s , of an inorganic substrate or extraneous material ($\rho_s = 2.5 \text{ g cm}^{-3}$) in an aggregate or agglomerate containing polypropylene or polyester. NaCl and ZnCl_2 show the approximate ranges of solutions of saturated sodium chloride and concentrated zinc chloride, respectively.

CRedit authorship contribution statement

AT: conceptualization; investigation; formal analysis; writing – original draft; writing – review and editing; project management.

JK: methodology; formal analysis; writing – original draft.

Data availability

Data will be made available on request.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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