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Article

Spatial Distribution of Microplastics in Surficial Benthic Sediment of Lake Michigan and Lake Erie

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ABSTRACT: The spatial distribution, concentration, particle size, and polymer compositions of microplastics in Lake Michigan and Lake Erie sediment were investigated. Fibers/lines were the most abundant of the five particle types characterized. Microplastic particles were observed in all samples with mean concentrations for particles greater than 0.355 mm of 65.2 p kg⁻¹ in Lake Michigan samples (n = 20) and 431 p kg⁻¹ in Lake Erie samples (n = 12). Additional analysis of particles with size 0.1250–0.3549 mm in Lake Erie resulted in a mean concentration of 631 p kg⁻¹. The majority of polymers in Lake Michigan samples were poly(ethylene terephthalate) (PET), high-density polyethylene (HDPE), and semisynthetic cellulose (S.S. Cellulose), and in Lake Erie samples were S.S. Cellulose, polypropylene (PP), and poly(vinyl chloride)



(PVC). Polymer density estimates indicated that 85 and 74% of observed microplastic particles have a density greater than 1.1 g cm^{-3} for Lake Michigan and Lake Erie, respectively. The current study provided a multidimensional dataset on the spatial distribution of microplastics in benthic sediment from Lake Michigan and Lake Erie and valuable information for assessment of the fate of microplastics in the Great Lakes.

1. INTRODUCTION

Microplastics are ubiquitous throughout the natural environment with their prevalence being observed in marine water, freshwater, groundwater, drinking water, remote mountains, Arctic sea ice, the atmosphere, and can be found within numerous organisms including humans.¹⁻⁹ Microplastics are plastic particles measuring less than 5 mm in diameter and are commonly categorized as fibers/lines, fragments, films, pellets/ beads, and foams. Microplastics originate from primary or secondary sources; primary sources fulfill a specific need or function (e.g., microbeads used in personal care products, abrasive cleaning particles, pre-production resin pellets, and microfibers used in manufacturing textiles),^{10,11} and secondary sources result from degradation, wear and tear, or fragmentation of larger debris. Secondary sources include litter fragments (e.g., plastic bags, bottles, wrappers, Styrofoam containers, cigarette filters), synthetic fibers from textiles, road salt, and tire wear particles.¹²⁻¹⁶ Pathways for microplastics to enter the aquatic environment can include stormwater runoff (e.g., litter and tire wear particles); domestic, industrial, and commercial wastewater treatment plant effluent; treated sewage sludge and illicit discharges (e.g., plastic shavings and dust, microbeads, and synthetic fibers); organic fertilizers from biowaste fermentation and composting (e.g., litter fragments, synthetic fibers, microbeads); and atmospheric deposition (e.g., synthetic fibers).^{13,17–21} The number of organisms with observed microplastics in their digestive systems continues to increase, but the consequences for ingestion of microplastics are not well understood. Plastic pieces of varying sizes have been ingested by many different marine and freshwater organisms including mammals,^{22–24} birds,^{25–28} crustaceans,^{29–31} mollusks,^{3,32–34} worms,^{35,36} and fish.^{37–42} Potential negative effects on organisms that have been documented include obstruction of the digestive system, clogging of feeding appendages, reduced food consumption and predator performance, nutritional deprivation, reduced immune response, substantial energy reduction, and impaired reproduction and survival.^{31,43–48}

Microplastics have been reported from various Great Lakes hydrologic settings and compartments, including tributary surface and subsurface water, and sediment, lake surface and subsurface water, nearshore and offshore sediment, and shoreline sediment.^{49–62} Comparison of water surface samples in the Great Lakes with water surface samples in tributaries indicate a large difference in the relative percent of different

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microplastic particle types. Fibers were much more prevalent in tributary samples (52 to 97% of particles)^{51,63} than in lake samples (<1-14%).^{53,55,56,61} A subsequent study of Milwaukee, Wisconsin, rivers and nearshore environments indicated that microplastics were distributed throughout the water column and density profiles reflected increasing polymer densities from the surface to the subsurface to the sediment.⁵⁹ Low-density microplastics ($<1.1 \text{ g cm}^{-3}$), such as foams from expanded polystyrene and fragments from polypropylene were shown to be the most abundant in water surface samples. Conversely, high-density (>1.1 g cm^{-3}) particles from synthetic fibers, such as poly(ethylene terephthalate) (i.e., polyester, PET) had the opposite result with particle abundance increasing with water depth down into the sediment.⁵⁹ Similar results describing vertical partitioning based on polymer density have been observed previously.^{64,65} These results indicate that microplastic particles with a density greater than 1.1 g cm⁻³, such as fibers, are likely to be settling out in the Great Lakes, and this could help explain the difference in the relative abundance of fibers in tributary samples compared with lake samples. While studies of lake sediment to date have begun to provide local-scale information, they have not been of sufficient scale to verify that high-density microplastic particles greater than 1.1 g cm⁻³ are indeed settling out in Great Lakes benthic sediment. The majority of Great Lakes sediment samples have been collected from shoreline, nearshore, and coastal environ-ments, 49,50,52,54,58,59 and the St. Lawrence River.⁶⁶ To further understand fate and transport within the Great Lakes Basin, a large-scale spatial representation of sediment microplastics presence in Great Lakes benthic sediment in the offshore lake environment is needed.

The goal of this study was to determine the spatial extent of microplastics contamination in Lake Michigan and Lake Erie surficial benthic sediment and the relative abundance of different microplastic types, sizes, and polymeric composition (microplastics properties). To do this, sediment was sampled from multiple locations within each of the two lakes for detailed analysis. Results may be used in future assessment of microplastics fate and transport in freshwater environments and for informing the assessment of potential biological effects for particles with varying microplastics properties.

2. MATERIALS AND METHODS

2.1. Sample Collection and Analysis. Sampling locations in Lake Michigan and Lake Erie were selected to provide a representative coverage of the different characteristics within the lakes, which include a range in water depth (15–240 m in Lake Michigan and 7.5–62.8 m in Lake Erie), spatial coverage around the lakes, and variable distance from the mouths of tributaries (Table S1).

Twenty sediment samples were collected from Lake Michigan in September 2013, and 12 sediment samples were collected from Lake Erie in September 2014. Adequate sediment samples could not be collected at six locations in Lake Michigan and eight locations in Lake Erie due to a lack of fine-grain sediment, or a high abundance of *Dreissena bugensis* (quagga) or *Dreissena polymorpha* (zebra) mussels (Table S1).

Lake Michigan and Lake Erie sediment samples were collected while on-board the U.S. Environmental Protection Agency's (EPA) R/V *Lake Guardian*. Lake Michigan sediment was obtained using a stainless-steel standard ponar with an 0.229 m \times 0.229 m sampling area (0.0524 m²) and a scoop

volume of 8.2 L (Ponar Grab, Standard, Wildco, Yulee, Florida). Immediately upon retrieval, sediment was transferred from the ponar dredge to a stainless-steel pan where a stainless-steel spatula was used to subsample approximately the top 2 cm of sediment. Sediment was then transferred to a 500mL baked amber glass bottle with a Teflon-lined lid, labeled and frozen. Lake Erie sediment was collected with a four-core multicorer (model MC-400, Ocean Instruments, Inc., San Diego, California). The coring tubes used in Lake Erie were 10 cm \times 60 cm polycarbonate tubes. Four sediment samples were collected per multicorer cast. Immediately upon removal from the multicorer, sediment cores were placed on a pneumatic sediment extrusion device to be sectioned into 2-cm intervals. The top 2 cm from two cores were composited into a 500-mL baked amber glass bottle and frozen.

Sediment samples were processed using an improved National Oceanic and Atmospheric Administration (NOAA) laboratory method⁶⁷ as modified by Zobkov and Esiukova⁶¹ with further details provided in the Supporting Information (Text S1). Briefly, sediment processing consisted of a threestep method: extraction of microplastics three times using a zinc chloride solution ($\rho = 1.6 \text{ g mL}^{-1}$), wet peroxide oxidation digestion of floating organic and plastic materials and calcite digestion using 4.5% hydrochloric acid. Digestions were done at room temperature to minimize the effects of the digestion process on the integrity of microplastic particles. After processing, the Lake Michigan samples were filtered through a stacked set of 8-in. diameter stainless-steel mesh sieves (4.750, 1.000, and 0.355 mm), while those for Lake Erie were filtered through sieves of smaller mesh sizes (1.000, 0.355, and 0.125 mm). In both cases, the solid material was separated into three size fractions, but the size fractions were different: 0.355-0.999, 1.000-4.749, and >4.750 mm for Lake Michigan, and 0.125-0.3549, 0.355-0.999, and >1.000 mm for Lake Erie. The different size fractions between Lake Michigan and Lake Erie provided information on smaller particle sizes to gain additional insight into microplastics presence. No microplastics were found in the >4.750 mm size fraction in Lake Michigan. As a result, the number of particles counted in each size fraction 1.000-4.749 and >4.750 mm were summed together so Lake Michigan sediment could have two comparable size fractions, 0.355-0.999 mm and >1.000 mm, to Lake Erie. Individual sieve contents from Lake Michigan and Lake Erie samples were transferred to Petri dishes using deionized (DI) water for microplastic abstraction. After filtering by size, microplastics were microscopically analyzed to enumerate and categorize microplastic particles according to plastic type: fragments, pellets/beads, fibers/lines, films, or foams. Results presented are likely to underestimate concentrations given the potential for some microplastics to adhere to settling material during the extraction process, the potential losses that could occur during sieving, and the potential loss of some polymers from the chemical digestions.

Plastic particle concentrations are reported as particles per kilogram (dry weight) sediment (p kg⁻¹). All sample results for this study are available online.⁶⁹

To confirm the polymeric nature of collected microplastic particles, a subset of Lake Michigan (9%, n = 37) and Lake Erie (5.5%, n = 44) particles were selected for further analysis by Fourier transform infrared (FTIR) attenuated total reflectance spectroscopy (Table S2). All particles chosen for FTIR analysis were randomly selected, but with regard to obtaining acceptable FTIR spectra, and were less than 1 mm in

size. An attempt was made to get good spectra on every particle selected for FTIR analysis. Whichever particles were first spotted under the microscope were extracted for FTIR analysis. Once 10-20 readings from a single sample were obtained, lab analysts would move on to another sample. Selected particles were analyzed using a PerkinElmer Spectrum Two FTIR with a universal attenuated total reflectance accessory attachment operating in reflectance mode at 64 scans with 4 cm⁻¹ resolution. Polymer identification was made by comparing sample spectra to an internal manufacturerprovided spectrum library, requiring a minimum 80% match for acceptance. The amount of FTIR analysis performed by particle type varied for Lake Michigan and Lake Erie microplastic particles, with fiber/line particles from Lake Michigan (7%) and Lake Erie (4%) being the lowest percentage of particle types analyzed (Table S2). All FTIR results for this study are available online.⁶⁹

2.2. Data Analysis. Because the percentage of particle types analyzed by FTIR were not exactly the same as the percentage of each particle type observed in samples, FTIR results were normalized to estimate the percentage of particles represented by each polymer in the observed samples for each lake. This computation was done using eq 1

$$PPE = \left(\frac{NP}{PT}\right) \times TP \tag{1}$$

where NP equals the number of particles identified as a specific polymer (e.g., polypropylene) per particle type (e.g., fiber/ line), PT equals the number of individual particles analyzed via FTIR by particle type, TP equals the total number of particles collected by particle type for each lake sampled (Michigan or Erie), and PPE equals the estimated number of particle types by polymer type. This normalization routine was intended to provide estimates for comparison, but accuracy is limited by the number of particles analyzed by FTIR. Computations were done separately for each lake with additional interpretation based on common density values for the polymers.⁵⁹

Statistical significance for concentrations from the overlapping size classes 0.355-0.999 and >1.000 mm from each lake was determined with a pairwise Wilcoxon rank-sum test with corrections for multiple comparisons (p < 0.05).⁷⁰

2.3. Quality Assurance and Quality Control. To determine recovery rates for microplastics in sediment using the NOAA laboratory method⁶⁷ recovery was evaluated in 2015, and again in 2017 using the revised NOAA method.⁶⁸ Briefly, in 2015, Lake Erie sediment was collected at Dunkirk Harbor, New York, using a ponar grab sampler specifically for methods testing. Microbeads were extracted from consumer facewash products by emptying entire tubes into a 0.125 mm stainless-steel sieve and were washed with deionized water until all of the soap residue was rinsed away. The remaining microbeads were transferred into a Petri dish, dried in a convention oven at 50 °C, and transferred to a glass vial for later use. Eight test samples were produced using 60 to 500 mL of Dunkirk Harbor sediment spiked with 10 to 100 extracted microbeads. These eight samples were processed in 2015 using the NOAA method⁶⁷ yielding recovery rates of 67 to 100% (mean = 90%, standard deviation (SD) 12%).

Two additional recovery tests were performed again in 2017 following the publication of the revised NOAA method⁶⁸ using Dunkirk Harbor sediment from the previous recovery tests. For these two tests, PET fibers were created by trimming thread purchased from a local fabric store. PET and

polypropylene fragments were created by cutting up a postconsumer cup and straw. Twenty-five fragments, half of all the spiked fragments, were greater than 1.0 mm, and the remaining spiked plastic particles ranged from 0.125 to 0.355 mm in size. Overall recovery results had a mean of 92% SD 7% from these two tests and were consistent with results reported by Zobkov and Esiukova using the revised NOAA method.⁶⁸ Mean recovery rates of 94% were obtained for fiber/lines and fragments. Further details regarding microplastic recovery tests are provided in Supporting Information Text S2.

To assess potential contamination from laboratory materials or ambient laboratory air, two laboratory blanks were collected and analyzed alongside the environmental samples and have previously been reported.⁵⁹ Laboratory blanks consisted of deionized water stored in open sample containers for periods of 1-14 days. The two laboratory blanks had fiber/lines in the two smallest size categories for a total of three and six fiber/ lines, respectively.

Duplicate sediment samples were collected at three different sampling locations on Lake Michigan and two sampling locations on Lake Erie. Relative percent difference (RPD) for the total microplastics concentration of the three duplicate sediment samples from Lake Michigan was 8, 28, and 116%, and the two duplicate sediment samples from Lake Erie was 11 and 126%.⁶⁹ Additional RPD details by particle type are provided in the Supporting Information (Text S3 and Table S3). The number of microplastics collected in each duplicate sediment sample pair varied by lake, sampling location, and particle type (Figures S1 and S2). These RPD duplicate sediment results represent a combination of variability from laboratory procedures, an incomplete mixture of sediment prior to transferring into the sample collection bottle, and sediment heterogeneity.

Polycarbonate core tubes were individually inspected prior to sampling. Visual evidence for potential contamination from these tubes was not apparent, but additional blank samples were not collected to verify this observation. Given that only one of the seven fragment particles analyzed by FTIR were found to be polycarbonate, it was considered unlikely that the core tubes introduced contamination of polycarbonate particles.

3. RESULTS

3.1. Sediment Microplastics. The smaller particle size fraction enumerated in Lake Erie samples (0.1250-0.3549 mm) was not enumerated in Lake Michigan samples. Results presented will first include the common particle size fractions analyzed in samples from both lakes (0.355-0.999 mm) and >1.000 mm and then the smaller particle size fraction for Lake Erie. In Lake Michigan no particles were greater than 4.750 mm, and therefore the two largest size fractions (1.000-4.749 and >4.750 mm) were combined to make a >1.000 mm size fraction for analysis herein. A full accounting of all sample results including concentrations for each particle size fraction, particle type within each sample, and polymers identified in a subset of particles is available online.⁶⁹

Microplastic particles were observed in every sediment sample collected. The concentration of particles of size fraction greater than 0.355 mm ranged from 9.1 to 318 p kg⁻¹ with a mean of 65.2 p kg⁻¹ in Lake Michigan (n = 20) and 77.6 to 1680 p kg⁻¹ with a mean of 431 p kg⁻¹ in Lake Erie (n = 12; Figures 1–3A). Concentrations of the additional smaller particle size fraction analyzed in Lake Erie samples ranged



Figure 1. Sediment sampling locations and concentrations by particle type for sediment samples collected from Lake Michigan. The black dots represent sampled locations, X indicates a location where bed material composition did not allow sample collection, major streams are indicated with blue lines, and state/provincial boundaries are shown with gray lines.^{71,72} Particles include the size fractions >0.355 mm. (p kg⁻¹, particles per kilogram dry weight sediment; mm, millimeters; >, greater than).

from 110 to 3200 p kg⁻¹ with a mean of 631 p kg⁻¹ (Figures 2, and 3A). The size fraction 0.355–0.999 mm represented 77% of particles from all size fractions for Lake Michigan and 36% for Lake Erie. The smallest size fraction 0.125-0.3549 mm accounted for 63% of observed particles from all size fractions in Lake Erie samples (Table S4).

Observed microplastics types were dominated by fibers/lines for all size fractions in samples from both lakes (Figure 3B). Particles that were greater than 0.355 mm consisted of 91 and 75% fibers/lines for Lake Michigan and Lake Erie, respectively. Particles in the small size fraction from Lake Erie consisted of 59% fibers/lines. Other microplastics that were greater than 0.355 mm in Lake Michigan and Lake Erie samples consisted of 1.5 and 10.2% fragments, 1.3 and 3.4% pellets/beads, 0.90 and 6.4% films, and 5.0 and 5.4% foams, respectively (Figure 3B). Microplastics observed in the smallest size fraction from Lake Erie consisted of 20.6% fragments, 10.5% pellets/beads, 1.7% films, and 8.6% foams (Figure 3B).

Mean concentrations for the combined comparable size fractions (>0.355 mm) were greater in Lake Erie samples than Lake Michigan samples, including total concentration and the individual particle type fragments, fiber/lines, and films (p < 0.05, Figure 4). Pellets/beads and foams were only present in

six and nine samples between both lakes, respectively; and therefore, results between the two lakes for these particle types were not significantly different.

3.2. Polymer Identification. FTIR analysis of 37 particles from Lake Michigan sediment samples (9% of all Lake Michigan sediment particles) resulted in identification of eight different polymers (Figure S3 and Table S5). Thirty-two percent of analyzed particles were identified as poly(ethylene terephthalate) (PET, "polyester"), 24% as high-density polyethylene (HDPE), and 19% as semisynthetic cellulose (S.S. Cellulose). The other five identified polymers each accounted for less than 8% of the total (Figure S3 and Table S5).

FTIR analysis of 44 particles from Lake Erie sediment samples (5.5% of all Lake Erie sediment particles) resulted in identification of 13 different polymers (Figure S3 and Table S5). Twenty-three percent of analyzed particles were identified as S.S. Cellulose, 14% as polypropylene (PP), and 11% as poly(vinyl chloride) (PVC). The other 10 identified polymers each accounted for less than 9% of the total, and polymers could not be identified for 9% of the particles (Figure S3 and Table S5).

Based on common densities for each polymer type and normalized FTIR results, an estimated 85 and 74% of observed plastic particles had estimated polymer densities greater than 1.1 g cm⁻³ for Lake Michigan and Lake Erie, respectively. Of those particles with polymer density greater than 1.1 g cm⁻³, 80 and 57% represent fibers/lines from Lake Michigan and Lake Erie, respectively (Figure 5A,B). The relative percentage of different polymer types varied by lake in these samples (Figure 5B).

4. DISCUSSION

The presence of microplastics in all sediment samples collected across Lake Michigan and Lake Erie demonstrates the ubiquitous presence of microplastics in the sediment of these two lakes. Concentrations in sediment from Lake Erie were greater overall compared to those from Lake Michigan for the common particle sizes analyzed (>0.355 mm). Contributing factors for this result could include the greater population density and urban and agricultural land cover in the Lake Erie basin, and collection of contributions from upstream areas that include the Detroit River and Lake St. Clair.

Microplastic particles have also been observed to be ubiquitous throughout tributaries from all Great Lakes^{51,59,62,63,77,78} and at the surface within all Great Lakes.^{53,55,56,61} These previous results established a difference in the distribution of particle types between water surface samples in tributaries compared with those from the open lakes: fibers were the dominant particle type in tributaries (45-100% of particles), 51,59,62,63,77,78 and fragments were dominant in lake water surface samples (42-97% of particles).^{53,55,56,61} Evidence that helps explain this difference was provided in a study of the vertical distribution of microplastics in Milwaukee, Wisconsin area streams, estuary, and Lake Michigan: Microplastic particles were present throughout the water column with polymer density increasing from the water surface to the water column to the sediment. Fibers were present throughout the water column and sediment, and fragments were more abundant at the water surface than in subsurface or sediment samples. Results from sediment samples in the current study support the notion that high-density microplastics (>1.1 g cm⁻³), such as PET fibers, observed at the water surface in tributaries tend to settle



Figure 2. Sediment sampling locations and concentrations by particle type and particle size fraction for sediment samples collected from Lake Erie. The black dots represent sampled locations, X indicates a location where bed material composition did not allow sample collection, major streams are indicated with blue lines, and state/provincial boundaries are shown with gray lines.^{71,72} Particles include the size fractions from 0.125 to 0.3549 mm (left bar) and >0.355 mm (right bar). (p kg⁻¹, particles per kilogram dry weight sediment; mm, millimeters; >, greater than).



Figure 3. (A) Mean concentrations and (B) percentages by size fraction and particle type for sediment samples collected in Lake Erie (12 samples) and Lake Michigan (20 samples). The smallest size fraction was only analyzed in Lake Erie samples. (n = number of samples; mm = millimeter; p kg⁻¹ = particles per kilogram; wt, weight).

through the water column and deposit in benthic lake sediment once reaching the more quiescent, low-turbulent, and deep waters of the Great Lakes.



Figure 4. Mean concentrations by particle type for all microplastics greater than 0.355 mm for Lake Erie and Lake Michigan. (n, number of samples collected; p kg⁻¹, particles per kilogram; wt, weight).

Consistent with the current study, a prevalence of fibers in different environmental settings has been observed in multiple studies from around the globe: stream water samples from eight countries, $^{51,62,79-87}$ lake water samples from four countries, $^{59,85,88-90}$ sediment samples from eight countries, $^{52,60,82,84,88,91-95}$ and atmospheric samples from four countries in addition to the Arctic $^{7,8,21,79,96-101}$ have documented that anthropogenic fibers are the most prevalent type of microplastic particle. Fibers can be primary and secondary microplastics, although they are predominantly thought of as secondary microplastics derived mainly from the breakdown of larger plastic material. Additionally, fibers can be composed of a diverse group of synthetic plastic polymers such as nylon or PET, natural cellulose, and/or artificial semisynthetic cellulose making fiber material challenging to

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Figure 5. (A) Sum of the estimated fraction of total particles and polymers by density. (B) Estimated fraction of total particles and polymers by lake, arranged by increasing density $(\rho)^{73-76}$ from top left to right within each color group. Only polymers that represented greater than 2% of particles collected are represented in individual polymer charts, and polystyrene (PS) and polycarbonate (PC) are not shown. PP = polypropylene; LDPE = low-density polyethylene; HDPE = high-density polyethylene; Nylon = nylon; PMMA = poly(methyl methacrylate); PAN = polyacrylonitrile; PVA = poly(vinyl acetate); PEG = poly(ethylene glycol); Azlon = azlon; PET = poly(ethylene terephthalate); PVC = poly(vinyl chloride); S.S. Cellulose = semisynthetic cellulose; unknown = polymer was not identified; (<, less than; >, greater than; ρ , density; g cm⁻³, grams per cubic centimeter).

define.^{80,101-103} Original sources of fibers can include microfibers used in manufacturing textiles, synthetic textiles (e.g., clothing, reusable bags/baskets, carpeting, window shades, towels, backpacks, parachutes, tents, nets, furniture, plastic teabags), cigarette filters, construction materials (e.g., fiber-reinforced cement composites, insulation) electrical components, and automotive components (e.g., airbags, upholstery, heavy-duty tires, cap ply for radial tires).^{104–107} Particles from these sources have multiple potential routes of delivery to the aquatic environment, including wastewater treatment plant (WWTP) effluent directly to water bodies,¹ treated biosolids (sewage sludge) application on land surfaces and subsequent runoff, 19,108 runoff from nonpoint sources,^{109,110} venting exhaust from laundry drying machines that can result in atmospheric transport,^{21,79,111} and atmospheric deposition directly to aquatic environments,^{21,99,100} and atmospheric deposition to land surfaces followed by runoff events.^{21,97,100} For example, WWTP facilities from around the United States contribute an estimated mean discharge of 4 million microplastic particles per facility per day, with fibers and fragments being the most prevalent in treatment plant effluent.¹⁹ Estimates of microplastics in sewage sludge have been reported from 15 385 p kg⁻¹ dry weight (dw) up to 34 000 p kg⁻¹ dw, and fibers were the most prevalent particle type.^{108,112} Research into atmospheric deposition of microplastics indicates that fibers and fragments are ubiquitous

within the atmosphere globally and the potential for redeposition (wet and dry) onto aquatic surfaces is high.^{7,8,96}

The range of microplastic concentrations in the current study from Lake Michigan was less than that from previous sediment studies of the Great Lakes basin, but previous studies have focused primarily on tributaries, estuaries, and nearshore environments that were closer to sources and not subject to dilution from larger undeveloped areas in the Great Lakes. In addition, previous studies have quantified microplastics into smaller size fractions (0.063 to >5.0 mm),⁵² (0.5–3.0 mm),⁵⁰ (0.063-2.0 mm),⁵⁸ (>0.125 mm),⁵⁹ and (0.063-2.0 mm)⁶⁰ than the current study from Lake Michigan (>0.355 mm). In contrast, concentrations and the size fractions used for quantifying microplastics from sediment samples collected from Lake Erie in the current study were more comparable to the results from previous samples^{50,52,58-60} collected from tributaries, and nearshore environments. This is likely a result of the greater influence of areas developed for urban and agricultural land use that contribute flow to Lake Erie, including flow from Lake Huron through the Detroit River, and direct drainage from the highly developed land surrounding Lake Erie. Like the current study, fibers and fragments have proven to be the most abundant particle types observed in these previous Great Lakes studies discussed above.

Much of the previous research on microplastics in the aquatic environment has focused on particle sizes greater than about 0.355 mm, and only a small number of studies have included smaller size fractions, although that number continues to increase. Many studies have enumerated microplastics particles in sizes greater than 0.355 mm. Of those particles, the concentrations in smaller size fractions (e.g., 0.355–1.0 mm) have commonly been greater than concentrations in larger size fractions; the percent of particles less than 1.0 mm in these studies ranged from 62-97% in sediment samples^{52,57,59,113,114} and from 59–93% in water samples.^{51,53,55,56,61} This pattern is maintained in studies where smaller sized particles were enumerated in water and in sediment samples. In the current study, the amount of microplastics in Lake Erie sediment samples with grain size 0.125-0.3549 mm was similar to the amount in the larger particle sizes (>0.355 mm) with median values of 309 and 312 p kg⁻¹, respectively. Similarly, concentrations of particles less than 0.125 mm in sediment samples from marine and freshwater environments in previous work were approximately equal to or greater than concentrations from size fractions greater than 0.125 mm.^{59,115-117} Results from surface water samples have indicated that concentrations of smaller particles may be even more prevalent; previous studies of marine waters, sampling with a mesh smaller than 100 μ m in size increased concentrations 1– 4 orders of magnitude over samples collected with a standard 333 μ m mesh.^{118–120} Similarly, studies of freshwater environments have shown higher concentrations when sampling with mesh sizes less than or equal to 100 μ m compared to the standard 333 μ m mesh.^{79,80,84,121} Collectively, these results have indicated smaller particles are at least as abundant as larger particles in sediment and in surface water, and this indicates that limiting analysis to size fractions greater than 0.355 mm can substantially underestimate overall microplastics concentrations in aquatic environments.

Identified polymers in more than 10% of the FTIR-analyzed particles were different depending on the lake. For example, 36% of the particles analyzed by FTIR in Lake Michigan were PET, while only 12% of particles from Lake Erie were PET. Similarly, 15% of FTIR-analyzed particles were HDPE in Lake Michigan compared to 1% in Lake Erie. Semisynthetic cellulose (S.S. Cellulose) and PP resulted in similar percentages between the lakes with 23 and 3% of FTIRanalyzed particles in Lake Michigan and 30 and 11% in Lake Erie, respectively. The variety of polymers observed supports the idea that "microplastics" are a suite of particles consisting of different chemical compounds with inherently different properties, including density, particle type, shape size, and chemical behavior. This information reinforces the notion that microplastics may be more accurately treated as a diverse group of emerging contaminants rather than one generic class of contaminants.¹

The relative abundances of different polymers in freshwater and marine environments have been observed to be vertically stratified with a diverse group of high-density polymers most abundant in the bottom sediment^{113,115,123–127} (including the current study), a combination of low- and high-density particles in the water column beneath the water surface,^{22,59,65} and low-density polymers most abundant at the water surface.^{55,61,121,128–130} The presence of polymers in these different aquatic compartments has not been exclusively limited in this manner: low-density particles such as polyethylene and PP ($\rho < 1.1 \text{ g cm}^{-3}$) have been observed in sediment, 50,52,58,59,115 and high-density polymers such as PVC and PET ($\rho > 1.1 \text{ g cm}^{-3}$) have been observed at the water surface. 59,85,90,121 Estimations of polymer density rely on values from the literature for the most common forms of each polymer. Density can vary from these values based on several factors. For example, manufacturing of plastics often includes addition of functional fillers such as calcium carbonate, silica, kaolin, talc, and mica or pigments such as copper(II) phthalocyanine that can change the density of polymers. 50,58 In addition, biofilm colonization has been observed on microplastics in the aquatic environment that changes the effective density of the particles. ${}^{131-134}$ These density modifications can cause microplastics with polymers of high or low density to appear in unexpected aquatic compartments.

With an abundance of evidence that microplastics are present in multiple environmental compartments from around the globe, and specifically in the Great Lakes basin, it is important to consider how this presence may influence ecological health.¹²² A review of published exposure studies over multiple levels of biological organization concluded that microplastics are causing ecological effects, and the assessment of these effects is complex with the nature and severity driven by dose, particle shape, polymer type, and particle size.¹³⁵ Observed effects are diverse and include multiple biological pathways with variable outcomes, such as reproductive disfunction, alteration of locomotion, intestinal damage, change in metabolic profiles, and mortality.¹³⁶ In addition, components of plastics including polymers and functional additives can have adverse effects,¹³⁶ and desorption of these components has been documented in biological organisms.¹³⁷⁻¹³⁹ Furthermore, microplastics can increase the concentration of harmful persistent organic pollutants (POPs) such as dichlorodiphenyldichloroethylene (DDE) and represent an understudied pathway for contamination among aquatic food-webs.¹⁴⁰ Even with the mounting evidence regarding adverse biological effects, bioassays with microplastics commonly use concentrations greater than those typically observed in environmental samples,¹³⁵ and have not been focused on exposure in benthic sediment. The current study provides information on microplastics particle size and type, concentration, and polymer composition in sediment of Lakes Michigan and Erie that can be used to assess potential for biological effects when sufficient data is available for such analysis.

Although research in the past decade has advanced the understanding of the behavior and effects of microplastics in the aquatic environment, there is still much to learn. There are many remaining gaps with respect to assessment of ecological effects. With regard to the presence and movement of microplastics, a more thorough understanding is needed for source characterization, atmospheric deposition, mass or particle loadings to and within receiving waters, fate and transport properties, and hydrodynamic modeling. In addition, understanding how exposure to microplastics may translate to a potential hazard for environmental organisms is not well understood. Additional relevant laboratory and field-based experiments for the complex matrix of environmentally appropriate microplastics characteristics are needed, including variable concentrations, size fractions, particle types, polymer composition, and organisms from multiple trophic levels. Further, a consistent approach for defining microplastics in the aquatic environment would allow comparability among studies, including field sampling as well as laboratory methods. Field

methods that would benefit from standardized methods include sampling at the surface, in the subsurface, and in sediment of rivers, estuaries, and lakes. Laboratory methods in need of standardization include enumeration, size fractions, classification of particle types (morphology and shape), and identification of polymer composition.

The current study fills an important gap in information for the Great Lakes system by quantifying microplastics in benthic sediment from the offshore lake environment. A multidimensional dataset was developed for evaluation of the spatial distribution of microplastics in benthic sediments from Lake Michigan and Lake Erie including concentrations, geographic distribution, particle types, particle sizes, and identified polymers. Lake Erie microplastic concentrations were greater than those observed for Lake Michigan for the comparable size fractions between the two lakes. There were similar amounts of particles in the smaller size fraction analyzed in Lake Erie compared to the large size fraction, emphasizing the importance of quantifying smaller particle sizes for a more thorough representation of microplastics. The observed tendency toward high-density particles and a majority percentage of fibers compared to other particles in sediment samples provides information that can be used in a future study of fate and transport of microplastics particles. These results indicate a plausible explanation for the difference in fiber abundance in tributaries that were not present at the water surface in the Great Lakes.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.est.0c06087.

Sample locations and characteristics for each location sampled, sediment processing methodology, total number of microplastic particles collected and total number of particles analyzed using FTIR table, duplicate environmental sample relative percent difference comparison of results, relative percent difference table, field environmental duplicate particle results figures, the number of particles collected by size fraction table, percentage of different polymers identified from FTIR analysis, and polymers identified for particles analyzed with FTIR by sample location and particle type (PDF)

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The manuscript was written with contributions from all authors. All authors have given approval to the final version of the manuscript.

Notes

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