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Hydrologic controls on the accumulation of different sized microplastics in the streambed sediments downstream of a wastewater treatment plant (Catalonia, Spain)

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Abstract

Wastewater treatment plants (WWTPs) act as a point source of microplastics (MPs) to freshwater ecosystems. Although MP abundance has been linked to high-density population areas, the mechanisms of how river hydrodynamics and particle size influence MP accumulation in streams are still largely unknown. This study investigated the spatial distribution of MPs within streambed sediments downstream of a WWTP effluent in Cànoves stream (Montseny, Catalonia) during baseflow conditions. MP concentrations from an upstream control site were compared to the WWTP bypass that added untreated wastewater at times when stream discharge exceeded capacity. The 450 m section investigated downstream of the WWTP consisted of three geomorphically altered sub-reaches interspersed between three unaltered buffer sub-reaches, each ∼75 m that provided a range in hydrologic conditions. Measurements of MP characteristics, hydrogeomorphic variables, and fine particles were simultaneously taken. MPs were quantified following the Nile red fluorescence method for large (>64 μm) and small (10–64 μm) particles. MPs in sediment samples downstream of the WWTP were mainly fragments with a higher abundance of small MPs (85 particles/g of sediment) vs large MPs (9 particles/g of sediment). While the abundance of large MPs in streambed sediments decreased with distance from the WWTP point source, the abundance of small MPs increased. Furthermore, the area of small MPs decreased with distance from the WWTP. MPs were most abundant at the WWTP bypass, suggesting these infrequent inputs during storm events represent an important source of MPs to the stream. Higher MP abundance coincided with increased organic matter content and smaller sediment grain sizes. Overall, our results present significant findings that could help explain differences in transport and accumulation patterns of MPs that influence their retention times in streambeds, suggesting a combination of preferential filtration in the streambed sediments, and fragmentation of larger particles.

1. Introduction

Plastic is an emerging pollutant, ubiquitous worldwide due to its widespread use and recalcitrant nature (Eerkes-Medrano *et al* 2015, Erni-Cassola *et al* 2017). The fate of microplastics (MPs, defined as 1–1000 μm, Hartmann *et al* 2019) in freshwater environments has recently gained more attention. Rivers have been identified as an important transport pathway of land-derived plastics to the ocean, accounting for 80% of global annual emissions (Meijer *et al* 2021) as well as systems where plastic waste is creating environmental impacts (Krause *et al* 2021, Kukkola *et al* 2021). However, rivers also function as a temporary sink of MPs, with long retention times of plastics in freshwater ecosystems,

especially within streambed sediments, providing the opportunity for modifications of these particles prior to reaching the oceans (Hoellein *et al* 2017, Drummond *et al* 2020, Liro *et al* 2020). All field studies targeting streambed environments to date have identified the presence of MPs in streambed sediments (Lambert and Wagner 2018, Bellasi *et al* 2020), with MP abundance in freshwaters strongly linked to population, hydrologic conditions, and stream geomorphic controls (Mason *et al* 2016, Hoellein *et al* 2017, Nel *et al* 2018, Watkins *et al* 2019, Krause *et al* 2021). However, mechanistic understanding of the drivers and controls of the trajectory of MPs from their sources to their eventual sinks in the oceans is lacking. Major knowledge gaps are remaining for instance on how close to the source MPs initially deposit in streambed sediments, how long they remain in place, and the processes that occur during this retention time that can alter their properties prior to their transport further downstream.

Wastewater treatment plant (WWTP) effluents are suspected to represent a major source of MPs (Bretas Alvim *et al* 2020), as well as organic matter and bacteria (Pascual-Benito *et al* 2020) to freshwater ecosystems, with estimates of approximately 5 million MPs released per WWTP per day (Mason *et al* 2016). The transport dynamics of MPs in riverine systems is expected to follow the same trajectory as other fine particles, with recent studies demonstrating similarities between MPs and naturally occurring allochthonous particles (Hoellein *et al* 2017). However, there are few studies that include simultaneous measurements of MPs with other types of fine particles. When taken together, this simultaneous measurement could help explain differences in transport and accumulation patterns that influence MP and non-plastic particle retention times in streambed sediments. The deposition of fine particles will be influenced by hyporheic exchange, the transport with solute from the water column into streambed sediments due to both advective flow and turbulence-driven exchange (Boano *et al* 2014, Drummond *et al* 2014a, 2020). However, the MP specific fate within sediments will be dependent on their polymer characteristics such as size, density, shape, and degree of biotic interaction through biofouling (Dietrich 1982, Kooi *et al* 2017). We here monitor multiple types of fine particles (synthetic polymers, organic and inorganic matter) simultaneously, downstream of a point source with regular fine particle inputs, in order to advance mechanistic understanding of the similarities and differences in their transport characteristics, which will be essential for improving our ability to predict MPs fate in freshwaters.

The initial hypothesis is that particle retention and downstream transport will be influenced by both local hydrologic conditions and MP characteristics (i.e. size, density, shape). For instance, MPs exceeding the pore size of the sediment will more likely be trapped at the sediment-water interface, while smaller particles are more likely to be transported via hyporheic porewaters into the streambed and either be retained in the sediments due to filtration/attachment processes or flow back to the water column (Drummond *et al* 2017, Hoellein *et al* 2019). Furthermore, the physical and chemical properties of smaller MPs may be preferentially altered within streambed sediments because of their higher surface area to volume ratios. Hyporheic abrasion or fragmentation of particles leads to increased surface area and potentially enhanced colonization/attachment of microbes, which could facilitate biodegradation or serve as a vector for pathogenic bacteria or viruses (Harrison *et al* 2018). MPs can also alter stream ecosystem functioning by affecting multiple trophic levels (Nelms *et al* 2018, Romera-Castillo *et al* 2018, Windsor *et al* 2019, Kukkola *et al* 2021), with increased potential of bioaccumulation with decreasing particle size (Rodrigues *et al* 2018, Krause *et al* 2021). For this reason, studies differentiating between MPs size ranges in their transport from source to sink, and especially the inclusion of smaller MPs sizes, are needed to assess risks associated with MPs in freshwater environments.

This study therefore aims to quantify MPs in the sediments downstream of a WWTP effluent, to assess the dependence of MP properties (i.e. abundance, size—length and area, shape) on distance from a point source, the local hydrologic conditions and the presence of other fine particles such as organic matter and fine sediments, in order to explain how the mentioned variables could influence transport and accumulation of MPs within streambed sediments. We expected higher MP accumulation downstream vs upstream of the WWTP effluent and that retention of MPs would relate to the hydrogeomorphic conditions (e.g. flow or water depth) within the sub-reaches and follow the same transport behavior as other fine particles measured within the study.

2. Methods

2.1. Site description

This study was conducted in Cànoves i Samalús (Catalonia, Spain, figure 1), a region with sub-humid Mediterranean climate and annual average rainfall between 700–1000 mm (Meredith *et al* 2021). Precipitation occurs mainly during Autumn and Spring with low precipitation in the Summer months. The study site is a 450 m reach of the Cànoves stream approximately 300 m above sea level, located downstream of the municipal WWTP (figure 1 and text S3 (available online at stacks.iop.org/ERL/16/115012/mmedia)). The WWTP represents the most upstream point of sewage effluent to the stream. The stream upstream of the WWTP is mainly intermittent during the summer months but then perennial downstream of the WWTP due to its almost continuous

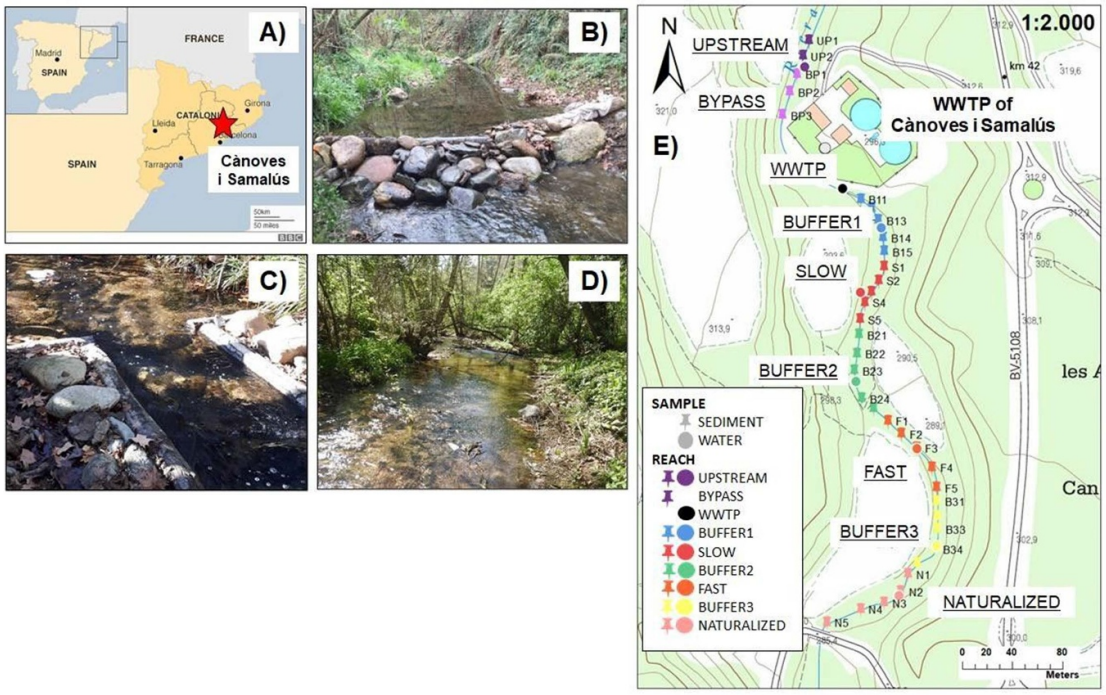

Figure 1. (A) Location of Cànoves i Samalús, Catalonia, Spain. Photographs of the modified sub-reaches: SLOW (B), FAST (C), and NATURALIZED (D). Site map with sampling locations in Cànoves stream with six 75 m sub-reaches downstream of the WWTP effluent identified (E).

effluent input (daily discharge ranges from 0.008 to 0.02 $m^3\ s^{-1}$) (Pascual-Benito *et al* 2020). The Cànoves WWTP treats the sewage waters from three villages, comprising a population of around 9200 inhabitants.

During the experimental period in March 2019, mean atmospheric temperature oscillated between 5 °C and 15 °C (Meteoblue 2019) with minimal rain (1.5 mm).

2.2. Experimental approach

We sampled streambed sediment every 15 m and surface water every 75 m within a 450 m long section of the Cànoves stream (Catalonia, NE Spain, figure 1) downstream of the local WWTP effluent and a 75 m sub-reach upstream. The 450 m downstream reach consisted of 75 m sub-reaches; three modified sub-reaches, each separated by unmodified sub-reaches (buffers). Morphohydraulic characteristics from these sub-reaches were modified by applying three types of interventions using bioengineering techniques and characterized as SLOW, FAST and NATURALIZED (figures 1(B)–(D)) (Meredith *et al* 2021). The bioengineering interventions were done in November 2015 by a local project (Naturalea 2017) in order to increase and improve the retention and auto-depuration capacity of the stream. Transversal logs were added to the SLOW sub-reach, generating small water dams. In the FAST sub-reach, logs were placed on the riverbanks to act as deflectors, thus increasing the water velocity and turbulence. In the NATURALIZED sub-reach, a widening of the channel and increase in sinuosity diversified stream habitats (figures 1(B)–(D)).

In the 75 m upstream sub-reach sampling consisted of one surface water sample at the start of the reach and a sediment sample every 15 m (figure 1(E)); three sediment samples near to where a bypass directly adds untreated wastewater to the stream when the capacity of the WWTP is exceeded (15, 30 and 45 m upstream of the WWTP effluent) and two sediment samples before the influence of the bypass (60 and 75 m upstream of the WWTP effluent). An additional water sample was taken directly from the WWTP effluent pipe. Each sample was analyzed for MP abundance and size (length and area). Streambed sediment samples were also analyzed for organic matter content and classified by grain size. Within each 75 m sub-reach, a conservative tracer injection was conducted within 1 week of sampling to characterize the discharge and the sub-reach-averaged hydrologic conditions (water level, channel width and average velocity).

2.3. Field methods

Streambed sediment and surface water sampling was conducted on 28 March 2019. 100 l of surface water from the aforementioned locations was filtered through a 64 μm sieve (eight samples). The material collected on the sieve was transferred with deionized (DI) water to a 20 ml glass vial and stored until lab analysis (section 2.4). Following the same procedure, WWTP effluent was sampled directly from the outlet pipe.

In total, 35 streambed sediment samples were taken—30 downstream, three at the bypass location,

and two at a control site before the influence of the bypass at 60 and 75 m upstream of the WWTP effluent (figure 1(E)). Prior to sampling the downstream sites, water depth and channel width were measured at each location. Then a bottom-cut metal bucket was used to restrict water flow and aid in collecting the sample with minimal loss, and approximately 170 g of streambed sediment was collected with a metal shovel and placed into a glass jar. Stones, branches and large materials (>2–3 cm) were removed by hand.

Discharge (Q, l s^{-1}) was estimated for each sub-reach from a conservative solute tracer injection (bromide as NaBr, Emsa, Barcelona, Spain) following the dilution-gauging method (Kilpatrick and Cobb 1985). Velocity (v) was estimated from the water flow measured at each sub-reach (Q_i), water depth (d_i) and channel width (w_i) from each sampling point as $v_i = Q_j/d_i/w_i$.

2.4. Laboratory methods

2.4.1. Sample processing—MP extraction, digestion, and staining

Sediment samples were oven-dried at 60 °C, while covered with perforated aluminum paper to avoid airborne contamination. The sediment was stirred once a day to facilitate the drying process, which took place over approximately two weeks. MPs were extracted from sediments using a zinc chloride ($ZnCl_2$, Sigma-Aldrich®) solution of 1.5 g cm^{-3} and a modified version of the sediment-MP-isolation (SMI) unit (Coppock *et al* 2017, Nel *et al* 2019). For each sediment sample, 50 g of sediment was mixed with 750 ml of $ZnCl_2$ solution and left to settle in the SMI unit for 15–30 min. The supernatant was then filtered through a 64 μm sieve. After that, the filtrate was mixed and 150 ml was filtered through a 10 μm sieve. Therefore, two subsamples of the different size fractions resulted from each sediment sample, and the >64 and 10–64 μm subsamples were treated separately from this point onward. The collected residue with the MPs in the 64 and 10 μm sieves were each rinsed with 30% hydrogen peroxide (H_2O_2) (Scharlab, Scharlau, EssentQ®) into separate glass jars, and filled to reach 20 ml of H_2O_2 and 2 ml Fe^{2+} (0.05 M) (modified method of NOAA, 2015; see supporting information (SI) for further details, text S2). The digestion solution was left for 24 h at room temperature. Post-digestion, each sample was filtered through the respective sieve (either 64 or 10 μm) and the residue with the MPs was transferred with DI water to a 20 ml vial. To each 20 ml vial, 100 μl of a Nile red stock solution (1 mg ml^{-1} in acetone, Maes *et al* 2017, Nel *et al* 2021) was added, shaken every 10 min for 30 min, and kept covered in aluminum foil in the dark at room T. After 24 h, the >64 μm sub-sample was filtered through a GF/D filter (Sigma-Aldrich®, Whatman® glass microfiber filters, ø 47 mm, pore size ø 2.7 μm), and the 10–64 μm sub-sample was filtered through a 10 μm polycarbonate track etch filter (Sigma-Aldrich®, Whatman®, Cyclopore® cyclopore PC circles, ø 47 mm, pore size ø 0.8 μm), placed in a clean polypropylene (PP) petri-dish and dried at 60 °C for 48 h. Surface water samples were processed by digesting and staining as described above. Negative controls for key steps throughout the MP analysis were taken to quantify contamination during lab processing. Air samples from the field site and the laboratory were also taken to confirm air contamination was minimal. Controls included sampling the $ZnCl_2$ solution, one before the first sample, another halfway through, and after the processing of the last sample. Controls were digested and stained following the same procedure (see SI, text S1 and table S2).

To avoid sample contamination, non-plastic materials were used whenever possible, all materials were rinsed three times with DI water prior to use, and a 100% cotton lab-coat was worn at all times in the lab.

2.4.2. MP counts and size analysis

A stereo microscope (Nikon SMZ-1000) fitted with a 0.75x objective lens, light source (Nikon Intensilight C-HGF1), and a 5 megapixels camera (Nikon DS-Fi1) was used to identify and count MPs. A green fluorescent protein-B filter set (excitation filter bandwidth of 470/40 nm, emission filter bandwidth of 535/50 nm, and a dichromatic mirror at 500 nm) was used in fluorescence mode. Nikon Intensilight C-HGF1 was adjusted as follows; shuttle open and neutral density scale was at 1 (100%) transmittance (Nel *et al* 2021). Imaging Software NIS-Elements Br (Nikon) was used to measure the fluorescence intensity (a.u.), length (μm), and area (μm^2) of each particle (see SI, figure S1).

Particles with a fluorescence intensity over 50 (a.u.) and 150 (a.u.) were considered as MPs, for >64 μm and 10–64 μm, respectively (Tamminga *et al* 2017, Nel *et al* 2021). In this way, false-positive readings were avoided since organic matter is not always completely removed during digestion, especially chitin and lignin substances which have reduced fluorescence after digestion (*quenching* effect) (Shim *et al* 2016, Erni-Cassola *et al* 2017, Maes *et al* 2017, Nel *et al* 2021). For large MPs (>64 μm), three subsamples with a field of view (FOV) equal to 0.22 cm^2 were analyzed. Thereafter the combined FOV total (0.66 cm^2) was extrapolated to the total area of the filter ($A_{GF/D\ filter}$ = 10.75 cm^2). For small MPs, four subsamples with a FOV equal to 0.031 cm^2 were analyzed and the combined FOV total (0.124 cm^2) was extrapolated to the total area of the filter ($A_{GF/D\ filter}$ = 10.75 cm^2). A length-area ratio (L/A ratio) was calculated as a descriptor for the shape of the particles. A lower L/A ratio suggests a more irregular particle shape, while a higher L/A ratio can indicate increased symmetry. A subset of suspected particles (>100 μm) (SLOW: N = 56; BUFFER 1: N = 16; FAST; N = 25) were extracted and analyzed

including polymer identification with a PerkinElmer Spotlight 400 Fourier-transform infrared spectroscopy (FTIR) system combination of a single element detector MCT 100 × 100 μm and range 4000–580 cm^{-1} by ALS Scandinavia AB, Sweden.

2.4.3. Organic matter quantification and grain size classification of sediment samples

The ash-free dry mass method was followed (American Public Health Association 1998) to estimate total particulate matter (i.e. inorganic and organic material, TPM) and particulate organic matter (POM) for each sediment sample. A known volume of dried sediment sample (20 g) was placed in pre-weighed aluminum weight pans. The sediment samples were then placed in a muffle furnace at 500 °C for 5 h and then a conventional oven at 60 °C for 24 h until the dry weight was stabilized. The difference in the pre-and post-weight is measured as POM (g).

For grain size classification, a 100 g sample of dried sediment was sieved through six sieves of different pores size (2, 1, 0.5, 0.25, 0.1 and 0.032 mm). The remaining sediment at each sieve was weighed and the corresponding percentage was calculated.

2.5. Statistical analysis

Descriptive statistics (mean, standard deviation, median, minimum and maximum values) of all parameters were calculated using R Studio Version 1.1.463. Non-parametric Kruskal Wallis test was used to test for significance between sub-reaches (IBM SPSS Statistics 25). Relationships between variables (MP characteristics, distance, and hydrologic parameters) were examined by applying bivariate regression models (linear, exponential, potential, and logarithmic). Fits were performed by ordinary least squares, and the goodness of fit (R^2) was used for model selection (Zar 2010). We refer only to the best-fit model in each case. A principal component analysis (PCA) was carried out with IBM SPSS Statistics 25, to determine the relationship between grain size classification, organic matter content, and MPs. The PCA was conducted with varimax rotation, and both Bartlett's Test of Sphericity was shown to be significant ($p = 0.000$) (Bartlett 1954), and that the Kaiser–Meyer–Olkin measure was below the acceptable limit of 0.5 (KMO = 0.303) (Kaiser and Rice 1974).

3. Results

3.1. MP abundance in sediment and surface water samples and dependence on the distance from the WWTP effluent

All of the analyzed sediment ($N = 35$) and water ($N = 8$) samples contained particles that were identified as MPs, with lengths ranging from 20 μm to 2.5 mm (see SI, table S1) and particle shapes being dominated by fragments (95.3%) over granules (4%) and fibers (0.7%). While fragments were observed in both the 10–64 μm and >64 μm size-ranges, granules were only identified in the 10–64 μm sub-samples and fibers only in the >64 μm sub-samples. Polyvinyl chloride (PVC) was the most dominant polymer type found (53.1%), followed by polyvinyl chloride acetate (PVCA) (28.7%), and polyethylene (PE) and polyester (5.10%). Negative lab controls (i.e. control plastic-free samples following the same lab process) and air control samples all evidenced minimal sample contamination (see SI, table S2).

Particles identified in surface water samples are representative of the MP quantity present at the respective time of sampling, while streambed sediments act as a sink or transient storage area for MPs and therefore, sediment samples represent an accumulation of particles over a longer time period (Liro *et al* 2020). A higher number of small MPs (10–64 μm, 26–601 items g^{-1}) were found in sediment samples as compared to large MPs (>64 μm 1–69 items g^{-1}) (table 1), figures 2(A) and (B) for the large and small MPs, respectively. The particle length and surface area of the >64 μm sample fraction in both sediment and water samples overlapped ranges in length and area of the MPs associated with WWTP effluent and sediments near the bypass pipe (table 1).

No trend between MP abundance in surface water and distance from the WWTP effluent was identified. The only relationship found between surface water MP properties and the observed hydrologic controls was a decrease in MP surface area with increased sub-reach-averaged water depth ($N = 6$, rho $= -0.89$, $p = 0.02$, $R^2 = 0.69$, $y = 16\,068\ln(x) + 58\,708$). The smaller size fraction of MPs (10–64 μm) was not analyzed in surface water samples since a 64 μm sieve was used in the field to facilitate the quick filtration of 100 l of surface water.

For MPs >64 μm, significant differences of particle length between different sub-reaches were identified by performing a Kruskal Wallis test ($p = 0.040$), but no differences were found between sub-reaches for any of the other MP properties investigated (i.e. abundance, area, and *L*/*A* coefficient). A low number of MPs were found UPSTREAM, with a large increase in MPs numbers near the BYPASS. A decrease in the number of larger MPs (>64 μm, items g^{-1}) with distance from the WWTP was observed (figure 2(A)). With inclusion of the BYPASS sediment samples, the statistical power of the relationship between MP abundance and distance increased ($y = -0.099x + 41.94$; $R^2 = 0.80$, $N = 7$, $p = 0.023$, figure 2(A)).

For the smaller sized MPs (10–64 μm), significant differences in particle length between sub-reaches were found according to a Kruskal Wallis test ($p = 0.027$), but no further statistically significant differences were identified between sub-reaches for other MP properties. However, MP abundance

Table 1. Particle number, length and surface area of large (>64 μm) and small MPs (10–64 μm) quantified in WWTP effluent (WWTP), surface water upstream/downstream (water up/down), sediment upstream, near the bypass pipe, and downstream (SED up, SED bypass, SED down). The data shown are medians [min–max].

	>64 μm MPs						10–64 μm MPs		
	Water up ($N = 1$)	Water WWTP($N = 1$)	Water down ($N = 6$)	SED up ($N = 2$)	SED bypass ($N = 3$)	SED down ($N = 30$)	SED up ($N = 2$)	SED bypass ($N = 3$)	SED down ($N = 30$)
Items: SED (#/g) or water (#/l)	0.24 [0–1]	1.68 [6–8]	3.27 [2–17]	11.81 [23–51]	45 [105–179]	8.94 [3–217]	55.49 [7–10]	176.25 [14–81]	84.86 [4–92]
Length (μm)	151 [131–177]	133 [68–292]	157 [64–730]	145 [75–647]	146 [64–963]	146 [64–1,790]	24 [11–49]	30 [11–64]	30 [10–64]
Area (μm^2)	11 13 140 [5 765–21 733]	9 290 [750–42 780]	13 364 [1 152–106 167]	9 822 [1 958–69 829]	10 758 [1 043–338 477]	11 731 [507–569 955]	481 [112–1 682]	636 [101–2 728]	661 [96–4 765]

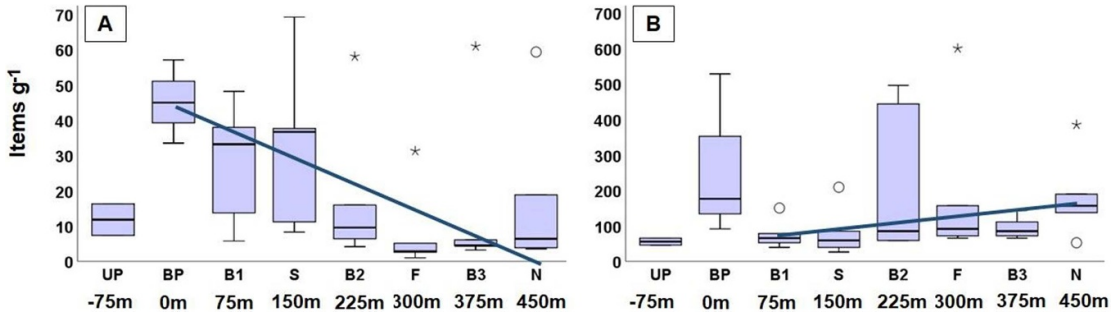

Figure 2. MP abundance within streambed sediment samples (items g^{-1}) for each sub-reach ($N = 35$, 5 per sub-reach) for (A) MPs >64 μm and (B) MPs 10–64 μm. The x-axis from left to right indicates the up to downstream gradient from the WWTP effluent input. The bold line inside the box indicates the median. Abbreviations: UP—upstream, BP—bypass, B1—buffer 1, S—slow, B2—buffer 2, F—fast, B3—buffer 3, N—naturalized, ◯—*outliers*, *—extreme *outliers*. The best fit line for items vs distance is shown in blue. Large MPs (A) $y = -0.099x + 41.94$; $R^2 = 0.80$, $N = 7$, $p = 0.023$ and small MPs (B) $y = 0.21x + 36.1$; $R^2 = 0.69$, $N = 6$, $p = 0.036$.

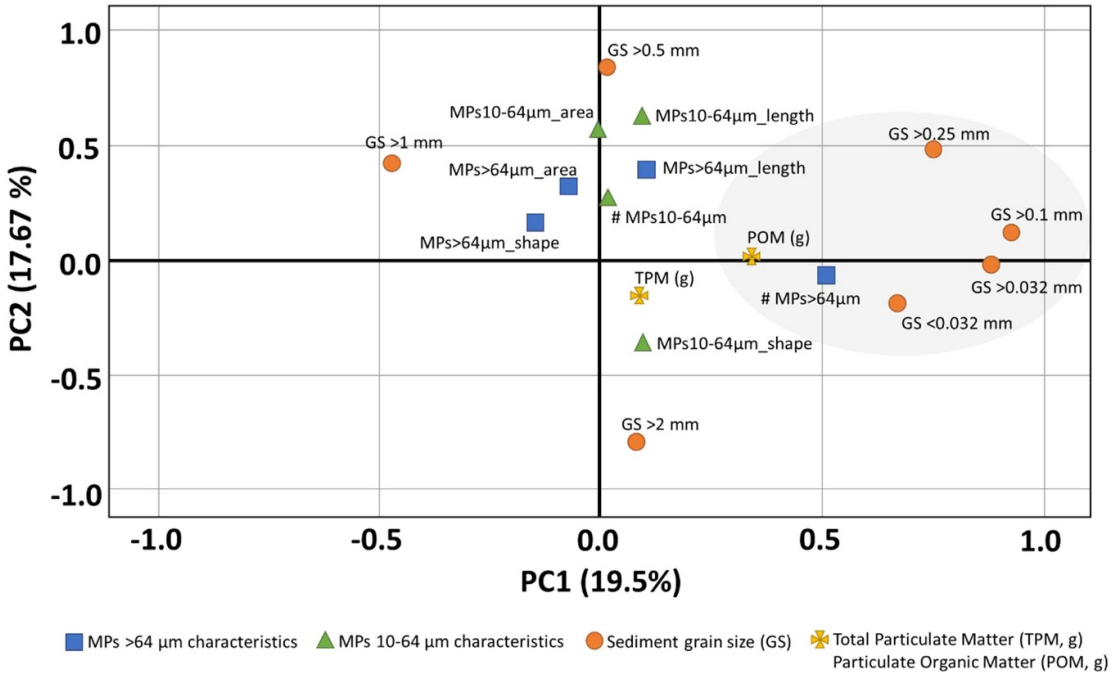

Figure 3. PCA results for the abundance of large (64 μm) and small (10–64 μm) MP particles, the particle length, surface area and shape coefficient (L/A) for MPs, organic matter (TPM, POM, g), and grain size classifications (mm). Total variance explained (%) is shown in the axis of the correspondent PC.

increased slightly with distance from the WWTP (figure 2(B)). Furthermore, the surface area of particles in the small MPs class decreased with downstream distance from the WWTP ($y = -1.0x + 914$; $R^2 = 0.68$; $N = 6$, $p = 0.005$). When the BYPASS location was included in the analysis of small MPs, no statistically significant trend for particle surface area or abundance with distance was identified.

3.2. Hydrologic influence on MP accumulation in sediments and relationship to other accumulated fine particles

Stream flow velocities at the field site differed by almost an order of magnitude in the downstream sub-reaches, with sub-reach-averaged maximum and minimum values of 0.13 ± 0.07 m s^{-1} (FAST) and 0.027 ± 0.01 m s^{-1} (BUFFER 3). Discharge (Q) ranged from 0.014 m^3 s^{-1} in BUFFER 3 to 0.060 $m^3 s^{-1}$ in FAST, representing the diversity of hydrologic conditions controlling particle transport and deposition. The number of large MP with >64 μm particle length as well as the length/surface area ratio (L/A) increased with increasing Q ($N = 6$, $p = 0.042$; $y = 133.17 \times 10^{2.26x}$; $R^2 = 0.31$ and $N = 6$, $p = 0.042$; $y = 0.024x^{0.045}$; $R^2 = 0.32$, respectively).

TPM (g) was similar between all sub-reaches (20.6 ± 0.22), but differences were observed between sub-reaches for POM (g; 0.30 ± 0.23). The highest value of POM was found in BUFFER 1 (0.58 ± 0.44) and the lowest value was observed in the FAST (0.20 ± 0.06). PCs 1 and 2 explained 19.5% and 17.7% of the total variance present within the dataset,

respectively (figure 3). PCA analysis indicates a low potential relationship between the smallest fraction of sediment (0.1–<0.032 mm), abundance of large MPs and POM (figure 3, gray shaded area).

4. Discussion and conclusions

4.1. Influence of WWTP continuous and infrequent inputs on the distribution of MPs in sediments

Our results clearly highlight the role of single-point inputs, such as WWTPs, to add plastic particles to the receiving stream, with MPs identified in all streambed sediment and water samples. At all sampling sites, small particles (10–64 μm) were found in a higher abundance than large particles (>64 μm), agreeing with recent work that demonstrate increased particle numbers to coincide with decreased MPs size in both marine and freshwater environments (Pabortsava and Lampitt 2020, Frei *et al* 2019). Small streams can lead to large contributions of plastic pollution (González-Fernández *et al* 2021), especially when they receive WWTP effluent (McCormick *et al* 2016). An unexpected result was the high abundance of MPs near to the bypass upstream of the WWTP effluent point source, which adds untreated wastewater inputs during infrequent storm events when the WWTP's capacity is exceeded. This bypass was found to constitute a substantial source of MPs to the receiving stream for both size fractions, even though storm events that exceed the capacity of the treatment plant and thus activated the bypass occur only on average 8–10 times per year (Pers. Comms Dr Francesc Sabater). The bypass was especially important for the addition of large MPs that may normally be removed within the WWTP filtration processes (Carr *et al* 2016), supported in our study by the improved correlation of large MPs with distance when the bypass site was included. WWTP mismanagement can also be a substantial contribution of macroplastics to streams, with estimates as high as 25 million MPs generated from fragmentation in the Besòs catchment, where our study stream is located (Schirinzi *et al* 2020). In contrast to most previous work that found mainly fragments and fibers as the most abundant shapes of MP in the natural environment (Hoellein *et al* 2017), 95% of MPs identified in this study represented fragments, while fibers represented less than 1% of the particles quantified. Possible explanations of the observed low fraction of fibers include their low density and slim shape that facilitate their transport downstream (Hoellein *et al* 2019), the high degradation rates of natural fibers (Stanton *et al* 2019), and the fact that polyamide or polyester fibers may not be detected using this method due to their lower fluorescence (Nel *et al* 2021).

Our analysis revealed the presence of a variety of polymers in the study reach, with PVC (53%) being the most abundant polymer type, followed by PVCA (29%), and PE and polyester (5%). Although our lab process used an extraction unit made of PVC, contamination in the blanks only had an average particle count per cm^2 of 5 $\pm$ 3 for large MPs and 9 $\pm$ 6 for small MPs, which was significantly lower than the counts in the environmental samples. In the natural environment, the most abundant MPs are PE, PP, polystyrene, and fibers, and WWTPs have been shown to be sources of all these polymer types and shapes (Rezania *et al* 2018, Gatidou *et al* 2019, Yang *et al* 2021). High-density MPs, such as PVC and PE terephthalate, can more easily be removed during WWTP treatment settling stages (Duis and Coors 2016). However, PVC is still commonly found in WWTP effluent possibly due to PVC-made pipes used in piping systems in some WWTPs and houses and external factors such as temporary building or industrial activities (Wagner and Reemtsma 2019, Tagg *et al* 2020). The excess flow during heavy rainfall episodes, which is expelled by the bypass pipe, could also help explain the presence of PVC in the streambed sediment since these infrequent releases to the stream provided opportunities to release larger and denser particles that would have otherwise been trapped by the WWTP. As a result of its high density, PVC would likely settle and accumulate within the streambed, while lighter particles are more likely to transport further downstream. In addition, other studies have detected dense polymers such as PVC and PVCA in sludge or biosolids (Crossman *et al* 2020, Okoffo *et al* 2020), which are applied to agriculture fields. MPs may then reenter freshwater bodies via runoff waters during irrigation and storm events (Henseler *et al* 2020).

4.2. Relationship of MP transport and retention compared to other fine particles

As several studies have demonstrated, fine particle dynamics play an important role in nutrient, carbon, and contaminant dynamics (Newbold *et al* 2005, Searcy *et al* 2006, Battin *et al* 2008, Bradford *et al* 2013, Harvey *et al* 2013, Grant *et al* 2014, Drummond *et al* 2014a). Fine particles are defined as particles <10 μm (colloids) or larger particles between 10 and 100 μm, whose deposition due to gravitational settling is negligible (Drummond *et al* 2014b). Fine particles include organic and inorganic matter, and microorganisms. Since MPs are part of the river environment and because of their characteristics (i.e. size, density) and their interaction with microorganisms (biofouling), they should be considered as part of the fine particles group. In fact, plastic is a novel form of allochthonous carbon (Romera-Castillo *et al* 2018, Hoellein *et al* 2019) and, because of its recalcitrant nature and chemical properties, can interact with aquatic organisms and influence ecosystem processes (Moore 2008, McCormick *et al* 2014). Therefore, it is essential to study the distribution of MPs, such as the hotspots of accumulation, and the effects that plastics

can have on environmental health to allow effective management and mitigation of MP pollution.

Interactions between fine particles, including MPs, and streambed sediments occur through a number of processes, including gravitational settling and hyporheic exchange (Drummond *et al* 2020). Cànoves is a turbulent stream, and therefore the rapid exchange from the surface water to the sediments via both turbulence and advective transport of solutes and fine particles is likely to be a dominant transport mechanism driving the MPs and other small and lower density particles into the sediment. However, a direct relationship was not found between velocity and MP accumulation downstream of the WWTP effluent. This result was surprising as a previous study in the same stream and study reach found an indirect relationship between velocity and the accumulation of fine POM (Meredith *et al* 2021). However, this may in part be explained by the difference in applied sampling methods. Meredith *et al* (2021) remobilized the sediment bed and then isolated the finer fraction of particles (<100 μm) and velocity was taken locally at each sampling location, whereas in our study the sediment cores included size fractions up to 2 mm and results were representative of the sub-reach and not unique to the sampling location. Also, while the size fractions of MPs can be classified, POM measured in this study was a bulk measurement encompassing all size fractions <2 mm. Since the larger size fraction (>1 mm) dominated the grain size distribution in our sediment cores, the transport and accumulation patterns of the finer POM could not be isolated and assessed separately within this study.

While large MPs decreased with distance from the WWTP effluent, the abundance of small MPs increased and the area of small MPs decreased. Particles quantified in the water represent a specific moment in time, while the particles found deposited in the sediments represent particle accumulation over time, from days to years. Therefore, the sediment results are representative of longer-term accumulation patterns of MPs downstream of the point source. Furthermore, although the statistical power was low, as Q increased, particles with longer lengths were found to accumulate within streambed sediments. These findings, all taken together, suggest a combination of preferential filtration of larger particles in the streambed sediments and/or fragmentation of particles as they transport downstream. For instance, smaller particles will be transported by water through the sediment pores and more easily participate in the two-way exchange between surface water and sediments, while larger particles will be preferentially filtered (Bradford *et al* 2006). Previous work by Simon *et al* (2018) and Mintenig *et al* 2017 found 19–447 #/l and up to 5×10^6 #/l of MPs in WWTP effluent, an increased number compared to ~2 #/l in the present study. The lower number in our work is likely due to the 64 μm sieve used in the field, whereas these studies measured down to a 10 μm particle size using μFTIR. We measured a comparable number of MPs accumulated in the streambed sediments downstream of a point source as the Roter Main river in Germany (Frei *et al* 2019), measuring ~300 #/g for MP particles (20–500 μm) compared to our range of 3–45 #/g, for >64 μm MPs and 55–176 #/g for 10–64 μm MPs. Other sampling downstream of WWTPs only measure the larger size fraction (>64 μm) and therefore are not directly comparable.

Transport and retention dynamics of MPs, similar to other fine particles, are influenced by streambed morphodynamics (Harvey *et al* 2012, Drummond *et al* 2014b, Phillips *et al* 2019) and biofilm presence at the sediment-water interface (Hoellein *et al* 2017). Studies on vertical hyporheic exchange fluxes around restoration cross vanes show that the hydraulic jump across the structures produces high downwelling fluxes around these structures, creating hotspots of hyporheic exchange (Drummond *et al* 2018). The river's morphology, and specifically the local differences in geomorphic conditions, will therefore determine the frequency in which hyporheic exchange processes happen and in turn influence the accumulation patterns. This is likely to explain why a higher variability of the number of large particles were observed in BUFFER 1 and SLOW subreaches, and an increase in particle counts in the NATURALIZED sub-reach with increased heterogeneity. Samples with higher abundance of fine sand or silt (Wentworth 1922) also retained larger MPs (>64 μm) and POM, suggesting streambed grain size is a key factor for MP accumulation in streambeds. Waldschläger and Schüttrumpf (2019) determined that both sediment grain size and particle characteristics (density, size, and shape) have a strong effect on the remobilization of MPs, with higher transport rates than natural sediments. The finding between fine sediments and presence of MPs can be especially important for biodegradable plastics, where enhanced degradation rates can occur in fine grain sizes such as mud and medium-fine sand (Lott *et al* 2020).

4.3. Conclusions

We demonstrate a clear longitudinal trend in MPs pollution downstream of a WWTP dependent on particle size fraction. Accumulation patterns of MPs downstream of a single-point input showed opposite trends for MPs classified as small (10–64 μm) and large (>64 μm), increasing and decreasing their abundance in sediments with increased distance from the MPs source. Presence of fine particles was related with higher abundance of large MPs (>64 μm), suggesting that hotspots of fine particle accumulation may act as a trap for MP particles. Small MPs surface area also decreased with distance, showing that processes such as filtration, particle trapping in the

streambed sediments, and fragmentation of larger particles are playing an important role in this stream.

Finally, our results demonstrate the infrequent inputs of untreated water during storm events via the bypass was especially important to the addition of large MPs. The combination of classifying MPs by particle size and measuring other fine particles commonly found in streams, provided additional insight into the transport dynamics of MPs from a point source that led to distinct accumulation patterns. It is essential to study the distribution pattern of MPs to understand hotspots of accumulation that influence environmental health, and to improve predictions of MP fate in freshwaters and mitigate MPs pollution. Our work provides insight into how hydrodynamics influence MP, and other fine particle, transport in streams, which will be especially important to consider under future environmental scenarios, with both expected increases in precipitation and MP production and pollution.

Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

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Author contributions

The study was conceptualized and designed by J D D., H N D, H M, and F S Field samples were collected by J D D, H N D, H M, S H S, and F S Laboratory analyses were carried out by J D D, H N D, and H M Statistical analyses were carried out by J D D and H M. All authors contributed to the development and writing of the manuscript.

Conflict of interest

The authors declare that they have no conflict of interest.

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