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Distribution and transport of microplastic and fine particulate organic matter in urban streams

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Abstract. Plastic litter is accumulating in ecosystems worldwide. Rivers are a major source of plastic litter to oceans. However, rivers also retain and transform plastic pollution. While methods for calculating particle transport dynamics in rivers are well established, they are infrequently used to quantify the transport and retention of microplastics (i.e., particles < 5 mm) in flowing waters. Measurements of microplastic movement in rivers are needed for a greater understanding of the fate of plastic litter at watershed and global scales, and to inform pollution prevention strategies. Our objectives were to (1) quantify the abundance of microplastics within different river habitats and (2) adapt organic matter "spiraling" metrics to measure microplastic transport concurrent with fine particulate organic matter (FPOM). We quantified microplastic and FPOM abundance across urban river habitats (i.e., surface water, water column, benthos), and calculated downstream particle velocity, index of retention, turnover rate, and spiraling length for both particle types. Microplastic standing stock was assessed using a habitat-specific approach, and estimates were scaled up to encompass the study reach. Spatial distribution of particles demonstrated that microplastics and FPOM were retained together, likely by hydrodynamic forces that facilitate particle sinking or resuspension. Microplastic particles had a higher downstream particle velocity and lower index of retention relative to FPOM, suggesting that microplastics were retained to a lesser degree than FPOM in the study reaches. Microplastics also showed lower turnover rates and longer spiraling lengths relative to FPOM, attributed to the slow rates of plastic degradation. Thus, rivers are less retentive of microplastics than FPOM, although both particles are retained in similar locations. Because microplastics are resistant to degradation, individual particles can be transported longer distances prior to mineralization than FPOM, making it likely that microplastic particles will encounter larger bodies of water and interact with various aquatic biota in the process. These empirical assessments of particle transport will be valuable for understanding the fate and transformation of microplastic particles in freshwater resources and ultimately contribute to the refinement of global plastic budgets.

Key words: *freshwater; microplastics; organic matter; pollution; retention; spiraling.*

Introduction

The production and disposal of plastic has accelerated since its industrialization in the mid-1900s (Geyer et al. 2017). A growing body of research shows plastic litter is a ubiquitous and pervasive environmental contaminant at a global scale (Barnes et al. 2009, Geyer et al. 2017). Studies documenting the sources, abundance, distribution, and biological impacts of plastic in marine ecosystems have dominated the literature for the last several decades (Thiel et al. 2003, Jambeck et al. 2015, Bergmann et al. 2017). In recent years, research has expanded to include measurements of plastic pollution to across a diversity of habitats (e.g., deep sea, rivers, atmosphere) and particle sizes (nano- and microplastics; Baldwin et al. 2016, Dris et al. 2016, Courtene-Jones et al. 2019). An array of empirical assessments of plastic sinks, sources, and transport mechanisms will be needed to generate a global plastic budget (Jambeck et al. 2015).

Microplastics (i.e., particles <5 mm) are a focus of plastic pollution research. Sources of microplastic pollution include fibers from synthetic textiles, fragments from the breakdown of larger plastic items, plastic particles from personal care products, and industrial pellets (i.e., "nurdles") from commercial manufacturing (Fendall and Sewell 2009, Andrady 2011, Browne et al. 2011). Particles are introduced to aquatic ecosystems via wastewater effluent, urban and agricultural runoff, combined sewer outflows, and atmospheric deposition (Barnes et al. 2009, Dris et al. 2016). Microplastics can

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interact with aquatic biota via ingestion of particles, leaching of chemical additives, transfer of persistent organic pollutants (POPs) into food webs, and the use of microplastic particles as a growth substrate for biofilms (Rochman et al. 2013, Wright et al. 2013, McCormick and Hoellein 2016).

Once considered simple conduits of microplastic from terrestrial sources to oceans (Jambeck et al. 2015), recent research demonstrates that rivers serve an active role in retaining and transforming (i.e., biofilm colonization, abiotic, and biotic breakdown) microplastics prior to export downstream (Browne et al. 2011, Hoellein et al. 2017). Documenting the dynamic nature of microplastic transport within river ecosystems is critical to inform global plastic budgets. In addition, retention and export of microplastics from rivers will drive the form, timing, and quantity of particles that enters oceans (Baldwin et al. 2016). Quantifying the transport of microplastics in flowing waters requires direct assessments of input, transport, and deposition in streams (Hoellein et al. 2019).

The movement of particles in rivers has long been studied for naturally occurring materials such as wood, leaf litter, fine sediment, and sand (Webster et al. 1999). "Spiraling" metrics are a group of calculations that quantify particle movement. By combining measurements of particle abundance, microbial respiration rates, and stream physical characteristics, particle spiraling metrics represent a mathematical approach for describing particle transport and biological turnover (Newbold et al. 1982, Ensign and Doyle 2006, Griffiths et al. 2012). Spiraling measurements have been applied to study the movement of naturally occurring particles in streams and provide critical insights into nutrient cycles, food webs, and energy flow (Newbold et al. 1982, Minshall et al. 1992, Webster and Meyer 1997, Griffiths et al. 2012), but have yet to be established as a method for assessing microplastic transport in freshwater rivers and streams.

The analytical framework of spiraling metrics presents a useful paradigm for exploring the transport of microplastics, a novel and highly recalcitrant form of allochthonous carbon in streams. Hoellein et al. (2019) used the spiraling framework to describe patterns in microplastic deposition in artificial streams, showing that plastic particles follow similar depositional patterns as other allochthonous particles, and deposition rates can be predicted to some extent based on particle properties such as size, density, and colonization by biofilms. This set of measurements has not previously been used to measure microplastic movement in situ, and the transport rates of microplastics has yet to be directly compared to the movement of fine particulate organic matter (FPOM) in streams.

The objectives of this study were to (1) quantify the distribution of microplastics in urban river habitats (i.e., surface water, water column, benthos) and (2) adapt nutrient spiraling metrics to describe microplastic transport concurrent with FPOM in urban streams. With respect to microplastic distribution, we expected plastic particles to be present throughout the study streams, with the highest concentration in the benthos. We hypothesized that the retentive capacity of benthic habitats would differ based on physical characteristics of the substrate (e.g., depositional vs. erosional habitats). With respect to particle transport, we predicted that downstream velocity of microplastics and FPOM would be similar. We hypothesized that microplastics would have a slower turnover rate than FPOM, and therefore, be transported father downstream than FPOM before mineralization.

Methods

Study sites

We collected data at three urban streams in the Chicago metropolitan region: North Branch Chicago River (Chicago, Illinois, USA), Higgins Creek (Des Plaines, Illinois, USA), and Springbrook Creek (Wheaton, Illinois, USA). Collection sites at Higgins Creek and Springbrook Creek were ~100 and ~300 m downstream of wastewater treatment plant outflows, respectively, a documented point-source of microplastics (McCormick et al. 2016). Sampling was conducted at each site in fall (November–December 2017), spring/early summer (May–July 2018), and late summer (August–September 2018). We conducted measurements three times to capture variability in seasonal factors that affect particle transport, including discharge, biofilm and macrophyte growth, and leaf litter inputs. On each date, we recorded stream depth and water velocity (m/s; Marsh-McBirney Flo-Mate 2000, Loveland, Colorado, USA) on 0.5–1.0 m increments to calculate discharge (m^3/s; Appendix S1: Table S1). We established one 100 m long study reach in each stream. Microplastic and organic matter samples were collected from three zones within each transect: upstream, mid-transect (~50 m), and downstream.

Microplastic collection: water and benthic substrates

Measuring particle transport dynamics requires collection of particles in the stream surface water (i.e., floating), water column, and benthic zone. We collected surface water samples using a drift net and water column samples via a 1–2 L glass bottle following previously published analyses (McCormick et al. 2016, Barrows et al. 2017, McNeish et al. 2018). Drift nets were deployed partially submerged (~22 cm) in water for 20–25 minutes (363 μm Nitex mesh). Depth, velocity (m/s; Marsh-McBirney Flo-Mate model 2000 Portable Flowmeter, Loveland, Colorado, USA), and net width were measured to determine the volume of water captured by each net. We transferred all material caught in the net to a pre-cleaned glass jar for storage (0.7-L Ball mason jar, Newell Brands, Hoboken, New Jersey, USA). We used

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squirt bottles to rinse trapped items from the sides of the net into the cod end, which was then detached, and material was transferred into the glass sample jar with rinsing and pre-rinsed forceps where needed. All rinsing was directed from the outside to avoid contamination. Water column samples were collected in the center of the stream using precleaned 1–2 L glass media bottles. We opened and capped sample bottles under the surface of the water to prevent atmospheric contamination. All samples were refrigerated until processing. Finally, three field controls were collected per sampling event to account for atmospheric contamination. To do so, a glass storage jar was opened next to the sample jar for the same duration the net samples were transferred into storage jars, and then closed at the same time. Prior to sampling, all glass jars and media bottles were rinsed three times with deionized (DI) water (previously filtered through 0.363-μm mesh), dried upside down to prevent atmospheric contamination, and capped for storage. In fall 2017, we collected 12 surface and 9 water column samples in each stream reach. Due to uniformity of microplastic abundance among replicate samples in fall 2017, we reduced the sample number for surface water and water column habitats in spring and summer 2018 ($N = 6$).

We collected benthic samples for microplastics in each of the habitat types in the study streams. First, we conducted a benthic survey to estimate relative coverage of habitat types in each 100-m reach on each sampling date. To do so, we set up 8–10 transects within the 100-m reach and recorded water depth and habitat type at 0.2–0.5 m increments, adjusted according to temporal variation in stream width and depth (Hoellein et al. 2009). Habitats recorded were sand, gravel, cobble, concrete, coarse benthic organic matter (CBOM), fine benthic organic matter (FBOM), and filamentous algae. Not all habitats were found at each site or during all seasons. We used a meter stick to determine differences between cobble and gravel where the substrata was classified substrate as cobble if the substrate remained stationary when pressed with the meter stick and as gravel if the rocky substrate moved easily with pressure from the meter stick. After completing the benthic survey, we collected triplicate samples of each substrate type for microplastic measurements. Habitats were sampled by inserting open specimen containers ~6 cm into the substrate, inverting, and capping immediately underwater (Hoellein et al. 2009). Samples were refrigerated until processing. Higgins Creek was the only site with a portion of the benthic zone as concrete. We could not collect a concrete substrate sample, so we used measurements of microplastic abundance from cobble as a surrogate for the concrete section of the reach.

Microplastic processing

We processed microplastics from water column, surface water, and benthic habitats in the laboratory according to previously published methods (McCormick et al. 2016, Barrows et al. 2018, McNeish et al. 2018). Processing procedures differed among sample types due to challenges in isolating microplastics from the matrix of materials that varies among habitats. The water column samples had the simplest procedure. Bottles were shaken to suspend any settled material, and 1 L was filtered (0.45-μm filters; Whatman, Pittsburgh, Pennsylvania, USA) for later quantification of microplastics. Surface water samples were transferred from glass jars onto stacked 4.75- and 0.3-mm sieves and rinsed at least three times with filtered RO water. Material on the 0.3-mm sieve was transferred back to the Mason jar, while material <0.3 mm and >4.75 mm was discarded. For benthic habitats, specimen containers with sand, gravel, FBOM, CBOM, and filamentous algae (i.e., all substrates except cobble) were shaken to homogenize the sample contents. A 70-mL subsample was taken from sand and gravel, and a 45 mL subsample was taken from FPOM samples for processing. This lower volume for FBOM was determined via pilot studies and was necessary for organic matter digestion and filtration processing. Material from benthic samples were rinsed over stacked sieves of 4.75, 1.0, and 0.3 mm. Material between 0.3 and 1 mm sieves was retained for analysis while the remainder was discarded. This was necessary due to large amounts of sand and inorganic material >1 mm that impeded sample digestion, filtration, and microplastic quantification. For cobble, individual rocks were transferred directly to a glass jar for drying and later processing with peroxide. We did not attempt to remove periphyton and attached particles with scrubbing to avoid contamination. Overall, we acknowledge differences in the size fractionation among the sample types, which we accounted for via careful measurements of particle dimensions (see Appendix S1).

Samples placed in glass jars from the net and benthic habitats were subject to further processing. Each was dried for 3 d at 70°C. Then, we added 30% H_2O_2 and 20 mL of 0.05 mol/L Fe (II) solution at 75°C to oxidize organic material (Masura et al. 2015). We added H_2O_2 in 20-mL increments until all organic material was digested or the sample failed to react following peroxide addition. Total volume of the digestion solution did not exceed 200 mL. Following the addition of peroxide to the cobble samples, cobbles were removed from the beaker using metal forceps and rinsed with filtered RO water to remove any microplastic particles on the substrate. Following the peroxide digestion, the material remaining in the surface water samples, as well as the benthic habitats of filamentous algae, CBOM, and cobble samples were filtered and dried in the same fashion as the water column samples described above. However, sand, gravel, and FPOM samples required density separation. Samples were supersaturated with NaCl, transferred to glass funnels, and left to settle for ~24 h. Liquid and floating particles, including microplastics were rinsed onto filters from the glass funnels (Masura et al. 2015).

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The final steps in sample analysis were filtration and counting. All samples were filtered onto gridded 0.45-μm filters (Whatman), placed in 20-mL aluminum weighing dishes, and covered with foil. Filters were dried at 70°C for 24 h. Each filter was assessed under a dissecting microscope (25× magnification) by two or three different researchers. Counts differing by a value of ≤2 particles were considered complete, but if there was a discrepancy >2, the filter was assessed by a third individual. Microplastics were classified by shape (i.e., fiber, fragment, pellet [bead], foam, and film), color, and length (Free et al. 2014, McCormick et al. 2014, Masura et al. 2015).

We randomly selected microplastic particles representing all study sites, sampling seasons, and sample types for particle identification analyses. Particles for analysis were selected at random by removing the first fiber identified on each filter. We did not control for particle color. Fibers identified represent ~7% of total fibers counted ($N = 85$). Particle identification was carried out by MicroVision Laboratories (Chelmsford, Massachusetts, USA) using Fourier Transform-Infrared Spectroscopy (FTIR) analysis. The Bruker LUMOS FTIR instrument assigns polymer types to unknown sample particles using reference spectra ranging from 7,000 to 600 cm^{-1} in the Bruker library. Particles were scanned at a spectral resolution of 4 cm^{-1} using OPUS software (Barrows et al. 2018).

Microplastic counts: contamination correction and in situ scaling

All sample counts were corrected for contamination. In addition to field controls collected during sampling, we completed 10 filter controls and 10 digestion controls per season to account for contamination in water column samples, and net and sediment samples, respectively, introduced during laboratory processing. Field control jars were rinsed with filtered RO water and poured directly onto a gridded filter. Bulk filter controls were prepared by placing a gridded microfiber filter on a filter tower and rinsing with RO water. Digestion controls consisted of 20 mL of H_2O_2 and 20 mL of 0.05 mol/L Fe (II) solution brought to 75°C alone in a Mason sample jar and rinsed with RO water onto a filter. As with environmental samples, all controls were filtered onto 0.45-μm filters and dried prior to counting. We documented only fibers as contaminants and found no field contamination. Thus, fiber counts were corrected using the average number of fibers on the filter controls (water column samples), and the digestion controls (surface water and benthic substrate samples), each generated on a seasonal basis. Water column filters were corrected by subtracting 0.90, 1.33, and 0.35 particles/sample from fall, spring, and summer, respectively. Mean contamination in surface water and benthic habitat samples was 2.82, 2.75, and 0.40 particles/sample in fall, spring, and summer, respectively. Together, field and laboratory controls account for ~12% of total filters assessed for microplastic.

We expressed particle density according to water volume and stream surface area. We calculated microplastic concentration in the surface water (no./m^3) by dividing the total number of microplastic particles by the volume of water that passed through each net. Water column concentrations were measured as number of particles per liter (L) of water filtered and converted to number per cubic meter (no./m^3). Microplastic standing stock (no./m^2) was determined for each benthic habitat. To estimate the total standing stock of microplastic (no./m^2) in each reach, we scaled up the benthic density (no./m^2) for each habitat in the 100-m stream reach according to the relative coverage of each habitat type (m^2). Areal coverage of each habitat was determined by multiplying the relative coverage of a habitat (i.e., from the benthic survey) by total area of the reach (Hoellein et al. 2009). Finally, we calculated a "hot spot index" as the proportion of total microplastics found in a habitat, divided by the habitat's relative coverage in the 100-m reach. A value >1 indicates a high concentration of microplastics relative to benthic coverage, where a value <1 indicates low proportion of microplastics relative to benthic coverage (Hoellein et al. 2009, Griffiths et al. 2012).

We acknowledge some differences in sample processing among habitat types (i.e., water column, net, and the various benthic habitat samples) due to the non-trivial challenges of collecting data across the wide variety of conditions in each habitat. First, there were differences in size fractionation between water column, net, and sediment samples. To address this, we measured the length of fibers and fragments in water column, net, and benthic habitats (Table 1). Second, we acknowledge that the density of NaCl (1.2 g/cm^3) may not suspend some microplastic particles of dense polymer types, so our sediment standing stocks represent conservative measurements. We note this is the first study to measure microplastics from all habitats in a stream reach. Inevitably, new combinations of analyses such as this will reveal important opportunities to refine approaches in sample collection and preparation in follow up studies, especially in a rapidly developing field of research. We offer detailed suggestions below (see *Discussion*).

Organic matter collection and processing

In addition to microplastic analyses, we collected FPOM samples for calculating spiraling metrics. Seston samples were collected in 1–2 L acid-washed glass bottles following the same methods and at the same time as samples for water column microplastics. Bottles were refrigerated until processing, which occurred within 7 d). In the lab, we shook the bottles to suspend settled material, filtered 1 L from each sample onto pre-ashed (i.e., 550°C for 1 h), and weighed glass fiber filters (0.7 μm, Merck Millipore). We placed filters into pre-ashed and weighed 20-mL aluminum pans (Fisherbrand, Waltham,

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TABLE 1. Comparison of method details and particle characteristics across habitats, according to subsample volume (mL) (if applicable), sieve limits (mm), use of digestion or density separation, and mean (±SE) fiber and fragment length (mm).

Sample type	Subsample (mL)	Size range (mm)	Digestion	Salinity separation	Fiber length (mm)	Fragment length (mm)
Surface water	N/A	0.3–4.75	yes	no	1.81 ± 0.09	0.98 ± 0.09
Water column	1,000	~0.05–5	no	no	1.21 ± 0.10	0.84
Sand	70	0.3–1	yes	yes		
Gravel	70	0.3–1	yes	yes		
FBOM	45	0.3–1	yes	yes	1.59 ± 0.11	0.78 ± 0.13
CBOM	N/A	0.3–1	yes	no		
Filamentous algae	N/A	0.3–1	yes	no		
Cobble	N/A	~0.05–5	yes	no	1.13 ± 0.11	N/A

Notes: Fiber length of sediment samples measured using sand, gravel, fine benthic organic matter (FBOM), coarse benthic organic matter (CBOM), and filamentous algae were consolidated. No fragments were measured in cobble samples. The fragment length of 0.84 applies to the single fragment counted in the water column samples. Volume of water captured by nets ranged 4.5–153.1 m^3. Subsample volumes are noted as N/A when whole samples (i.e., not a subsample) were processed.

Massachusetts, USA), dried the samples at 70°C, and measured dry mass (DM). Pans were combusted at 550°C for 3 h and cooled in a desiccator overnight before calculating ash-free dry mass (AFDM).

We used a benthic core to collect samples of fine benthic organic matter (FBOM) to measure AFDM and respiration. Four sampling locations were chosen randomly throughout each 100-m reach. The core (0.13 m^2 at North Branch and Higgins Creek, 0.06 m^2 at Salt Creek; adjusted according to stream depth) was inserted ~10 cm into the sediment (Cook and Hoellein 2016). At locations with a concrete benthic surface in Higgins Creek, one researcher held the core tightly in place by pushing down on the rim of the core while another researcher conducted sample and data collection. First, we measured the water depth in the core ($N = 5$) to calculate water volume (Entrekin et al. 2008). Next, we manually agitated the substrate to suspend organic matter. All coarse benthic organic matter (CBOM) was removed using a 4.75 mm sieve. Then, we collected FBOM samples by filling 150-mL specimen containers in the core and capping containers underwater. We collected four FPOM samples to measure AFDM, and four samples to measure respiration (Entrekin et al. 2008).

We measured respiration by incubating FPOM samples in situ. "Controls" (i.e., water only; $N = 3$) and specimen containers with FPOM ($N = 4$) were incubated in the stream in the dark (i.e., covered by black plastic bag) to maintain a consistent temperature and prevent exposure to sunlight (Hoellein et al. 2009). We measured dissolved oxygen (DO; HQ40d portable meter, Hach, Loveland, Colorado, USA) before and after 3 h of incubation. In the laboratory, we measured AFDM of FBOM. Samples were placed in pre-ashed and weighed tins, and then dry mass and AFDM were measured as described above.

Spiraling calculations

We adapted organic matter spiraling equations to quantify movement of microplastics and FPOM (Newbold et al. 1982, Minshall et al. 1992, Griffiths et al. 2012). We used microplastic and suspended sediment concentrations from the surface water, water column, and benthic habitats, as well as measurements of stream discharge (Q), width (w), depth (z), and water velocity (V_{water}) to calculate transport and retention metrics. Downstream particle velocity (Eq. 1; $V_{particle}$), index of retention (Eq. 2; $IR_{particle}$), and spiraling length (Eq. 4; $S_{particle}$) were calculated for both particle types. We measured the FPOM turnover rate (Eq. 3; k_{FPOM}) via respiration (Entrekin et al. 2008, Griffiths et al. 2012). We used a published value for the turnover rate of polyethylene to estimate plastic turnover rate ($k_{plasttic}$; Pritchard 1998, Gewert et al. 2015). We recognize the limitations of using a published value for $k_{plastic}$ and acknowledge that the turnover of plastic is likely to vary by polymer type and environmental conditions

$$V_{particle}(\text{mm/s}) = \frac{\text{WC} \times \text{Q}}{\text{BC} \times \text{w}} \quad (1)$$

$$\text{IR}_{particle} = \frac{V_{water}}{V} \quad (2)$$

$$k_{FPOM}(\text{d}^{-1}) = \frac{R}{\text{BC} + (\text{WC} \times \text{z})} \quad (3)$$

$$S_{particle}(\text{km}) = \frac{V}{k} \quad (4)$$

In the equations above, WC is the sum of the surface water and water column microplastic concentrations, BC is microplastic standing stock for the total reach, and R is respiration. Particle parameters are hereafter defined by particle type where appropriate (e.g., V_{FPOM}, $V_{plastic}$).

Data analysis

We used one-way ANOVA to compare microplastic concentration (no./m^3), suspended sediment, and

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benthic FBOM among fall, spring, and summer in the surface water and water column for each study stream. We compared benthic microplastic concentrations (no./m^2) among habitats (e.g., sand, cobble, algae) and seasons using two-way ANOVAs for each stream. Last, we compared mean hot spot indices among habitats using a one-way ANOVA. Following a significant P value ($\leq$0.05), we used Tukey's multiple comparison test to identify differences among seasons or habitats. All data were assessed to meet the assumptions of ANOVA and regression. All statistical analyses were completed using SYSTAT 13 (SYSTAT Software, Cranes Software International, Chicago, Illinois, USA).

Results

Surface water and water column habitats

Microplastic concentrations in the surface water and water column habitats were analyzed for each site individually. Across all sites, particle concentration was consistently higher in the water column than the surface water (Fig. 1). There was no consistent temporal pattern in microplastic abundance. Surface water concentration was highest in Higgins Creek in spring ($P \leq 0.001$) and in the North Branch in summer ($P = 0.004$; Fig. 1A, B). In the water column, particle concentration was significantly higher in fall at 7732.2 no./m^3 compared to spring and summer (555.6 and 1825.0 no./m^3, respectively) in the North Branch ($P = 0.002$; Fig. 1E), while Higgins and Springbrook Creeks showed no differences among dates (Fig. 1D, F).

Microplastics in the benthic zone

We also examined microplastic patterns in the benthic habitats for each stream individually. At Higgins Creek, microplastic standing stock varied among habitats (two-way ANOVA $P = 0.007$), but not by season (two-way ANOVA $P = 0.356$; Table 2). Gravel and filamentous algae at Higgins Creek had significantly more microplastics than other benthic habitats (Fig. 2A), and filamentous algae and concrete covered ~85% of the benthic surface across all seasons (Fig. 2B). The hot spot index showed gravel retained a large proportion of microplastics relative to its low coverage within the reach, while concrete, sand, and cobble retained little plastics relative to benthic coverage (Fig. 2C). Total microplastics at Higgins Creek (No./reach) was lowest in summer (~250,000 pieces) and highest in fall (~480,000 pieces), when filamentous algae was also highest (Fig. 2D).

Patterns of microplastics in benthic habitats of the North Branch were different than Higgins Creek, largely due to the lack of a concrete channel. Microplastic standing stock in the North Branch varied by benthic habitat (two-way ANOVA Habitat $P = 0.025$) but not season (two-way ANOVA Season $P = 0.957$; Table 2). FBOM had a significantly higher microplastic concentration relative to cobble and CBOM, while sand and gravel were intermediate (Fig. 3A). Benthic coverage in the North Branch was dominated by gravel (~50%), followed by cobble, sand, and FBOM (Fig. 3B). While the substrate hot spot indices were not significantly different among habitat types, we noted a trend of lower retention in gravel and higher retention in FBOM (Fig. 3C). Total microplastics in the 100-m reach of the North Branch ranged from ~600,000 pieces in fall to ~1 million particles in summer (Fig. 3D).

Benthic habitat patterns at Springbrook Creek were similar to the North Branch. Microplastic standing stock varied significantly by season (two-way ANOVA, season $P = 0.037$; Table 2) and habitat (two-way ANOVA, habitat $P \leq 0.001$; Table 2). CBOM had significantly higher microplastic standing stock than gravel, cobble, and filamentous algae, while sand and FBOM were intermediate. (Fig. 4A). Gravel and cobble were the most common substrates in Springbrook Creek, covering >60% of the benthic surface (Fig. 4B). Hot spot indices demonstrate that sand, FBOM, and CBOM had a significantly higher proportion of microplastics relative to their small distribution within the study reach, while cobble retained little plastic (Fig. 4C). Total microplastics ranged from ~130,000 pieces to ~250,000 pieces in the 100-m reach (Fig. 4D).

Last, we considered the hot spot index for each substrate with all of the collection dates as replicates. The mean hot spot index varied significantly by habitat (one-way ANOVA $P = 0.025$; Fig. 5). FBOM had significantly more retentive capacity for microplastics than cobble, which was the least retentive of microplastics relative to surface area (−0.90). This pattern followed our expectations for microplastic deposition in depositional (i.e., FBOM) and erosional (i.e., cobble) habitats. Future studies may benefit from an assessment of pore size to determine the relationship with microplastic.

Relative abundance by shape and fiber color

Microplastic composition by shape was dominated by fibers and fragments across the three sites. At the North Branch, fibers and fragments accounted for ~79% and ~21% of the particles collected, respectively. Pellets were rare (<1%), and foam and film were absent in all seasons (Appendix S1: Fig. S1B). Higgins Creek and Springbrook Creek, which are both downstream of wastewater treatment facilities, had similar compositions. At Higgins and Springbrook Creeks, fibers were most common (~68% and ~51%, respectively), followed by fragments (~28% and ~39%, respectively), and finally pellets (~3% and ~10%, respectively). Foam and film were rare at both sites, representing <1% of all microplastics collected across all seasons (Appendix S1: Fig. S1A, C).

Relative abundance of fiber colors was similar among samples. Blue, clear, red, and black fibers account for ~85–89% of all fibers across collection sites and controls. However, blue and clear fibers alone represented ~42–58% of fibers in environmental samples and were more

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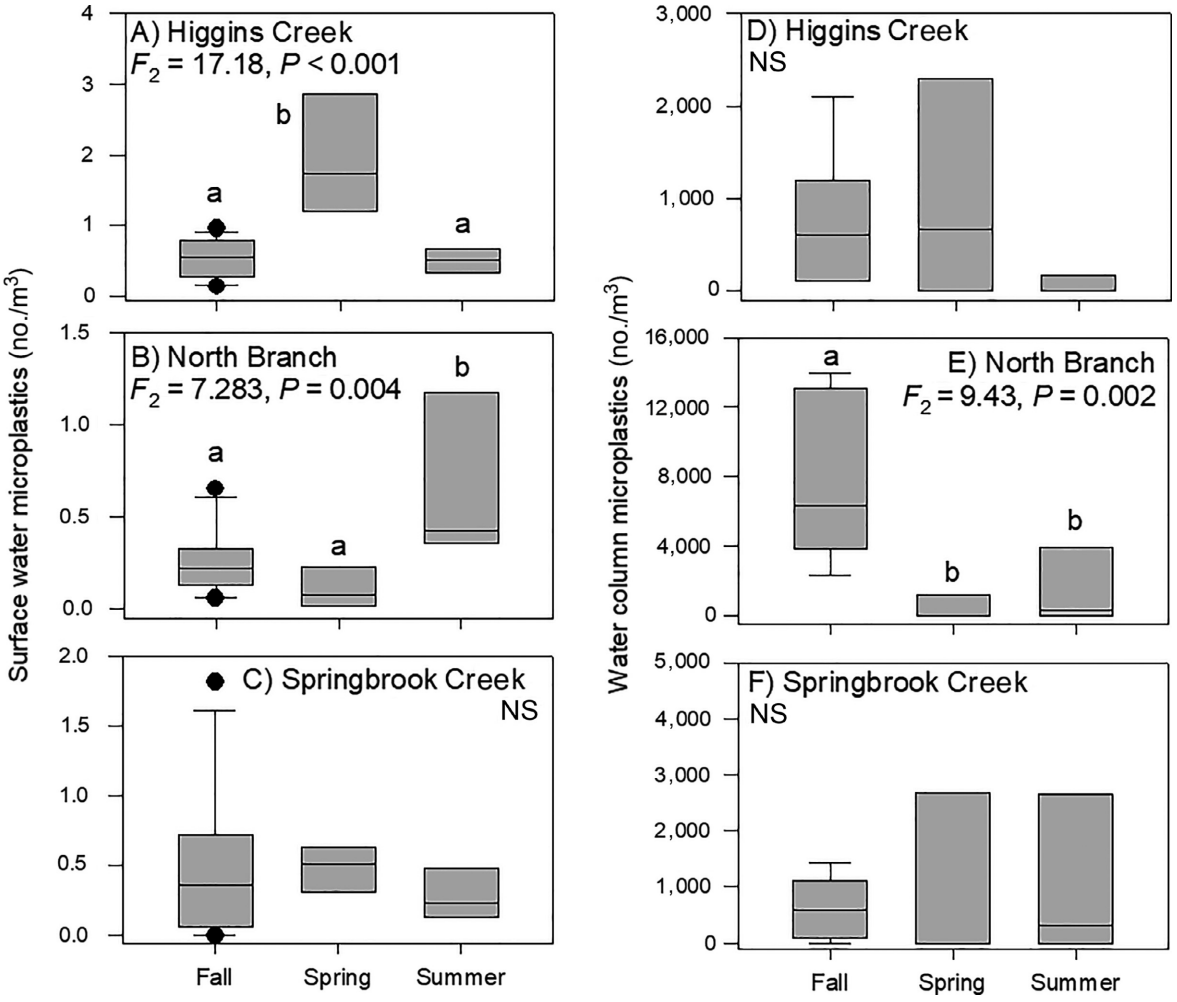

FIG. 1. Median (center line), 25th and 75th percentile (box edges), and data outliers beyond 25 and 75 percentiles (dots) for microplastic concentration in the (A–C) surface water and (D–F) water column habitats at Higgins Creek (A, D), North Branch (B, E), and Springbrook Creek (C, F) during three seasons. *F* statistics and *P* values are from one-way ANOVA among seasons. F-statistic subscript represents degrees of freedom for the numerator. Denominator degrees of freedom is 18 for all. Different letters indicate significant differences from Tukey's test results. NS, not significant.

TABLE 2. Results from two-way ANOVA comparing microplastic standing stock (no./m^2) among seasons and benthic habitats at each of the three study sites.

	Season		Habitat		Interaction	
Site	*F*	*P*	*F*	*P*	*F*	*P*
Higgins Creek	1.06	0.356	4.00	**0.007**	0.82	0.589
North Branch	0.04	0.957	3.11	**0.025**	1.53	0.176
Springbrook Creek	3.54	**0.037**	6.83	**≤0.001**	1.38	0.218

Notes: Significant *P* values are in boldface type. Degrees of freedom for season, habitat, and interaction at Higgins Creek and North Branch are df = 2, df = 4, and df = 8, respectively. Springbrook Creek degrees of freedom for season, habitat and interaction are df = 2, df = 5, and df = 10, respectively (the difference is due to the additional habitat of filamentous algae at this site). Denominator degrees of freedom for all *F* ratios is 47 at Higgins Creek, 42 at North Branch, and 46 at Springbrook Creek.

abundant (~75%) in controls. Gray, striped (i.e., blue/black and clear), purple, and other colors (e.g., red and clear, pink, yellow, orange, and neon green) were rare across all samples (Appendix S1: Fig. S2).

Microplastic length and particle identification

Particle size measurements showed overlap in length (mm) across each of the methods used to isolate microplastics from the different riverine habitats. Mean fiber length ranged from 1.13 to 1.81 mm (Appendix S1: Fig. S3). Mean fragment length ranged from 0.78 to 0.98 mm (Table 1). Results support direct comparisons of measurements across habitats, despite the variation in sampling and processing methods.

The chemical composition of particles varied across sample types. Of the particles selected for analysis, 85

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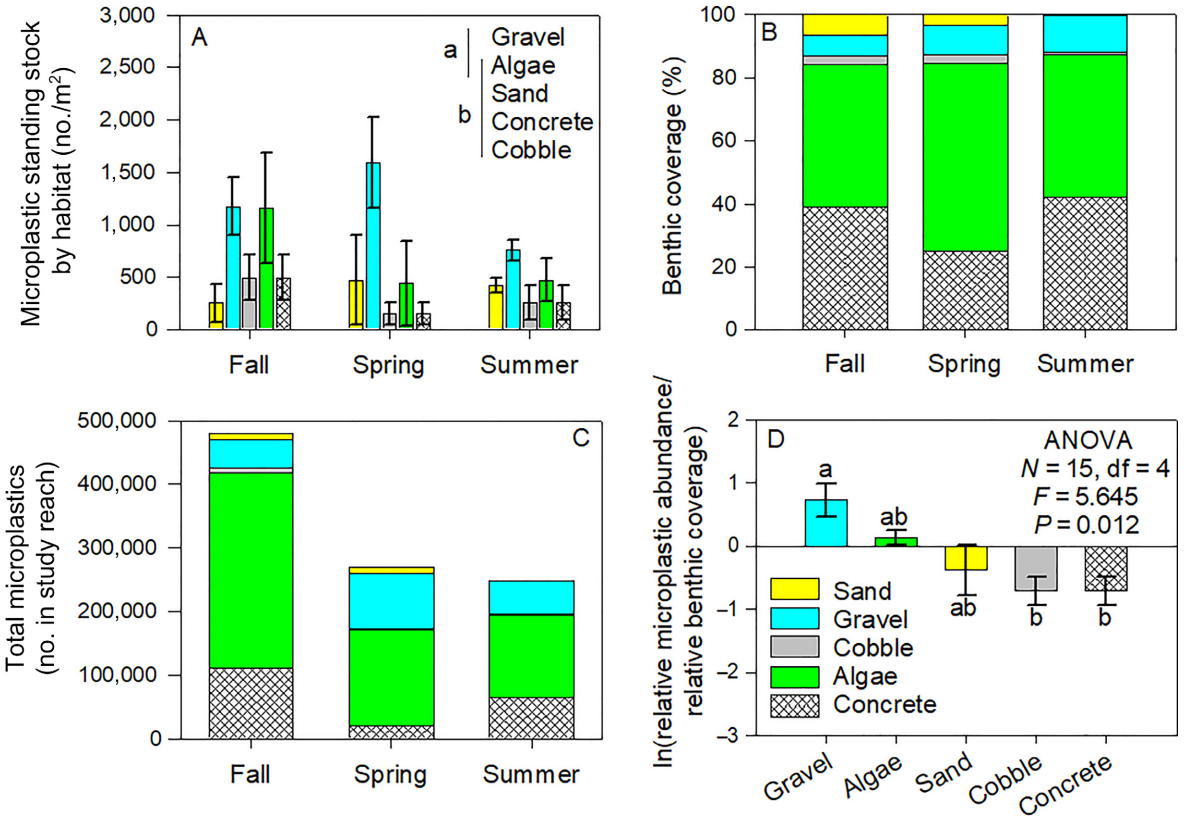

FIG. 2. Higgins Creek (A) microplastic standing stock (mean ± SE) by habitat, (B) benthic coverage by habitat, (C) total microplastics in study reach represented as the sum from benthic habitats, and (D) hot spot index calculated for each habitat during three seasons. Different letters indicate significant differences among habitats from Tukey's test. FBOM, fine benthic organic matter; CBOM, coarse benthic organic matter. Numerator and denominator degrees of freedom for the hot spot index F statistic are 4 and 10, respectively.

fibers were identified as plastic polymers (37.6%), processed natural materials (23.5%), or natural materials (39%). Plastic fibers were comprised of largely of polyester (84% of the plastic particles), with acrylic, polypropylene, and nylon also detected (Appendix S1: Fig. S4). Processed natural particles included dyed cotton, synthetic cellulose, and dyed cellulose-based fibers. Natural fibers included animal-based materials (e.g., wool), cotton, and cellulose-based products (Appendix S1: Fig. S4).

Organic matter

We compared suspended sediment and FBOM among sampling dates for each site. Suspended sediment (g AFDM/m^3) varied among seasons at all sites (one-way ANOVA, $P \leq 0.001$; Appendix S1: Table S2). At Higgins Creek and North Branch, suspended sediment was highest in summer, and did not differ in fall and spring. At Springbrook Creek, suspended sediment was highest in fall, spring was intermediate, and summer was lowest. Standing stock of FBOM (g AFDM/m^2) was significantly higher in fall in the North Branch, and spring and summer did not differ (one-way ANOVA; $P = 0.005$).

Spiraling metrics

We applied spiraling metrics to both microplastics and FPOM in each stream and each season. Downstream velocity of plastics ($V_{plastic}$) was faster by one to two orders of magnitude and more variable than FPOM (V_{FPOM}), ranging from 53 to 2,468 mm/s and 0.2 to 7.0 mm/s, respectively (Table 3). Indices of retention (IR) also showed higher retention for FPOM (40–1,384) than for microplastics (0.1–7.4; Table 3). In six out of the nine measurements, $IR_{plastic}$ was >1, demonstrating net microplastic retention. Where $IR_{plastic} < 1$, plastic was transported out of the study reach without retention. This occurred in fall and spring at the North Branch, and summer at Springbrook Creek (Table 3). The turnover rate of FPOM (k_{FPOM}; d^{-1}) measured via respiration (mean = 0.051 d^{-1}), was much higher than the turnover rate of plastics, which was a published value for polyethylene (2.74×10^{-6} d^{-1}; Pritchard 1998, Gewert et al. 2015). Finally, spiraling length (km), or the potential distance a particle moves before mineralization, ranged from 2 to 82 km for FPOM (S_{FPOM}). Due to slow rates of biological processing, plastic spiraling length ($S_{plastic}$)

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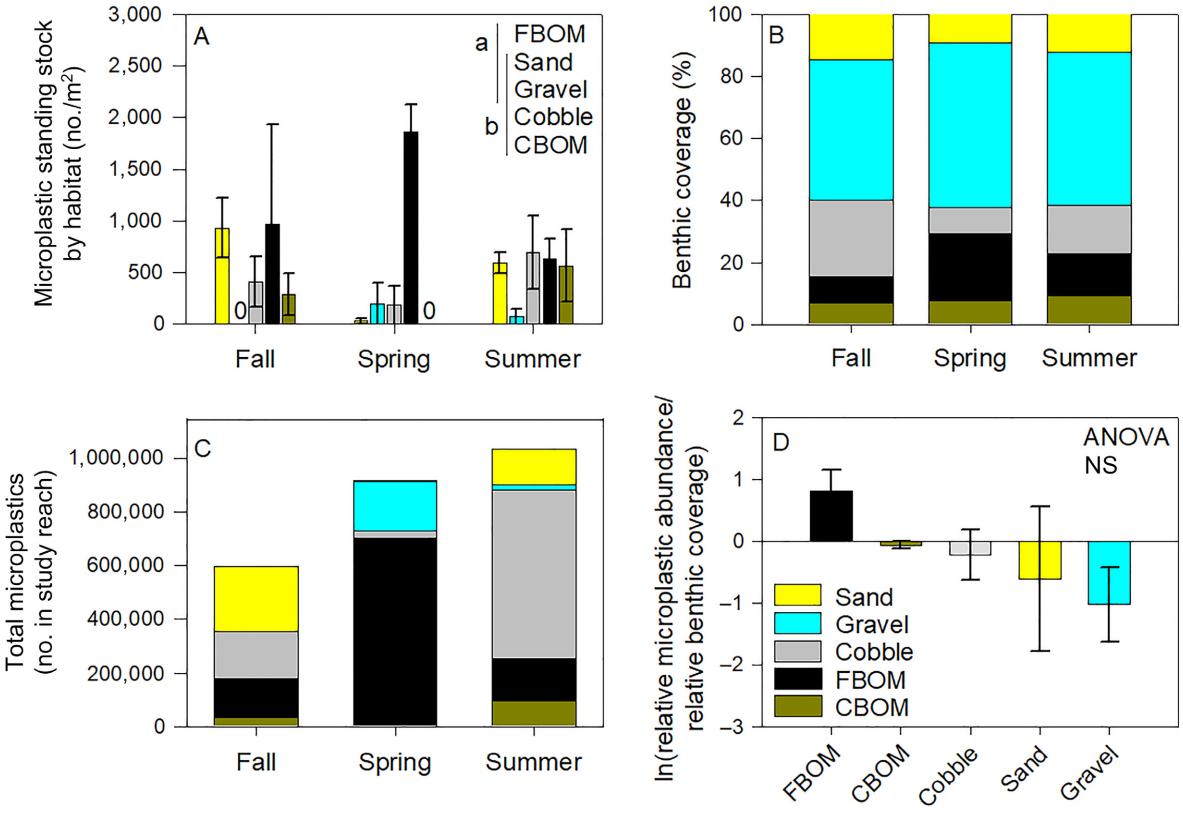

FIG. 3. North Branch Chicago River (A) microplastic standing stock (mean ± SE) by habitat, (B) benthic coverage (%) by habitat, (C) total microplastics in study reach represented as the sum from benthic habitats, and (D) hot spot index calculated for each habitat during three seasons. Different letters indicate significant differences among habitats from Tukey's test. Zeros indicate no data.

measurements were $>1.7 \times 10^6$ km, demonstrating that microplastics will not break down within the stream, and particles have the potential to move very long distances downstream prior to mineralization (Table 3).

DISCUSSION

Documenting the distribution and transport of microplastics in urban rivers is required to understand ecological dynamics of the particles. While spiraling metrics are fundamental in explaining particle transport, this study represents the first time the concept has been used in urban streams to compare movement of FPOM and microplastics, a novel form of allochthonous carbon. Transport results demonstrated that microplastic "spiraling" is dynamic, where particles have the capacity to be retained within streams, and also have the capacity to be transported far downstream prior to mineralization.

Spatial distribution of microplastics in streams

Microplastics were documented across all stream habitats (i.e., surface water, water column, and benthic zone) at all sites. While we observed no consistent seasonal patterns, microplastic abundance and composition varied among stream habitats. A clear pattern was that the concentration of suspended microplastics was much higher in the water column than the surface water habitat. This pattern of microplastic distribution can be explained by factors including particle density, biofilm colonization, habitat volume, and the methods used to collect the samples. While particles less dense than freshwater (1 g/cm^3) such as polypropylene (density = 0.946 g/cm^3) and foamed polystyrene (i.e., Styrofoam; density = 0.05 g/cm^3) will be transported near the water surface, many plastic polymers including polystyrene and acrylic (1.04 and 1.17 g/cm^3, respectively) will be transported while suspended in the water column before deposition or retention (Hoellein et al. 2019). Although, biofilm colonization of microplastics (Zettler et al. 2013, Harrison et al. 2014, McCormick et al. 2014, 2016, Hoellein et al. 2017), and turbulent conditions can enhance deposition, even of lightweight materials (e.g., polyethylene; Morét-Ferguson et al. 2010, Andrady 2011, Lobelle and Cunliffe 2011). Last, our estimates of surface water and water column microplastic concentration are affected by the methodology, including the collection devices and spatial distribution of the habitat

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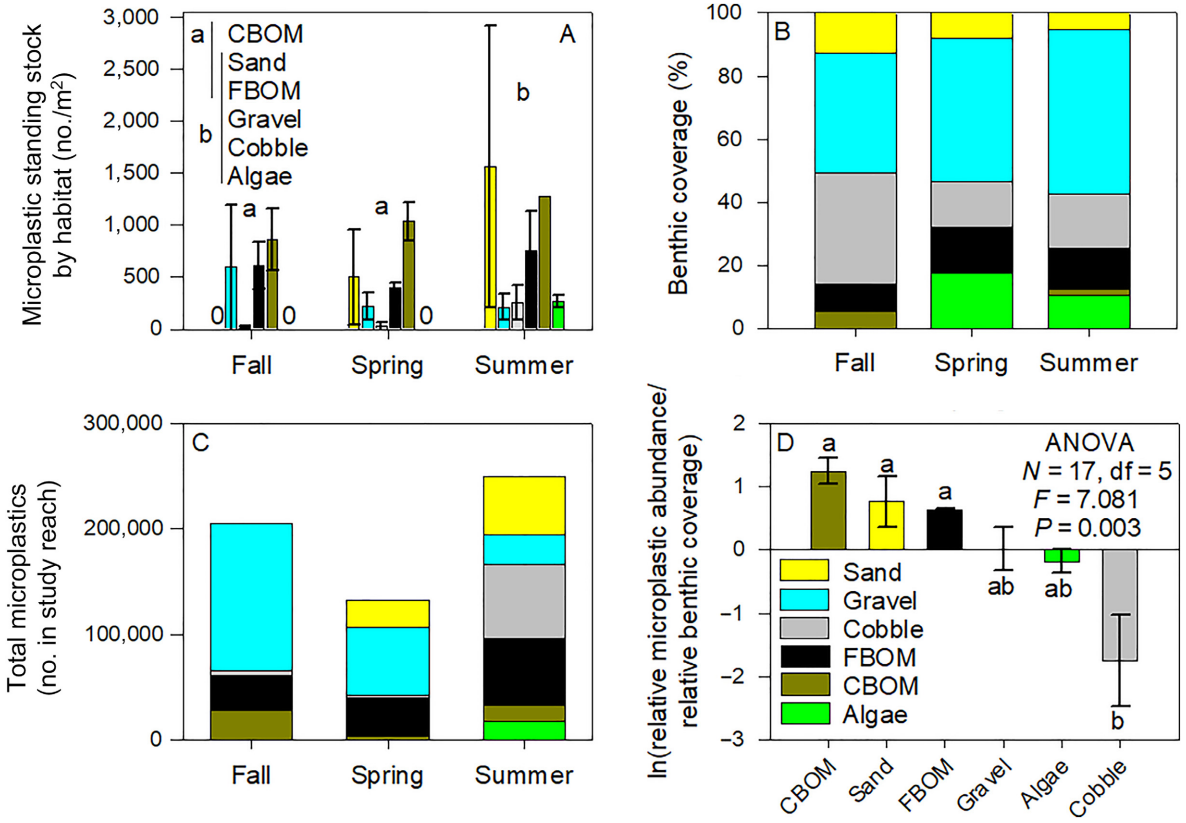

FIG. 4. Springbrook Creek (A) microplastic standing stock (mean ± SE) by habitat, (B) benthic coverage (%) by habitat, (C) total microplastics in study reach represented as the sum from benthic habitats, and (D) hot spot index calculated for each habitat during three seasons. Different letters indicate significant differences among habitats from Tukey's test. Zeros indicate no data. Numerator and denominator degrees of freedom for the hot spot index F statistic are 5 and 11, respectively.

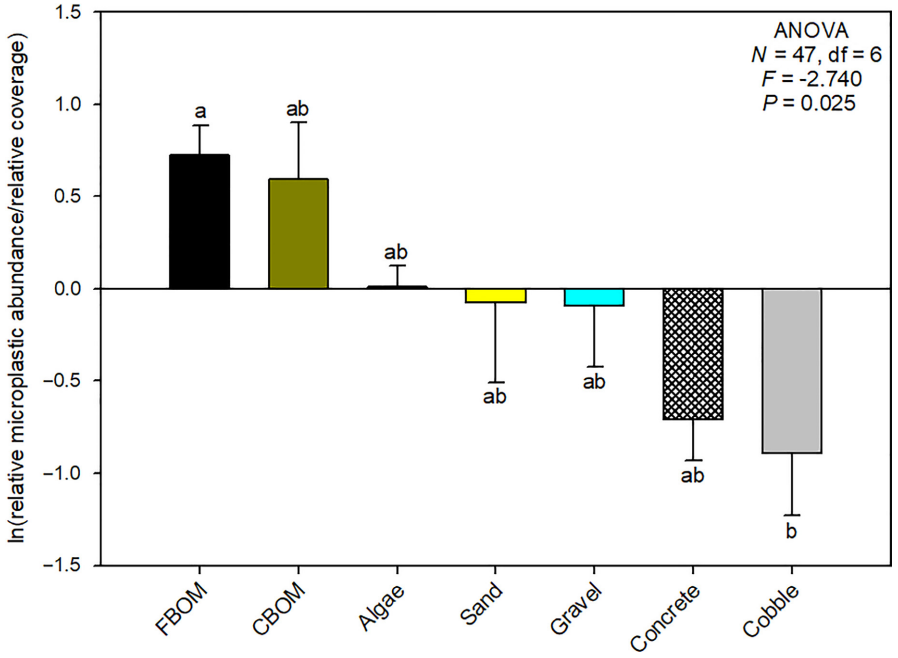

FIG. 5. Hot spot index for benthic habitats. Each bar is average among study sites and seasons (N = 9). Different letters indicate significant differences among habitats from Tukey's test.

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TABLE 3. Contributing variables for microplastic and FPOM used in spiraling calculations at three sites and time periods.

	Higgins Creek			North Branch			Springbrook Creek		
Variable	Fall	Spring	Summer	Fall	Spring	Summer	Fall	Spring	Summer
Discharge (Q, m^3/s)	0.85	0.67	1.20	1.87	0.94	0.87	0.26	0.38	0.19
Water velocity (m/s)	0.35	0.28	0.39	0.34	0.13	0.07	0.16	0.27	0.17
Width (m)	5.83	5.67	6.00	17.30	17.70	17.60	7.07	6.40	6.50
Depth (m)	0.31	0.34	0.38	0.43	0.49	0.55	0.19	0.19	0.14
Respiration ($g\,C{\cdot}m^{-2}{\cdot}d^{-1}$)	−0.52	−4.42	−0.32	−1.49	−0.45	−1.39	−0.53	−0.18	−0.19
Surface water MP ($no./m^3$)	0.53	2.01	0.50	0.26	0.13	0.66	0.49	0.47	0.29
Water column MP ($no./m^3$)	789	1028	108	773	556	1825	626	1067	992
Benthic MP ($no./m^2$)	823	467	414	339	527	564	334	206	391
Suspended sediment ($g\,AFDM/m^3$)	0.43	0.82	1.50	0.52	0.23	2.02	2.79	2.02	0.46
FBOM ($g\,AFDM/m^2$)	9.01	14.07	51.91	230.06	10.82	57.21	44.95	109.26	68.14
$V_{plastic}$ (mm/s)	140.3	260.6	52.8	2467.8	56.0	159.2	68.3	306.7	74.7
V_{FPOM} (mm/s)	7.0	6.9	5.8	0.2	1.1	1.7	2.3	1.1	0.2
$IR_{plastic}$	2.5	1.1	7.4	0.1	2.3	0.4	2.3	0.9	2.3
IR_{FPOM}	49.8	40.9	67.3	1383.7	113.5	40.4	70.7	246.5	854.8
$k_{plastic}$ (d^{-1})	2.74×10^{-6}	2.74×10^{-6}	2.74×10^{-6}	2.74×10^{-6}	2.74×10^{-6}	2.74×10^{-6}	2.74×10^{-6}	2.74×10^{-6}	2.74×10^{-6}
k_{FPOM} (d^{-1})	5.64×10^{-2}	3.08×10^{-1}	6.1×10^{-3}	6.50×10^{-3}	4.12×10^{-2}	2.38×10^{-2}	1.17×10^{-2}	1.70×10^{-3}	0.0021
$S_{plastic}$ (km)	4.4×10^6	8.2×10^6	1.7×10^6	7.8×10^7	1.8×10^6	5.0×10^6	2.2×10^6	9.7×10^6	2.4×10^6
S_{FPOM} (km)	11	2	82	3	2	6	17	57	8

Note: Abbreviations are FBOM, fine benthic organic matter; FPOM, fine particulate organic matter; IR, index of retention; k, turnover rate; MP, microplastic; no., number; Q, discharge; S, spiraling length; V, particle velocity.

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sampled. Surface water samples are limited to particles captured by the net travelling within the narrow section of water at the stream–atmosphere interface. Due to the small area of the neuston net opening, as well as net mesh size, measurements of microplastic concentration in the surface water are likely underestimates (Barrows et al. 2017). In addition, low-flow zones (e.g., pools, debris dams, and stream edges) are retention sites for floating microplastics that are not included in net measurements (Hoellein et al. 2019). Floating particles not captured by the net could represent a significant contribution to estimates of riverine microplastics in the context of a plastic budget. In contrast, water column samples were collected at varying depths throughout the water column and incorporate sections of streams not captured by neuston nets. However, the volume of water included in each water column sample is smaller than the net measurements, so estimations of microplastic concentration within the water column are susceptible to wide variation among replicate samples. This is consistent with concurrent measurements of net and water column samples collected elsewhere (Barrows et al. 2017, 2018, Miller et al. 2017). Recent developments in techniques for stream water microplastic collection (e.g., larger volumes, depth-integrated sampling Lenaker et al. 2019, Grbić et al. 2020), will help resolve discrepancies.

Another consistent pattern for microplastic spatial distribution in the study streams was the high density of microplastics in benthic zone relative to the water column. High estimates of microplastic standing stock suggest the benthic zone acts as a microplastic sink. Other studies have suggested the same pattern, where benthic microplastic standing stock exceeds suspended concentrations in freshwaters (Castañeda et al. 2014, Klein et al. 2015, Hoellein et al. 2017). In addition, factors including microplastic shape, density, and biofilm colonization alter deposition dynamics and result in faster assimilation into the benthos. For example, Hoellein et al. (2019) added microplastics of varying polymer types, with and without biofilm colonization, to show the effects of environmental conditions, material, and particle size on deposition in streams, and reported that polystyrene fragments were retained faster than polypropylene pellets. Colonization by microbial biofilms enhanced the "stickiness" of the particles and enhanced deposition (Hoellein et al. 2019). Over long time periods, microplastics in the benthos may become buried and will remain in place until the sediment is disturbed by flooding and microplastics are resuspended (Nizzetto et al. 2016, Hurley et al. 2018).

Distribution of benthic microplastics

The benthic zones of streams are heterogeneous mixtures of substrate types reflecting depositional and erosional habitats. We therefore expected to find differences in microplastic abundance among substrates. While previous research has measured microplastic contamination in marine and freshwater sediment in general (Cauwenberghe et al. 2013, Corcoran et al. 2015, Klein et al. 2015, Ballent et al. 2016), few studies have assessed microplastic standing stocks among different benthic habitats (Corcoran et al. 2020). Thus, our benthic habitat-specific assessments are a novel approach to explaining the distribution of benthic microplastic in rivers. As expected, benthic surveys and hot spot index calculations showed that the benthic zone is composed of a patchwork of different habitats that vary in their retentive capacity for microplastic particles. Distribution of these habitats is a result of hydrologic patterns, where depositional habitats (e.g., FBOM, CBOM) tend to occur in low flow sections of the stream near pools or fallen debris, while erosional habitats (e.g., concrete, cobble) are more common in higher-flow areas (Fig. 5). Depositional habitats (e.g., FBOM) located along river edges contained higher concentrations of microplastics, relative to erosional habitats in the main channel (Corcoran et al. 2020).

Our results are consistent with several inferences generated about spatial distribution of benthic microplastics compared to point sources, benthic organic matter, and water velocity in previous research. Hoellein et al. (2017) noted variation in sand and fine benthic organic matter as a potential correlate with microplastic density, but the authors did not measure microplastic concentrations for the sediment types separately. Areas of low water velocity were associated with higher concentrations of fine sediment and microplastics in the Rhine River, Thames River (Canada), and along the Italian coast (Vianello et al. 2013, Mani et al. 2016, Corcoran et al. 2020). In a similar fashion, Castañeda et al. (2014) reported little to no microplastic in areas of the St. Lawrence River with coarse benthic particles (e.g., gravel and cobble). In our study, high variability within and among substrates was common, but we also found depositional habitats retained more microplastics than erosional habitats relative to their benthic coverage, thus following published patterns for the retention of FPOM and CBOM (Webster et al. 1999, Minshall et al. 2000).

Microplastic polymer identification

Fiber identification results demonstrate the prevalence of synthetic microplastic fibers, semi-synthetic materials, and processed natural fibers in the environment, and we acknowledge that the term "microplastics" include a wide diversity of material types (Rochman et al. 2019). Of the plastic polymers identified, polyester was the most abundant and appeared across all sample categories. Our results are indicative of trends in global plastic use where polyester is one of the most common fiber types documented in the environment and is used for ~50% of the fiber market (Browne et al. 2011, Courtene-Jones et al. 2019). Although natural cotton and cellulose-based fibers were also documented in tested samples, the presence of *processed* forms suggest that

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microfiber pollution includes plastic as well as highly processed materials with added dyes, waxes, and synthetic chemicals (Corcoran et al. 2020). Despite being classified as "plant-based" microfibers, the mixture of chemical additives indicated the particles were environmental pollutants and are thereby included in this study under the umbrella term microplastics (Laskar and Kumar 2019, Rochman et al. 2019).

Microplastic "spiraling"

A key objective of this study was to compare the transport of FPOM with microplastics in urban streams. Nutrient and carbon "spiraling" has long been a focus of research on flowing waters, with well-developed methods, data syntheses, and models to explain factors that drive element transport including hydrologic conditions and biological activity (Webster 1975, Newbold et al. 1982, Griffiths et al. 2012). Some early studies have used the spiraling concept to quantify microplastic transport dynamics in streams (Hoellein et al. 2017, 2019), but microplastics and fine organic particles have not been compared directly.

Contrary to our expectations, microplastics were transported downstream faster than FPOM. Variability in FPOM and microplastic particle velocity among sites is due in part to differences in environmental and physical conditions among study streams and dates. For example, higher estimates of particle velocity are found at our North Branch site, which was 9–10 m wider than the other two sites and had higher discharge. Comparing between particle types, indices of retention demonstrated that both FPOM and microplastics were retained within our study reaches in most cases, but FPOM was retained to a greater degree than microplastics. Differences in V_{FPOM} and V_{plastic} were strongly affected by the ratio of material in the water column to that in the benthos. The ratio was higher for microplastic because there as more microplastic in the water column relative to the streambed compared to FPOM. A key factor affecting the ratio may be the enumeration method for each particle type (i.e., plastic counts vs. FPOM mass), and the ratio may change if microplastic mass was estimated by measuring for the distribution of particles according to size and density. In addition, separation methods may underestimate benthic microplastics, further coloring interpretation of this ratio. Our approach is a key step towards quantifying microplastic spiraling, but the next logical steps for future research will be to quantify microplastic particle mass across all ecosystem compartments and habitat types.

The spiraling length (S) for FPOM was much shorter than for microplastics, attributed to the lower turnover rate and higher downstream velocity of plastic. While on average FPOM will be metabolized within ~21 km of downstream transport, microplastics can *theoretically* travel >12 million km before being mineralized to carbon dioxide. The exceptionally long value therefore does not estimate a specific travel distance, but instead the result indicates the potential travel length of microplastics is effectively unlimited. The conclusion drawn from this value is that, although on average microplastic particles will be retained in our 100 m study reaches (mean IR = 2.15), the same particles have the capacity to also be transported very far downstream before being respired. In fact, the same microplastic particle can be an active and potential long-term resident of flowing waters, and then later do the same in downstream bodies of water such as estuaries and oceans. This pattern is logical given the durability of plastic is intentional to its design but offers a clear contrast when estimates of degradation relative to transport are placed in direct comparison to naturally occurring fine particles in streams.

Calculating turnover requires estimations of mineralization rates for both FPOM and microplastics. Methods and expected ranges for FPOM are much better established than for microplastics. We quantified respiration and concentration of suspended and benthic fine particles according to widely used protocols (Tank et al. 2010). Our estimations of in situ FPOM respiration ranged from −9 to −216 mg $O_2 \cdot m^{-2} \cdot h^{-1}$ and are similar to respiration rates for FPOM in other streams in the region (−20 to −50 mg $O_2 \cdot m^{-2} \cdot h^{-1}$; Hoellein et al. 2019). In contrast, the mineralization rate of microplastics has much greater uncertainty. We used a published rate of polyethylene mineralization as the value for microplastic turnover rate in our calculations (0.1% per yr; Pritchard 1998, Gewert et al. 2015). This estimation of plastic mineralization was conducted under optimal lab conditions and may not represent in situ mineralization rates. Other studies have noted plastic degradation in the environment differs by polymer and is dependent on linked abiotic and biotic processes including UV-radiation, physical abrasion, and enzymatic breakdown by microbes (Gewert et al. 2015, Rummel et al. 2017). For example, O'Brine and Thompson (2010) showed that colonization of polyethylene bags by marine biofilms decreased UV exposure by ~90%, thus slowing the breakdown of the polymer. In our study, microplastic concentrations were highest at benthic habitats (e.g., FBOM, CBOM), where low oxygen and light conditions may further restrict breakdown rates (Fig. 5). We may therefore expect microplastics within cobble or concrete habitats to break down or be transported downstream at a faster rate than plastics stored within organic matter depositional zones. However, this has not been measured. We suggest more empirical and modeling studies are needed to refine estimates of mineralization rates for microplastic polymers under a wide variety of conditions and substrates typical of stream ecosystems.

Method evolution: Critique and suggestions for harmonization among many habitat types

This is the first study to compare microplastic concentrations in surface water, water column, and seven distinct benthic habitats, which presents non-trivial

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challenges for alignment of methods. We used different collection devices (i.e., nets, bottles, and sediment grabs), sieve sizes, and salinity separations. These were necessitated by the wide variety of conditions presented by each of the sample types. For example, while water column and cobble samples were not sieved prior to processing, net samples were rinsed over 0.3- and 4.75-mm sieves, and other sediment types were rinsed over 0.3-, 1.0-, and 4.75-mm sieves. We note that, despite differences in sample processing methods for water column, net, sediment (i.e., non-cobble), and cobble samples, mean fiber length ranged from 1.13 to 1.8 mm across all sampling types (Appendix S1: Fig. S3). In addition, fragment length ranged from 0.78 to 0.98 mm across the sample types (Table 1). These results suggest that processing methods did not generate large discrepancies in microplastic size among sample types. However, we recommend future studies can avoid this complication by using sieves with the same mesh size for all sample types. In addition, methodological artifacts inherent to processing of various stream habitat types can be minimized by newly emerging methods for separation of sediment and microplastics, although each have additional caveats to consider (Claessens et al. 2013, Crichton et al. 2017). For example, we used sodium chloride for separation of microplastics in some benthic habitats (sand and gravel), which means we may have underestimated benthic microplastic standing stock in those habitats, which can result in an overestimation of V_{plastic}. We suggest future assessments use salts such as sodium iodide and zinc chloride, which offer more dense solutions. However, these come with important considerations for purchasing costs and hazardous waste management compared to sodium chloride, and may not be feasible for all researchers (Claessens et al. 2013, Coppock et al. 2017).

Finally, we note that despite a potential underestimate of benthic microplastic standing stocks, benthic density estimates were orders of magnitude higher than net and water column results in eight out of the nine sample dates. Thus, we argue that our conclusions for microplastic deposition rates are conservative, and that future studies will benefit from this initial approach as a comprehensive assessment among stream habitats. Overall, this field of study is under rapid development, which includes the need for method standardization and comparison across ecosystem and habitat types (Hartmann et al. 2019).

Implications for global plastic budgets

The delivery of microplastics from rivers to oceans is a key contribution to global accumulations of plastic. Past research focused on plastic abundance and movement in marine ecosystems, thereby considering rivers as simple conduits that transport microplastics from terrestrial environments to the ocean (Browne et al. 2011, Jambeck et al. 2015, Schmidt et al. 2017). While the capacity for rivers to retain, transform, and redistribute plastic particles is understudied, recent work has demonstrated the dynamic nature of rivers as a source and sink for plastic litter from inland regions (McCormick and Hoellein 2016, Hoellein et al. 2017, McNeish et al. 2018). Global budgets of plastic pollution cannot be established before greater assessments of the dynamic nature of microplastic movement in flowing waters are completed.

Our research sought to inform the overarching question, "are rivers a source or sink of microplastics to downstream ecosystems?" and unexpectedly, our data suggest that the answer is "both." Together, the high estimates of microplastic standing stocks, values for indices of retention, and spiraling lengths all suggest that the same microplastic particles can be retained, transported long distances, and released into a larger river system or body of water, all prior to being mineralized into carbon dioxide. Similarly, the differences in hot spot indices among stream benthic habitats showed the simultaneous retention and transport of microplastics according to hydrodynamics. While erosional habitats (e.g., concrete, cobble) may only temporarily retain microplastics, depositional habitats (e.g., FBOM, CBOM) can withhold plastics for longer durations until particles are suspended following a disturbance or high-flow event (Hurley et al. 2018). Retention in stream ecosystems and slow rates of biodegradation support the classification of microplastics as a highly recalcitrant form of allochthonous carbon in streams, and should be studied using well developed protocols for naturally particles (Hoellein et al. 2019). This work contributes to the ongoing effort to document microplastic distribution among global ecosystems, including river habitats. By using spiraling metrics to quantify fine particle transport and microplastics, these data will inform models that clarify the role of rivers in global plastic budgets.

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