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Spatio-temporal trends in microplastic presence in the sediments of the River Thames catchment (UK)

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ABSTRACT

This study investigated the spatio-temporal variability of microplastics (MPs) in the sediments of the River Thames (UK) catchment over 30 months (July 2019 – Dec 2021). The average MP concentration was 61 items kg^{-1} d.w., with fragments <1 mm being dominant and polyethylene (PE) the most common polymer. Adjacent land use influenced MP concentrations and types, with industrial sites showing particularly high levels and a prevalence of small beads and industrial polymers. MP concentrations generally decreased after higher winter flows, likely due to sediment rearrangement or winnowing. This study describes the seasonal concentrations and characteristics of MPs present in sediment from the River Thames catchment, and attempts to identify their likely origin. Further, the study provides new insights into the mobility and fate of MPs in riverine settings under varying flow conditions, which is vital given the predicted increases in flooding under various global heating scenarios.

1. Introduction

Microplastics (MPs; plastics between 1 μm - 5 mm) have been detected in all environments, from the deep ocean (Kane et al., 2020) to the Arctic sea ice (Bergmann et al., 2019; Kanhai et al., 2020). This includes freshwater systems, with substantial levels of MPs found in even the most remote locations, such as the lakes of Tibet and river catchments in the French Pyrenees or Northern Botswana (Zhang et al., 2016; Jiang et al., 2019; Allen et al., 2019; Kelleher et al., 2023). In particular, river sediments form important MP reservoirs; Scherer et al. (2020) found MP concentrations in sediments to be 600,000 times higher than those found in the overlying water column and Eo et al. (2019) found sedimentary MP concentrations 2827 times higher compared to the water column. With evidence suggesting that high-flow events are able to remobilise legacy MPs (Hurley et al., 2018; Ockelford et al., 2020), the elevated levels of MP pollution in riverbeds raises concerns over the potential risks these contaminants might pose to aquatic biota (de Sá et al., 2018; Gray and Weinstein, 2017; Sulaiman et al., 2023) and to human populations that rely on rivers for drinking water abstraction. Although the weight fraction of MPs in plastic waste is small, they can interact with a wide range of species due to their size (Fossi et al., 2014) and emerging evidence points at the potential of MP to adversely impact human health (Jenner et al., 2022; Leslie et al., 2022; Ragusa et al., 2021).

Preventing or reducing MP emissions into rivers will require the mitigation and elimination of their release at-source (Whitehead et al., 2021), and as such, it is crucial to assess the main sources of MPs into the riverine environment. However, inputs vary on spatial and temporal

Abbreviations: MP, microplastic; d.w., dry weight; WWTW, wastewater treatment works; CSO, combined sewer overflow; MSW, municipal solid waste; FTIR, Fourier-Transform Infrared Spectroscopy; PE, polyethylene; PEMA, poly(ethyl methacrylate); PMMA, poly(methyl methacrylate); PBM, poly(butyl methacrylate); PEA, poly(ethyl acrylate); PBT, poly(butylene terephthalate); PDP, poly(diallyl phthalate); PVP, poly(vinyl pyrrolidone); PVA, poly(vinyl alcohol); PP, polypropylene; PS, polystyrene; PVDC, poly(vinylidene chloride); PCT, poly(1,4-cyclohexanedimethylene terephthalate); PA, polyamide/Nylon 6; PC, polycarbonate; PAA, polyacrylamide; EVA, ethylene-vinyl acetate; PVC, poly(vinyl chloride); PB, polybutylene; PVS, poly(vinyl stearate); PVF, poly(vinyl fluoride); PVCA, poly(vinyl chloride/vinyl acetate.

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scales, making the assessment of specific source contributions an analytical challenge. While some MPs are intentionally produced in microscopic size (*primary* MPs, e.g., pre-production pellets, commonly known as nurdles), most MPs present in the environment originate from degradation of larger (macro) plastic items (*secondary* MPs) (Barnes et al., 2009). In practice, MPs can be emitted into the environment through point or diffuse sources. While point sources emit MPs from easily identifiable, confined areas (e.g. Wastewater Treatment Works [WWTW] effluent outfalls), diffuse sources of MPs are spread over larger areas (Eerkes-Medrano et al., 2015; Wang et al., 2017; Campanale et al., 2019). The main diffuse sources of MPs include discarded litter, tyre and road-marking wear and agricultural activities (Talbot and Chang, 2022). MPs accumulated on land enter rivers through surface runoff, or aeolian (wind-blown) inputs (Campanale et al., 2022). Such sources can be associated with different land uses; for example, urban areas are typically characterised by higher MP loads from WWTW (which receive greater sewage loads), road runoff and discarded litter, whereas waterways located in rural areas can receive considerable inputs from nearby agricultural land (Schmidt et al., 2020; Harley-Nyang et al., 2022).

Simultaneously, MP concentrations in sediment change over time. Both precipitation and wind can re-entrain and deposit MPs that have been previously deposited on floodplains or riverbanks (Horton and Dixon, 2018; Lechthaler et al., 2021). The input of MPs into rivers can increase with the onset of precipitation in the early autumn/winter (in the northern hemisphere), following MP accumulation on land during the drier months (Schmidt et al., 2018). This 'first flush effect' can not only lead to a rapid increase in diffuse inputs of MPs into rivers (e.g., input of MPs deposited on floodplains/riverbanks), but also from point sources of MPs (e.g., storm drains and/or CSOs (Maniquiz-Redillas et al., 2022; Schernewski et al., 2021). While some studies suggest that such delivery of MPs into rivers can be reflected in increased MP deposition in sediment (e.g., Chen et al., 2021), most point at MPs being remobilised from riverbeds under high river flows (Hurley et al., 2018; Eppehimer et al., 2021; Kiss et al., 2022). Nevertheless, most field MP studies have so far been conducted under steady flow conditions, failing to account for the hydrological controls on MP concentrations in sediment (Drummond et al., 2022; Krause et al., 2021). Simultaneously, bedform structures found in coarse-granular riverbeds, such as pool-riffle sequences, can facilitate the transport of MPs into deeper, hyporheic sediment (Frei et al., 2019; Boos et al., 2024). The reworking of such morphological units therefore further convolutes our already limited understanding of MP transport through river channels (Russell et al., 2023).

The River Thames (UK) is an ecologically and culturally important catchment, which supplies about two thirds of the drinking water for the local population (Greater London Authority, 2021). The river receives treated wastewater effluent from over 13 million inhabitants (Rowley et al., 2020), which is reflected in the considerable levels of MP pollution found in its surface waters (Rowley et al., 2020; Devereux et al., 2023). However, only one study to date has quantified the abundance of MPs in the sediments of the River Thames; Horton et al. (2017a) found average MP concentrations as high as 660 particles kg^{-1} in the sediment of three tributaries of the River Thames (the River Leach, the River Lambourn and the Cut). However, this study did not quantify the presence of small (<1 mm) fractions of MPs, which are thought to be the most harmful due to their higher capacity to adsorb toxic pollutants (Lee et al., 2014) and their higher potential to be ingested by aquatic biota, including commercially important species (Setälä et al., 2014; Horton et al., 2017b). To our best knowledge, the sources of MPs into the River Thames and the seasonality in their inputs are not yet established.

This study aims to address these research gaps through the following objectives: (*i*) Understand the extent of River Thames sediment contamination with MPs, including small MPs <1 mm; (*ii*) Assess the link between the concentration and characteristics (shape, size and chemical composition) of MPs retained in River Thames catchment sediments and adjacent land use; (*iii*) Based on MP characteristics, determine their possible sources into the River Thames catchment; and (*iv*) understand the seasonality in the presence of MPs in the sediment of the River Thames catchment. Through investigating MP levels on a seasonal basis over seven sampling events, we aim to provide a more representative assessment of MP presence in the sediments of the River Thames catchment, establish the most important MP sources year-round, and inform appropriate mitigation measures. We hypothesise that sediments of the River Thames catchment will contain a substantial degree of MP pollution originating from a mixture of point and diffuse MP sources associated with adjacent land-use. More specifically, we expect MP concentrations to increase from rural to urban to industrial sites and theorise that each land-use will be associated with different MP sizes, morphologies and polymeric compositions. On a temporal scale, we predict that MP concentrations will vary seasonally, increasing in the summer (i.e., under low-flow conditions). Finally, due to the changes in flow conditions that control MP deposition, we hypothesise that spatial trends will be more evident in the summer (under low-flow conditions that facilitate particle settling) than in the winter (high-flow conditions that facilitate particle resuspension).

2. Materials and methods

2.1. Sample collection

Sediment samples were collected from twelve sites within the River Thames catchment over a total of seven sampling events in July 2019, January 2020, March 2020, August 2020, March 2021, August 2021 and December 2021. Although the sampling events were originally planned to take place in January and July each year, the COVID-related lockdowns in the UK made it impossible to collect samples at consistent time increments. Two of the sampling sites (5 and 6) were located on the main River Thames and the remaining sites (1–4, 7–12) were situated on eight of its tributaries (Table 1). In line with previous freshwater MP studies (e.g., Chen et al., 2021; He et al., 2020; Soltani et al., 2022), sampling sites were classified into three main land use categories: rural-RUR (4 sites), urban-URB (5 sites) and industrial-IND (3 sites) (Table S1) (see Fig. 1).

Site classification was based on riparian land use, in the immediate vicinity of each sample site (i.e., river bank land use) and along a virtual 100 m-wide buffer strip for a further 1 km upstream. This information was gathered using a combination of catchment walkovers, cross-referenced against satellite imagery (Google Earth) obtained during

Table 1
Chemical composition of MPs ($n = 317$) extracted from sediment samples ($n = 101$) at rural (RUR), urban (URB) and industrial (IND) sites across the River Thames catchment throughout July 2019 - Mar 2021, based on μATR-FTIR/ATR-FTIR analysis. PE – polyethylene, Acrylates *(a group consisting of: poly(ethyl methacrylate) (PEMA), poly(methyl methacrylate) (PMMA), poly(butyl methacrylate) (PBM), poly(ethyl acrylate) (PEA))*, PBT – poly(butylene terephthalate), PDP – poly(diallyl phthalate), PVP – poly(vinyl pyrrolidone), PVA – poly(vinyl alcohol), PP – polypropylene, PS – polystyrene, PVDC - poly(vinylidene chloride), PCT – poly(1,4-cyclohexanedimethylene terephthalate), PA – polyamide/ Nylon 6.

	Rural	Urban	Industrial
PE	62.5 %	56.5 %	12.0 %
Acrylates	6.3 %	8.7 %	21.4 %
PVP	3.1 %	1.4 %	13.7 %
PVA	6.3 %	5.8 %	10.3 %
PCT	0.0 %	1.4 %	14.5 %
PP	3.1 %	13.0 %	4.3 %
PB	6.3 %	4.3 %	3.4 %
Phenoxy resin	0.0 %	0.0 %	6.0 %
EVA	0.0 %	2.9 %	3.4 %
PC	3.1 %	0.0 %	4.3 %
Other	9.4 %	5.8 %	6.8 %

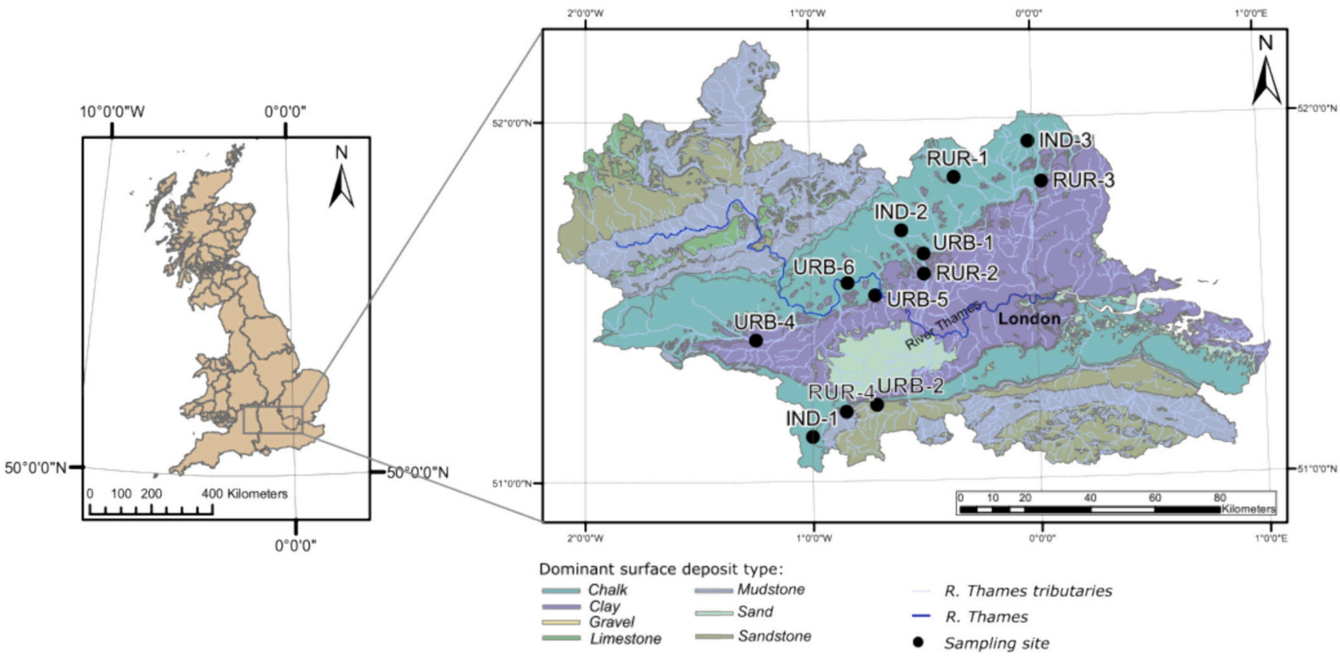

Fig. 1. Map of the River Thames catchment (UK) and the location of sampling sites included in the study, showing underlying geology and site category (RUR - rural; URB - urban; IND - industrial). Sourced from Ordnance Survey and Environmental Agency.

the study period. This allowed predominant land usage and management to be captured and potential MP sources (e.g., industrial sites) to be identified. However, the authors appreciate that whilst such an approach to land use classification is useful for capturing point sources it may be less effective at capturing diffuse or intermittent inputs such as those arising from road run-off, WWTPs or combined sewer overflows (CSOs) (Kukkola et al., 2024).

Samples were taken from coarse-granular sections of the catchment using a metal 'cookie cutter' sampling device (0.5 m in diameter and 0.75 m high; Klingeman and Emmett, 1982), which shielded the collection area from the river flow and prevented the wash-out of fine sediment and MPs. At each site, the cylinder was worked into the streambed to a depth of approx. 5 cm. To obtain an optimal representation of both coarse and fine sediment grains, a minimum of 3 kg of sediment was manually extracted using a steel shovel and subsequently transferred into a Ziplock bag. With the exception of July 2019, 2–3 sediment samples were taken from each sampling site. During summer sampling, sediment was collected across the channel cross-section (mid-channel and beside both riverbanks). However, access to some sites became limited in the winter months due to increased water depths and flows. Where the water was too deep to safely access the whole cross-section of the river, samples were taken from the inner banks of each river. This was unlikely to skew the results of the study, as MP concentrations were found not to significantly differ between morphological units (inner bank, outer bank and mid-channel; Kruskal-Wallis; $p = 0.83$).

2.2. Hydraulic parameter data

Water depth data were used as an indicator of changes to flow in the studied rivers due to gauging station flow measurements not being available for all of the sampling sites. These data were obtained from the National River Flow Archive, Environmental Agency (https://nrfa.ceh.ac.uk/) (Table S3) (CEH, 2022). For gauging stations where both river flow and depth data were available, both measurements revealed a statistically significant positive correlation (Spearman; $R = 0.91$, $p < 0.0001$). Water depth data were averaged over a period of one month prior to sample collection, which was considered to represent a reasonable optimum between the residence time of MPs in sediment (usually several months; Drummond et al., 2022) and the temporal resolution of the water level data (daily measurements). Although in-situ water velocity data was also collected from each sampling site using a flow meter, gauging station flow measurements allowed a better assessment of MP transport as they provided information about changes to flow patterns that occurred prior to sampling events.

2.3. Sample preparation

Prior to analysis, sediment samples were wet sieved into 0.5ϕ fractions (63 μm - 45 mm) and oven-dried at 40 °C overnight. The separation into 0.5ϕ fractions was required due to the large volumes of sediment processed, and the poor sorting of the sediment. Each sieve was then weighed to record the grain size distribution. Only material between 63 μm and 5 mm in size was taken for further analysis, in agreement with the most widely implemented definition of MPs (Frias and Nash, 2019). In addition, the sediment was separated into two fractions: 63 μm – 1 mm, and 1–5 mm, in line with the size classification of MPs (small and large MPs; Erni-Cassola et al., 2017).

2.4. MP isolation

Two separate methods were used to extract MPs from coarse (1 mm - 5 mm) and fine (63 μm – 1 mm) sediment fractions (Fig. 2). In accordance with the procedure described by Horton et al. (2017a), at least 200 g of >1 mm fraction was separated into 20-g subsamples and investigated under a binocular light stereomicroscope (Nikon SMZ800) at 10× magnification. Presumptive MPs (i.e., not yet confirmed to be plastic) were then removed using stainless steel tweezers and placed in a separate 90 mm glass Petri dish for subsequent visual analysis at a higher (20–50×) magnification.

MPs were extracted from the fine (63 μm – 1 mm) sediment fraction using a modification of the density flotation protocol outlined in Hurley et al. (2018), which incorporated using saturated NaCl solution (table salt; d = 1.2 g cm^{-3}) as the flotation solution. In total, 100 g of fine sediment was taken for analysis from each sample, in line with previous studies (e.g., Scherer et al., 2020; Eppehimer et al., 2021; Margenat

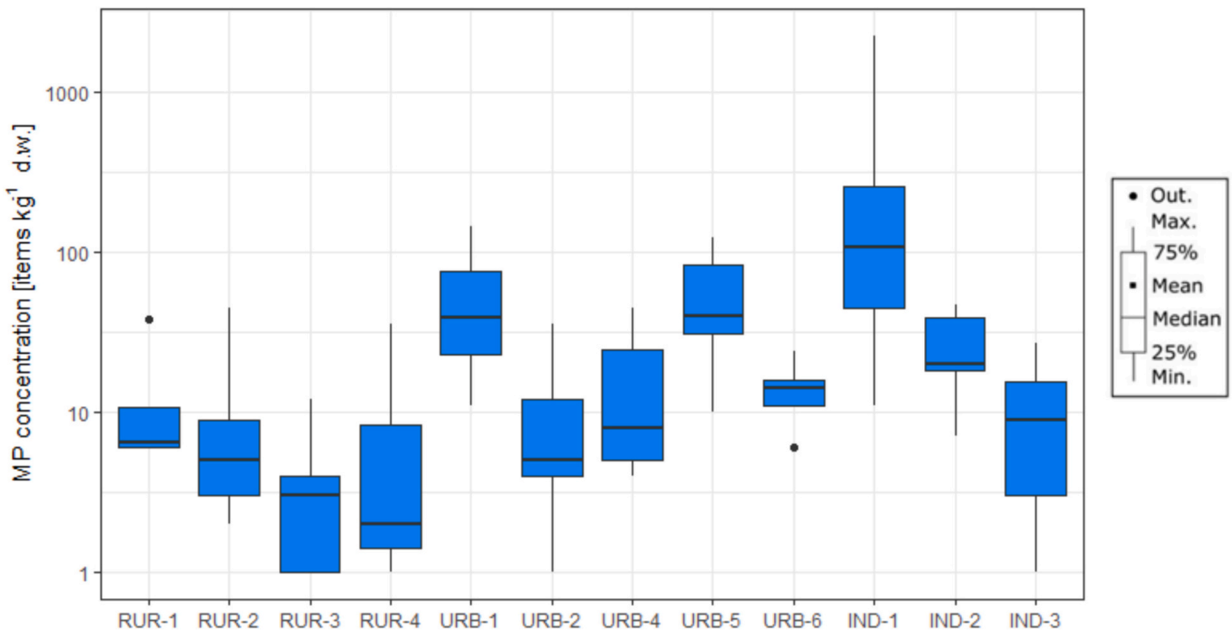

Fig. 2. MP concentrations (y axis log10-transformed) found in sediment samples ($n = 185$) taken from 12 sampling sites in the River Thames catchment throughout July 2019 – Dec 2021. MP – microplastic, RUR – rural, URB – urban, IND – industrial, d.w. – dry weight, out. – outlier (defined as >1.5 times the interquartile range above the third quartile or below the first quartile).

et al., 2021). Where the total volume of fines was <100 g in weight (9 out of the 185 samples), the total volume of fine material present in the sample (20–90 g) was analysed. Preliminary trials of different methods of organic matter digestion using these sediments did not produce good results, with insufficient organic matter removal to justify the additional process step. As the analysed material was not digested, the samples were divided across multiple filters to prevent the overlapping of particles and obtain an optimal amount of material for microscopic analysis.

2.5. MP identification

2.5.1. Visual analysis of MPs

Particulate matter extracted from the fine sediment fraction (63 μm – 1 mm) and presumptive MPs extracted from the coarse sediment fraction (1–5 mm) were visually investigated under a stereomicroscope (Nikon SMZ800) at 20–50× magnification. MPs were identified based on commonly employed criteria outlined in Hidalgo-Ruz et al. (2012). Where all of the criteria were met, the particles were counted as presumptive MPs and categorised according to their size and shape. MPs were categorised into the lowest number of morphologies possible (beads, fibres and fragments) to simplify the future inter-comparison of studies (Lusher et al., 2020). Items below 63 μm in size (measured along the longest dimension of the particle) were excluded from the total counts, in line with the lower cut-off point used in sediment sieving and because such small items could not be reliably identified using the visual method (Hurley et al., 2018). In total, about 5550 filter papers (from the analysis of the fine sediment fraction) and 185 sub-samples of suspected MPs extracted from the coarse sediment fraction were visually investigated. This resulted in the identification of 5280 small (63 μm – 1 mm) presumptive MPs and 782 large (1–5 mm) presumptive MPs.

2.5.2. Spectroscopic analysis of MPs

13 % of all items identified using the visual method over July 2019 - Mar 2021 were analysed (117 large and 349 small particles), in line with the proportion of MPs taken for spectroscopic analysis reported in previous literature (Baechler et al., 2020; Harris et al., 2022; Talbot et al., 2022).

The analysis was conducted using a Perkin Elmer Spotlight 400 Imaging system (4000–700 cm^{-1} IR spectral range) equipped with an ATR accessory. PerkinElmer Spectrum 10 software was used to compare the measured infrared spectra to the reference spectra from a polymer library (spectra database from S.T. Japan-Europe GmbH, Germany/Japan). Only particles with minimum 70 % similarity between spectra were considered to be polymers.

Overall, 82 % of large and 64 % of small particles originally selected as MPs were confirmed to be synthetic polymers, resulting in an average MP identification rate of 68 %. The final MP concentrations (of combined small and large MPs) reported in this study were adjusted using this correction factor.

2.6. Quality assurance and control

Quality control measures were taken throughout sample collection and subsequent analysis to prevent the cross-contamination of samples with extraneous MPs (Coppock et al., 2017). To prevent the contamination of samples during collection, all equipment was thoroughly rinsed with local river water prior to sampling to prevent cross-contamination between sites (Stanton et al., 2019; Scherer et al., 2020) and the persons involved in sample collection were stood downstream of the sampling point at all times to minimise potential contamination from plastic items (e.g., waders) or re-suspended substrate (Baldwin et al., 2016). In the lab, nitrile gloves and a 100 % cotton lab coat were worn at all times during sample handling. Wherever possible, work was carried out in a laminar flow cabinet (Bassaire UK, P-range). Work surfaces were frequently wiped with 70 % ethanol (Molecular Biology Grade, Fisher Scientific). Glassware was used instead of plastic wherever possible, but where the use of plastic labware was unavoidable (polypropylene Falcon tubes and Petri dishes), the items were new and unused. All vessels were covered with aluminium foil when not being used. Sieves were washed with a natural (coconut fibre) bristle brush prior to treatment of each sample. All liquid reagents used during the analysis were filtered using 0.22 μm cellulose nitrate filters (Whatman™) to remove any existing particulate contaminants. Preliminary trials of soaking the Ziplock bags in MilliQ water for several days did not show the release of any particles, therefore we believe contamination would have been negligible. Nonetheless, as a precaution, translucent MPs were discounted during the visual investigation of samples.

2.6.1. Calculation of procedural blanks

To calculate procedural blanks, triplicate blank samples containing only saturated NaCl solution were run in parallel during density flotation described in section 2.4. MPs present on the filters were then counted under a binocular light stereomicroscope, in line with the quantification procedure implemented for sediment samples.

As wet sieving was impossible to perform under laminar flow, potential contamination was monitored by placing dampened sterile cellulose nitrate papers (Whatman™) within the working area inside an open Petri dish. The filters were subsequently visually investigated under a stereomicroscope and the number of MPs counted on the filter area (4.7-cm diameter) was then extrapolated to obtain the hypothetical number of particles that could have entered the sieve area (21-cm diameter). Blank filters were also used during microscopic analysis of samples under a stereomicroscope to account for any airborne MPs.

As fibres were the only MP shape detected in the prepared blanks, the cumulative mean number of MPs found in the procedural blanks implemented in the different stages of sample analysis was subtracted from the total number of fibres found in environmental samples. The average concentration of fibres (found throughout all quality tests implemented prior to analysis of samples collected during each sampling event) in the blank samples was estimated at 8 fibres per sample and this number was subtracted from the total number of fibres present in each sediment sample. The Limit of Detection (LOD) was calculated in line with the procedure implemented by Horton et al. (2021) (LOD = average + 3.3 SD). Where the blank-corrected number of fibres present in the sample was lower than the calculated 3.3 SD (30 fibres per sample), it was assumed that the concentration of fibres in the environmental sample was below the limit of detection.

2.7. MP content calculation and statistical analysis

All MP concentrations found in sediment samples have been rounded down to the nearest whole number and normalised to the mass of sediment in the 63 μm - 5 mm size range (Keisling et al., 2020). The final values are expressed as the number of particles per kg of dry weight of sediment (items kg^{-1} d.w.), in line with the most commonly applied protocols (Masura et al., 2015). As data were determined to be non-normally distributed (based on the Shapiro-Wilk test), MP concentrations are presented using median values in addition to the average values typically reported in MP studies (Tibbetts et al., 2018).

Due to four of the sampling sites being inaccessible during some of the sampling events, only 8 (out of the 12) sampling sites were considered in the statistical analysis and calculation of seasonal averages included in section 3.2.1 (in order to remove the possible bias associated with different size data sets and more accurately represent the temporal changes in MP concentrations in the sediment). However, all available data points were included in calculations presented in section 3.2.2.

Due to the non-normality of the data, the Kruskal-Wallis non-parametric test was used to test for differences between the concentration of MPs at sites characterised by different land use. Where the results of the test were statistically significant, Dunn's Test was further conducted to determine which groups were different. Due to the non-normal distribution of the data sets, the strength of the relationships between MP concentrations and predictor variables was tested using Spearman's rank correlation coefficient.

Descriptive statistics (average, standard deviation, median, min, max) were calculated using Excel 2016 and R Software (vers. 1.3.959). All statistical tests were performed in R Software (vers. 1.3.959).

3. Results

3.1. Concentrations and characteristics of MPs in the river Thames sediment

Detectable levels of MPs were present in 89.7 % (166 out of 185) of sediment samples (i.e., in both the coarse and fine sediment fractions). MP concentrations varied substantially across the catchment (SD = 347 items kg^{-1} d.w.; Fig. 2) and ranged between 0 and 4241 items kg^{-1} d.w., averaging at 61 items kg^{-1} d.w. across all sampling seasons.

Sediment samples contained three main MP shape categories: fragments (58 %), beads (32 %) and fibres (10 %) (Fig. 3). While the morphology of most MPs (67 %) was consistent with that of degraded larger plastics, a third of the particles were 'primary' MPs, such as microbeads (Fig. 3h), glitter (Fig. 3c) or nurdles (Fig. 3i). Also of note was the presence of road marking paint fragments (incorporating glass beads ca. 100–250 μm in diameter) in some of the samples (Fig. 3d).

The majority (90 %) of MPs were below 1000 μm in diameter, with the median size falling into the 750 μm - 1000 μm size category (34 %) (Fig. S4). Beads were almost entirely composed of particles between 63 μm and 100 μm in size (99.7 %), with only scarce >1000 μm particles. Fragments were the most heterogeneous category of MPs in terms of their size distribution and included both small (68 % particles between 63 μm – 1000 μm) and large (32 % between 1000 μm – 5000 μm) MP fractions.

Only 68 % of particles originally selected for spectroscopic analysis were confirmed to be MPs. In total, 24 different polymers were detected in the samples. Polyethylene (PE) was the dominant polymer overall (33 % of total particle count), and acrylates were the second most abundant type of polymer (15 % of total particle count). Small numbers of biodegradable polymers, such as poly(vinyl alcohol) (PVA) or PVP (poly (vinyl pyrrolidone), were also detected (7 % and 8 % of total particle count, respectively). Fibres were composed of PE and PP (50 % of total particle count each), while beads were mainly PBT and PDP (44 and 29 % of total particle count, respectively) (Fig. S5). Fragments contained a highly heterogeneous mixture of polymers dominated by PE (38 % of total particle count).

3.2. Variations in MP presence and characteristics across sites and sampling events

3.2.1. Temporal variations

MP concentrations in the sediment increased during periods of low flow and decreased in high-flow conditions over July 2019 – March 2021 (Fig. 4). The second and third sampling events (January 2020 and March 2020) were conducted following storm events (Storm Brendan and Storm Dennis), which was reflected in a gradual reduction of MP concentrations in sediment. However, a particularly striking, 40-fold, decrease in average MP concentrations following the onset of high flows in the winter occurred between August 2020 and March 2021 (average MP concentrations of 301 and 7 items kg^{-1} d.w., respectively).

Interestingly, the final summer-winter couplet (August 2021 – December 2021) exhibited an opposite trend, with slightly elevated MP presence in the winter (average MP concentrations of 34 and 48 items kg^{-1} d.w. in August 2021 and December 2021, respectively). This observation resulted in the lack of significant difference detected between MP levels found in the sediment in the summer (low-flow) and winter (high-flow) seasons (Kruskal-Wallis, $p > 0.05$).

Although seasonal variations in flow appeared to strongly impact MP concentrations in the sediment, they had no appreciable influence on the shape composition, chemical composition, or sizes of MPs (Fig. S6 – S8).

3.2.2. Spatial variations

MP concentrations were significantly higher at urban and industrial locations relative to rural sites throughout the whole study (average MP concentration of 179 ± 528, 32 ± 37 and 8 ± 13 items kg^{-1} d.w. for

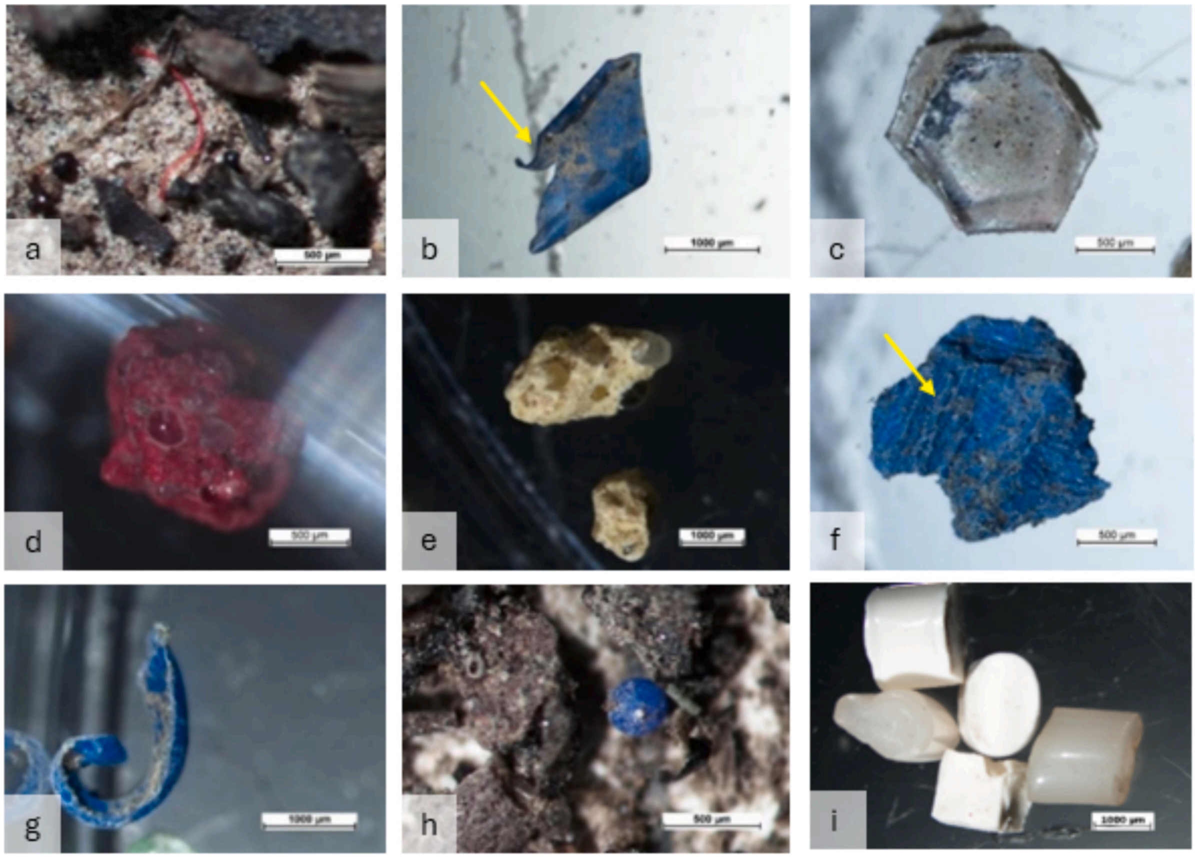

Fig. 3. Examples of MPs found in sediment samples taken from the River Thames catchment over July 2019 – Dec 2021: (a) fibre, (b) film-shaped fragment, (c) fragment of glitter, (d-e) fragments of surface marking paints with visible glass beads, (f-g) fragments present at industrial sites; (h) bead, (i) raw plastic pellets (nurdles). Signs of MP degradation are indicated with arrows.

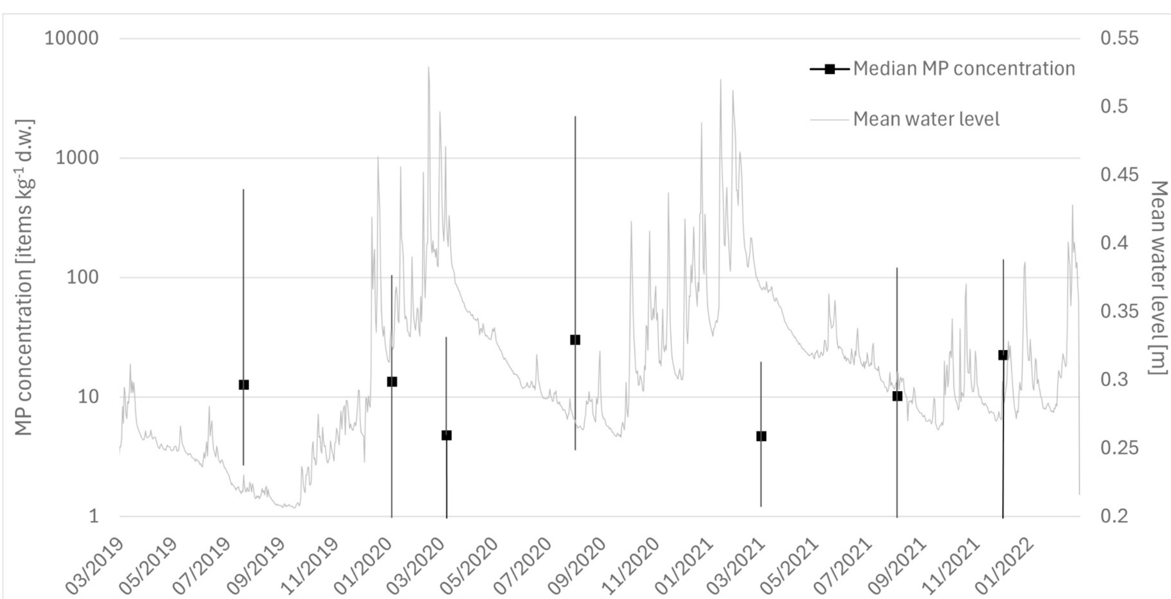

Fig. 4. MP (microplastic) concentrations detected in the sediment samples ($n = 144$) taken from eight of the twelve sampling sites in the River Thames catchment throughout seven sampling events (July 2019, Jan 2020, Mar 2020, Aug 2020, Mar 2021, Aug 2021, Dec 2021), plotted against mean water values found at the sampling sites. The black squares indicate median MP concentrations, and the whiskers indicate the minimum and maximum MP concentrations recorded at the sampling sites.

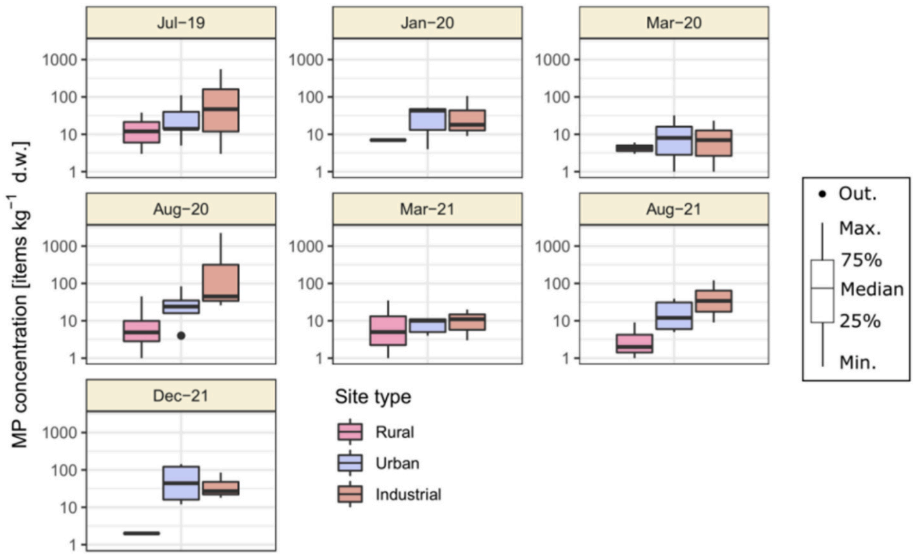

Fig. 5. MP (microplastic) concentrations detected in the sediment samples (n = 185) taken from sites characterised by different land use (rural, urban, industrial) across the River Thames catchment over the seven sampling events (July 2019, Jan 2020, Mar 2020, Aug 2021, Dec 2021).

industrial, urban and rural locations, respectively; Kruskal-Wallis, $p < 0.005$), although this trend was generally less pronounced during high-flow (Dec - Mar) periods (Fig. 5).

The influence of surrounding land use was also reflected in the variability of MP shapes, sizes and compositions found in sediment samples. Although fragments were the dominant shape of MPs irrespective of land use, industrial sites contained higher quantities of beads compared to sites dominated by rural and urban land use (Fig. 6). Although their exact numbers were not quantified, some of the MPs present at industrial sites showed patterns of possible mechanical abrasion, such as a curved morphology (Fig. 3g) or grooved surfaces (Fig. 3f), which have been previously linked to industrial processes as opposed to the naturally occurring degradation patterns (e.g., cracking or flaking due to interactions with sediment, see Fig. 3b; Warrier et al., 2016). Beads extracted from industrial sediment samples differed from those found at urban sites; while both were brightly coloured, beads found at industrial sites were much smaller (63–100 μm) than those present in urban areas (ca. 250 μm). Samples taken from industrial sites also contained the smallest MPs compared to urban and rural sites, with 79 % of MPs falling into the 63–100 μm size category (Fig. 7). In contrast, rural samples were dominated by MPs below 1000 μm in diameter (93 %), whereas urban samples presented a more evenly distributed spread of MP sizes and contained both small (<1000 μm) and large (>1000 μm) plastic particles (72 and 28 % of total, respectively).

PE was the dominant polymer at both rural and urban sites, accounting for 62.5 and 56.5 % of total particle count, respectively

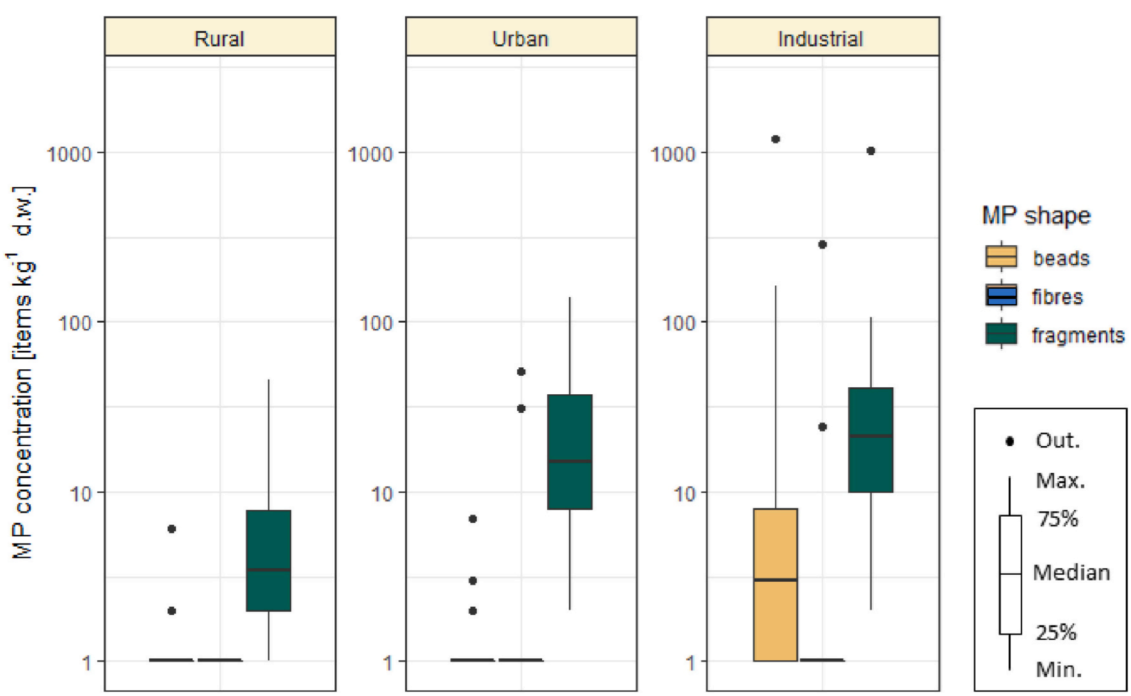

Fig. 6. MP concentrations split by the contributions of the different MP shapes (beads, fibres and fragments) detected in sediment (n = 185) at rural, urban and industrial sampling sites across the River Thames catchment over July 2019 – Dec 2021. MP – microplastic d.w. – dry weight, out. – outlier (defined as >1.5 times the interquartile range above the third quartile or below the first quartile).

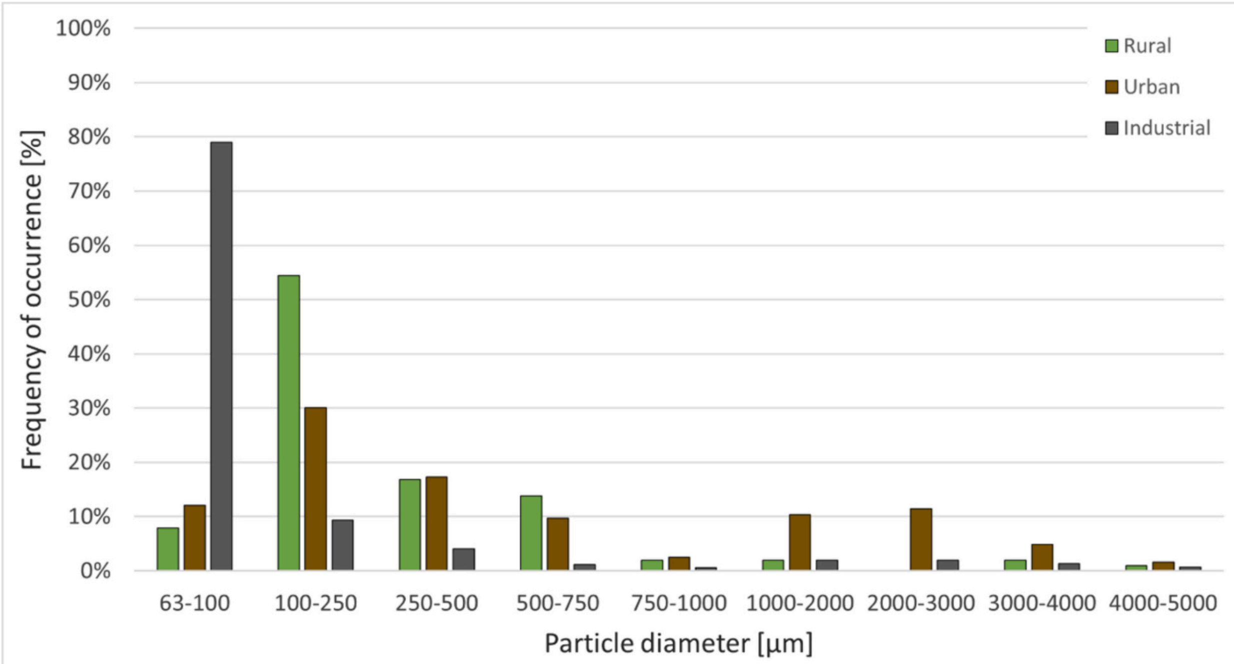

Fig. 7. Size composition (as % total count) of MPs (microplastics) found in the sediment of the River Thames catchment with respect to site type (rural, urban and industrial) sites throughout July 2019 – Dec 2021 (n = 185 samples).

(Table 1). In comparison, industrial sites contained a much broader spread of polymers, with a higher percentage of acrylates, PBT, PDP and PVP.

4. Discussion

4.1. Presence and characteristics of MPs in the river Thames catchment sediment

MPs were pervasive in the sediment of the River Thames catchment. Despite the large variation observed between samples and sites, values reported here (0–4241 items kg^{-1} d.w., average = 61 items kg^{-1} d.w.) are in the range typically found in the literature. Similar MP concentrations have been found in sediments of other British rivers, such as the River Tame (UK) (average = 165 items kg^{-1}, lower MP size cut-off of 63 µm; Tibbetts et al., 2018) or River Kelvin (Scotland) (161–432 items kg^{-1}, lower MP size cut-off of 63 µm; Blair et al., 2019), as well as in other rivers in Europe, e.g., Elbe, Mosel, Neckar and Rhine (Germany) (34–64 items kg^{-1}, lower MP size cut-off not reported; Wagner et al., 2014), the River Antua (Portugal) (100–629 items kg^{-1}, lower MP size cut-off of 55 µm; Rodrigues et al., 2018) and other parts of the world, such as the River Yangtze (China) (25–300 items kg^{-1}, lower MP size cut-off of 48 µm; Di and Wang, 2018) or the Neuse River Basin (USA) (0–808 items kg^{-1}, lower MP size cut-off of 64 µm; Kurki-Fox et al., 2023). Notably, much higher MP concentrations (average = 660 items kg^{-1}) were found in the previous study conducted in the River Thames by Horton et al. (2017a). Whereas sites sampled by Horton et al. (2017a) were located elsewhere in the catchment and were chosen based on the received loads of sewage effluent, this study included a wider range of sites that were not selected based on their proximity to nearby municipal wastewater inputs. The original study also used different analytical methods and only accounted for large (1–4 mm) MPs. Higher MP concentrations can generally be expected if a lower cut-off size is implemented, as it is generally thought that the smallest MP are the most abundant in the environment (Enders et al., 2015; Horton et al., 2021). On the other hand, the previous study incorporated the use of $ZnCl_2$ as flotation medium, while the use of NaCl in this study likely resulted in lower MP recovery overall.

The lower than previously reported numbers of MPs found in this study could also be a result of the sampling strategy. River sediment samples are typically collected through wading into a river, and most field studies that monitor MP concentrations in the sediment are conducted under low-flow conditions due to the ease and safety of sampling (including Horton et al. (2017a)). Under low-flow conditions, MPs are more likely to settle and be retained in sediments compared to high flows, which can remobilise MP from sediments and yield much lower MP concentrations (Hurley et al., 2018; Eppehimer et al., 2021; Kiss et al., 2022). As this study reports on MP concentrations taken over both low and high flow conditions, it likely represents a more realistic picture of MP retained in the sediments of the River Thames compared to studies that report on MP levels under low-flow conditions based on a one-off sampling event. Nevertheless, the average MP concentration found under low-flow conditions in this study were still lower than those previously found in the River Thames by Horton et al. (2017a) (141 items kg^{-1} and 660 items kg^{-1}, respectively).

Fragments were the most abundant shape of MPs present in the sediment, in agreement with findings reported in many recent studies (Wagner et al., 2014; Tibbetts et al., 2018; Gupta et al., 2021; Fan et al., 2022). At the same time, this observation was in contrast with that reported in the River Thames by Horton et al., 2017a, as well as other authors who found fibres to represent the most abundant MP shape retained in bed sediments (Murphy et al., 2022; Sá et al., 2022). While the small percentage of fibres could have reflected the true abundance of fibres in the sampled sediment, this observation could have been caused by the retention of fibres in deeper layers of sediment than sampled here (fibres have been found as deep as 0.6 m below sediment surface; Frei et al., 2019), or by sample processing artefacts (i.e., fibres could have been lost during sample sieving due to their cross-sectional diameter being smaller than the lower MP cut-off point implemented in this study, although this potential loss has not been quantified).

The size composition of MPs observed in this study is in line with the typical observation of small (<1 mm) MPs being more abundant in the environment (Horton et al., 2021; Sekudewicz et al., 2021; Tibbetts et al., 2018). Similarly, the dominance of PE in the sediment was in agreement with most previous freshwater studies (Jones et al., 2020). This is unsurprising, as PE is the most used polymer in the packaging industry and accounts for the largest share (61 %) of plastic waste emissions world-wide (PlasticsEurope, 2020). Most MPs found in the samples (76 %) were composed of low-density polymers. While this could have been a result of using NaCl as flotation media (Way et al., 2022), natural interactions that MPs undergo in the environment, such as biofouling or degradation, alter the intrinsic properties of polymers (Parrish and Fahrenfeld, 2019). The visual inspection of MPs revealed varying extents of their degradation, suggesting their density has likely not corresponded to that of virgin polymers.

An unexpected observation was the presence of low numbers of biodegradable polymers, such as poly(vinyl alcohol) (PVA) or PVP (poly (vinyl pyrrolidone) at all site types. PVA is used to produce biodegradable packaging, laundry detergent pods and contact lenses, whereas PVP is added to personal care products including shampoos and toothpaste (Gebreselassie, 2021). Although both polymers are widely marketed as biodegradable, studies have found their biodegradation in aquatic settings to be negligible (Alonso López et al., 2021) and a number of studies have reported their presence in freshwater settings (Gupta et al., 2021; Huppertsberg et al., 2020). Due to being water-soluble, PVA and PVP do not meet the definition of MPs (i.e., solid, water-insoluble particles), which can no longer be used in rinse-off cosmetics in the UK (European Commission, 2017). However, we chose to include them in the total MP counts due to their synthetic origin and likely prevalence in aquatic settings, which should be explored further.

4.2. Temporal variations in MP presence

Although it has previously been predicted that the residence time of MPs decreases with increasing river flows (Drummond et al., 2022), the data collected showing high concentrations early after the onset of high flows (December 2021) suggests that MPs are only remobilised once the critical shear stress required to remobilise the sediment fractions present in the riverbed is exceeded. It is believed that such substantial shear stresses occurred prior to sampling events conducted in January – March 2020 and March 2021, which coincided with the second and third greatest flooding events (after winter 2013/2014) in the River Thames catchment in this decade.

This phenomenon could be explained by the occurrence of sediment rearrangement under increasing shear stresses. Low excess shear stresses (e.g., in the beginning of a flood event) are associated with kinematic sorting of sediment, whereby fine grains infiltrate the riverbed, leading to the formation of an armour layer (Parker and Klingeman, 1982; Cooper and Tait, 2009). As found in flume experiments investigating MP movement in sediment under flood waves, this stabilises the bed and traps the fine fractions (including MPs), enhancing the sink behaviour of the sediment. Once armour layer breakup occurs under much higher excess shear stresses, MPs (and fine sediment) are rapidly released and the riverbed finally transitions from being a sink to a source of MPs (Ockelford et al., 2020).

At the same time, it has been determined that increased flows can also drive small particles ($\leq$100 μm) deeper into the riverbed through hyporheic exchange (Drummond et al., 2022). As such, MPs can be retained in the deeper, less mobile regions of the sediment over longer time periods (Drummond et al., 2020). Given the generally small sizes of MPs detected in this study, it is possible that this winnowing process (rather than remobilisation) was responsible for the apparent loss of MPs from the investigated surface layers of sediment in the winter months. Indeed, previous studies reported high concentrations of pore scale MPs (20–50 μm) in freeze cores as deep as 0.6 m (Frei et al., 2019). While these harmful fractions of MPs could persist in the sediment over long timescales, the predicted increase in flooding raises concern over a potential increased spread of plastics through the environment in the future (Kundzewicz et al., 2017).

4.3. The influence of land use on MP presence and characteristics

The sites included in this study revealed different degrees of MP pollution depending on adjacent land-use, with rural sites being statistically less contaminated compared to urban and industrial locations. This is in agreement with the widely reported trend of MP concentrations in riverbeds increasing in urbanised areas relative to less populated locations (Chen et al., 2021; Fan et al., 2022; Schell et al., 2021; Talbot and Chang, 2022; Xu et al., 2021; Zhang et al., 2021).

Although the association between MP concentrations and the presence of WWTW/CSOs upstream of the sampling sites was not investigated, the morphology of MPs found at rural and urban sites was consistent with that typically attributed to wastewater discharges, with a mixture of both secondary (fibres and fragments) and primary (beads and glitter) particles being present (Horton and Dixon, 2018; Hurley et al., 2018). Notably, the size and morphology of beads matched those reported for personal care products (e.g., facial scrubs or toothpaste) (Prata et al., 2018). As cosmetic products containing such microbeads have not been sold in UK since June 2018 (European Commission, 2017), this may suggest either a continued use of such products, or the occurrence of a greater degree of microbead persistence and retention in riverbeds and the wider river catchment than previously assumed (Weber and Lechthaler, 2021).

Sediment extracted from a number of urban sites also contained fragments of yellow road marking paint incorporating glass beads, which are added to such paints to enhance their reflectivity. The presence of identical MPs was also reported in the River Thames by Horton et al. (2017a), who attributed them to storm drains carrying runoff from nearby roads into rivers. Indeed, some polymers detected in urban sediment samples (PDP and acrylates) have been previously found in road dust (Kitahara and Nakata, 2020), urban canals and storm water channels (Boni et al., 2022), suggesting a possible surface runoff contribution. At the same time, it should be noted that UK sewerage systems can receive MPs from municipal wastewater, industrial effluents, stormwater and even landfills (Okoffo et al., 2019), making it challenging to pin-point the exact source of MP.

Industrial sites included in this study were associated with particularly high MP levels (Talbot and Cheng, 2022), in line with previous studies that found industrial wastewater to contain three-fold the number of MPs present in domestic wastewater (Long et al., 2021). Discharge limits for industrial facilities are often poorly regulated and the pollutant levels present therein are generally not disclosed, particularly for emerging contaminants, such as MPs (Franco et al., 2020). In comparison to urban and rural sites, industrial sites contained much smaller MPs in the bed sediment, most of which were fragments and microbeads between 63 and 100 μm. Such a decrease in MP size with increasing contribution from industrial sources is a well-described phenomenon (Su et al., 2020; Wang et al., 2020). The visual analysis of MPs found in industrial areas also pointed at the likely industrial wastewater origin; for example, the curved shape and mechanically abraded surface of fragments was in line with the previously described morphology of MPs produced in the process of drilling and machining plastic materials (Prata et al., 2018; Long et al., 2021). Fragments were composed of a wide range of polymers used across food packaging (PE, PP, PS), as well as in industrial applications. For example, PC, PP, PA and PBT found in the sediment are the among most used polymers in the electrical industry (Kousaiti et al., 2020), while PA, PC, PMMA (poly (methyl methacrylate) and PVC (poly(vinyl chloride) are widely utilised in building and construction (Lahtela et al., 2019). Although all three of the industrial sites included in this study contained partially submerged sewage outlet pipes, only one site (IND-3) was publicly reported to also receive industrial discharges. Simultaneously, all of the sites were littered with industrial waste, such as PVC piping or Styrofoam, the degradation of which could have produced MPs composed of polymers used in industry.

A particularly striking observation was the presence of high numbers of beads at industrial sites. Despite the general belief that the use of microbeads had been banned in the UK (see also discussion above), the 2018 MP legislation only included the incorporation of plastic beads in rinse-off cosmetic products, such as facial scrubs or toothpaste (European Commission, 2017). As such, microbeads continue to be intentionally added to a plethora of products and are widely used in industrial processes (Ding et al., 2019; Woodward et al., 2021). Some of the most common applications in the industrial sector include plastic media blasting, whereby MPs are used as abrasives, e.g., to clean vehicle surfaces or strip paint, where less aggressive stripping (compared to media such as sand or steel grit) is required (Galafassi et al., 2019). Beads also represent an important ingredient of paints due to their ability to enhance colour or improve the resistance of the paint to scratches (Lassen et al., 2015). The small size and intense colouring of beads found at industrial sites was in line with the characteristics of those added to paints, which have a diameter between a few and a hundred microns (Galafassi et al., 2019). Nevertheless, as microbeads are also often found in sewage, it is impossible to make conclusions regarding their origin (Prata et al., 2018).

5. Conclusions

Despite offering undoubted societal benefits, the ever-increasing production of plastics over recent decades has led to their accumulation in terrestrial and aquatic environments. This study is the first to investigate the extent and characteristics of MP pollution in River Thames catchment sediment over both space and time. MP were prevalent and present in a range of sizes, shapes and chemical compositions, all of which were influenced by the surrounding land use. In particular, industrial and urban locations were substantially more contaminated than rural areas, with some industrial sites representing MP hotspots. MPs retained in sediment at industrial sites appeared to originate from industrial processes, such as plastic media blasting. Better control of industrial effluent at source (e.g., through the use of ultrafiltration) could therefore greatly help prevent their environmental release.

Findings from this study confirm that riverbeds can likely act as both sinks and sources of MPs depending on the flow conditions, although further research will be required to account for the mechanisms that underpin the loss of MPs from sediments under high flows. In light of ongoing climate changes and the predicted increase in flooding, it will be crucial to conduct further research to understand the influence of such extreme weather events on the dispersal of MPs through the environment.

This study provides new insights into the mobility of MPs in coarse-granular river sediments and sheds light on the main sources of MPs into the River Thames, contributing to a better collective understanding of MP fate in freshwater settings.

CRediT authorship contribution statement

Karolina Skalska: Writing – original draft, Visualization, Methodology, Formal analysis. **Annie Ockelford:** Writing – review & editing, Supervision, Funding acquisition, Conceptualization. **James Ebdon:** Writing – review & editing, Supervision, Conceptualization. **Andrew Cundy:** Writing – review & editing, Supervision, Conceptualization. **Alice A. Horton:** Writing – review & editing.

Declaration of competing interest

The authors declare no competing interests.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.marpolbul.2024.116881.

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