Testimony on

Plastic Pollution in the Great Lakes

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Before the

Subcommittee on Oceans, Atmosphere, Fisheries, and Coast Guard
U.S. Senate
115th Congress of the United States of America

Hearing on the
Efforts on Marine Debris in the Oceans and Great Lakes
Russell Senate Office Building, Room 253, Washington, D.C.

Tuesday, July 25, 2017

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The findings and perspectives presented in this testimony represent the author's own professional assessment as an independent academic researcher. They should not be taken to reflect the views of the University of Michigan, the author's past affiliations, or funders present or past.

SUMMARY STATEMENT

I wish to thank Chairman Sullivan, and my Michigan Senator, Ranking Member Peters, as well as the members of the Subcommittee for inviting me to today's hearing. As a representative of the basic research community, I appreciate being at this table and part of these discussions.

My name is Melissa Duhaime and I am an assistant professor at the University of Michigan in the Department of Ecology and Evolutionary Biology. I studied biology at Cornell University and hold a doctorate from the Max Planck Institute for Marine Microbiology in Germany.

I have worked in ocean and freshwater sciences for over a decade, studying plastics across the world's oceans, and most extensively in the Great Lakes, where I began my career in Michigan 5 years ago--in fact, that time marked the very inception of this young research field.

Plastic hit the consumer market after WW2, when the economics of this cheap good and the convenience of a throw away culture took off. 60% of plastic ever produced--5 billion tons--still remains in landfills or dispersed in the environment today. This is equivalent to 10 times the biomass of all humans on Earth. For each of us in this room, there are 10 of us made of plastic out there. Each year, 5-13 million tons of plastic enter the oceans. These numbers will continue to rise the global production of plastic goods continues to increase exponentially.

The trends are no different in the Great Lakes.

In 2014, we carried out the largest survey to date of Great Lakes surface plastic pollution, traversing Lakes Superior, Huron, St. Clair, and Erie. We collected surface-floating plastic down to one-tenth of a millimeter. We found plastic at every site sampled. The sample with the highest total concentration of plastic (in the Detroit River) contained almost 2 million particles per km, 4-times higher than yet reported in the surface of the Great Lakes.

The highest concentrations of plastic were found near populated Great Lakes cities, in river plumes, directly at the effluent of wastewater treatment plants, and following storm events.

As with all plastic pollution, the smallest plastics dominated all samples. Given this trend, it is essential that more attention be paid to the smallest size classes of plastic, especially the nanoscale, of which we know near-nothing about, but whose health risks will be highest.

The vast majority of plastic detected with secondary plastic fragments, broken down from larger pieces—not the microbeads reported to dominate in the first study of Great Lakes plastic.

Plastic floating in water serves as sponges of toxic persistent organic pollutants (or "POPs") that are consumed when plastics are. Two carcinogens, polyaromatic hydrocarbons (PAHs) and polychlorinated bisphenyls (PCBs), were detected on plastic from Lake St Clair, the Detroit River plume, and Cleveland WWTP effluent. Also, antibiotics, herbicides, fungicides, and insecticides have been detected on plastic in Lake Erie. The implications of these of findings have not yet been explored.

In a U-M study of fish and mussels collected from the Great Lakes, roughly one-quarter of all Great Lakes fishes and one-third of bivalves examined contained plastic fibers in their stomachs.

In laboratory studies, Lake Michigan Quagga mussels and Chironomid worms consume nano-sized plastic, mistaking them for food. These organisms, especially the worms, are central to the Great Lakes food web. They are a food source for all the foraging fish, which are then consumed by greater "fish-eating fishes", such as salmon, trout, bass, and walleye.

Research is needed to define the effects of consumption and to determine the economic and public health impacts of plastic pollution in the Great Lakes.

In summary, basic research has shown the plastic is everywhere, in all oceans on the planet, remote alpine lakes, in the Great Lakes, and in beer and fish sold for human consumption. It is near certain that humans are consuming plastic.

In the wake of these discoveries, the United Nations has declared plastic pollution among the most critical emerging environmental issues of our time. The scientific consensus is that plastic pollution must be reduced to avoid the risk of irreversible ecosystem harm.

The direct human health consequences of plastic pollution are unknown, but this is the essential frontier of basic research.

As put by environmental toxicologist, David Sedlak, "Although we are all responsible for microplastics in the environment, getting the entire world to rethink the way it uses synthetic polymers would be a long, arduous process requiring compelling evidence of severe environmental risks."

Basic research is critical to our ability to understand the extent and implications of this issue. I look forward to sharing future findings with you and continuing to be a resource to the Committee. I look forward to your questions now and in the future. Thank you.

Plastic Pollution in the Great Lakes

I. INTRODUCTION

The accumulation of plastic debris in nature is "one of the most ubiquitous and long-lasting recent changes to the surface of our planet" [1]. Since plastic hit the consumer markets in the 1950s, 60% of plastic produced—4.9 billion metric tons—still remains in landfills or is inadvertently dispersed in the environment[a]. That is 10 times more than the biomass of humans on the planet. Each year, **5-13 million tons of plastic find its way into our oceans** [2]. In the absence of mechanisms to incentivize improved waste management and behavior change, this number will continue to rise, reflecting the **exponentially increasing global production of plastic goods** [3].

Aquatic organisms ingest plastic pollutants [4,5], which results in energetic and fitness costs [6,7] and other morbid impacts [8]. Microscopic plastic is found in fish and shellfish sold for human consumption at seafood markets around the world, including in Europe [9] and in the U.S. [10]. There is a high likelihood that humans are consuming this plastic. The health consequences of this are unknown.

In the wake of these discoveries, the **United Nations has declared plastic pollution among the most critical emerging environmental issues of our time** [11]. The scientific consensus is that plastic pollution must be reduced to avoid the risk of irreversible ecosystem harm [12].

While most research has focused on the distribution and impacts of *marine* litter, most plastic pollution originates on land [13]. As such, **freshwater bodies serve as conduits for the transport of plastic litter to the ocean**. Humans live in close contact with freshwater. 90% of the world's population lives 6 miles from a freshwater body [14].

Recently, **plastic has been documented in the Great Lakes at some of the highest concentrations seen on the planet**. Yet, too little is known about the fate of this plastic and its role in ecosystem dynamics to assess environmental risk and predict the impacts on one fifth of the world's surface freshwater and arguably one of our most valuable national security assets.

This discussion focuses on recent findings led by our team at the University of Michigan regarding plastic pollution in the Great Lakes. It (1) reports the quantification, distribution, and modeled transport of Great Lakes plastic debris, (2) describes the carcinogenic toxins that hitch a ride on Great Lakes plastic, (3) demonstrates that organisms central to the Great Lakes food web consume plastic, and (4) explores new frontiers in the detection of nano-sized plastic. The report concludes by highlighting recommendations for future research directions. These aim at

[a] https://www.nytimes.com/2017/07/19/climate/plastic-pollution-study-science-advances.html?mcubz=0

addressing current knowledge gaps in our ability to assess environmental risks of this pervasive, persistent pollutant—in the Great Lakes and beyond.

II. Plastic Pollution in the Great Lakes

In 2014, we carried out the largest survey to date of Great Lakes surface plastic pollution, quantifying plastic in over 100 samples collected across Lakes Superior, Huron, St. Clair and Erie [15]. With funds from the University of Michigan Water Center and Erb Family foundation, as well as a generous donation of time, research vessel, and fuel by citizen scientist, David Brooks (resident of Chelsea, MI), we traversed these lakes and collected surface-floating plastic down to 100 μm—one-tenth of a millimeter, smaller than a period on this page.

We have **worked for four years with NOAA's Marine Debris Program to develop an Action Plan for the Great Lakes**. The Great Lakes plastic research community is incredibly collaborative and connected, in large part due to the organizing efforts of NOAA's Marine Debris Program in the region. I have worked with the International Joint Commission to establish recommendations on how to address the problem of plastic pollution in our Great Lakes. Our data have contributed to follow-up research programs and private funding, remediation action plans, and new knowledge disseminated to the public through outreach initiatives around the Great Lakes. Our work has been published in peer-reviewed journals [15-17] and key elements are summarized below.

A. Abundance and Distribution

While floating plastic bottles and bags, styrofoam coolers, straws, old tires, and cigarette butts disrupt our intrinsic connection with "pristine" natural spaces, most Great Lakes plastic is small, nearly invisible "microplastic" (<5 millimeters in size).

Figure 1. A calm and seemingly clean Lake Erie (left), photo credit, Melissa Freeland; particles collected following a storm event from the surface of Lake Erie at the Cleveland wastewater treatment plant effluent site (right), many of which proved to be "microplastic" (defined as plastic <5 mm in size).

What we collected in our field survey were not the pristine samples we had collected previously across the world's oceans, which consisted primarily of plastic and little else. Rather, with each surface trawl, we pulled up pounds and pounds of biomass—such as algae, insect larvae, sticks, and leaf litter. Enmeshed in this was microscale plastic trash (Figure 2).

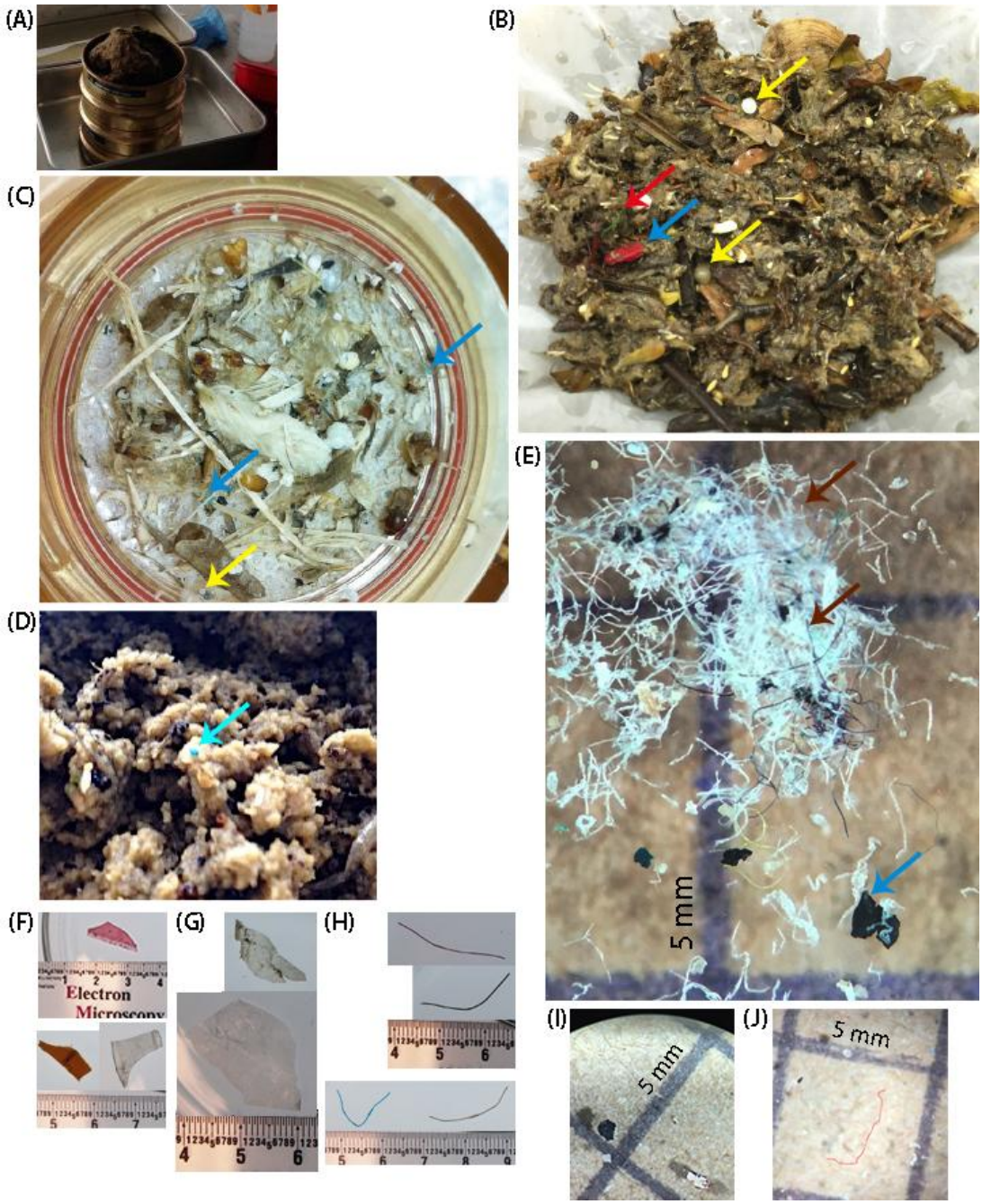

Figure 2. From [15]. Samples from Great Lakes plastic survey of 2014 at various stages of processing, including examples of different shape classes. Arrows indicate plastic amidst co-sampled nonplastic organic matter; blue: fragment, dark red: line, yellow: nurdles, cyan: sphere/bead, brown: fiber. **(A)** Bulk sample directly upon retrieval from surface net on a stack of a series of sieves. This sample contained an abundance of algal biomass. **(B)** Bulk sample drying on a 53 µm mesh net. **(C)** Sample after enzymatic processing, which included an incubation in hydrogen peroxide that bleached much of the non-plastic organic matter. This bleaching aided in differentiating plastic (tended to retain color) from non-plastic (prone to bleaching) particles. **(D)** Examples of plastic of sphere class; zoomed in subset of sample in **(B)**. **(E)** Smallest size fraction (106–1,000 µm) after hydrogen peroxide treatment. Note colored plastic fibers (brown arrows) enmeshed in mass of natural fibers bleached white from hydrogen peroxide treatment. **(F–H)** Examples of plastic of fragment, film and line shape classes, respectively; ruler markings are in cm units. **(J,I)** Examples of plastic of paint chip and fiber shape classes, respectively; grid squares are in 5 mm units.

We found plastic at every site sampled in this Great Lakes study (Figure 3). The sample with the highest total concentration of plastic (in the Detroit River) contained almost 2 million particles km^{-2}, a **4-fold higher concentration than yet reported in the surface of the Great Lakes** [18,19].

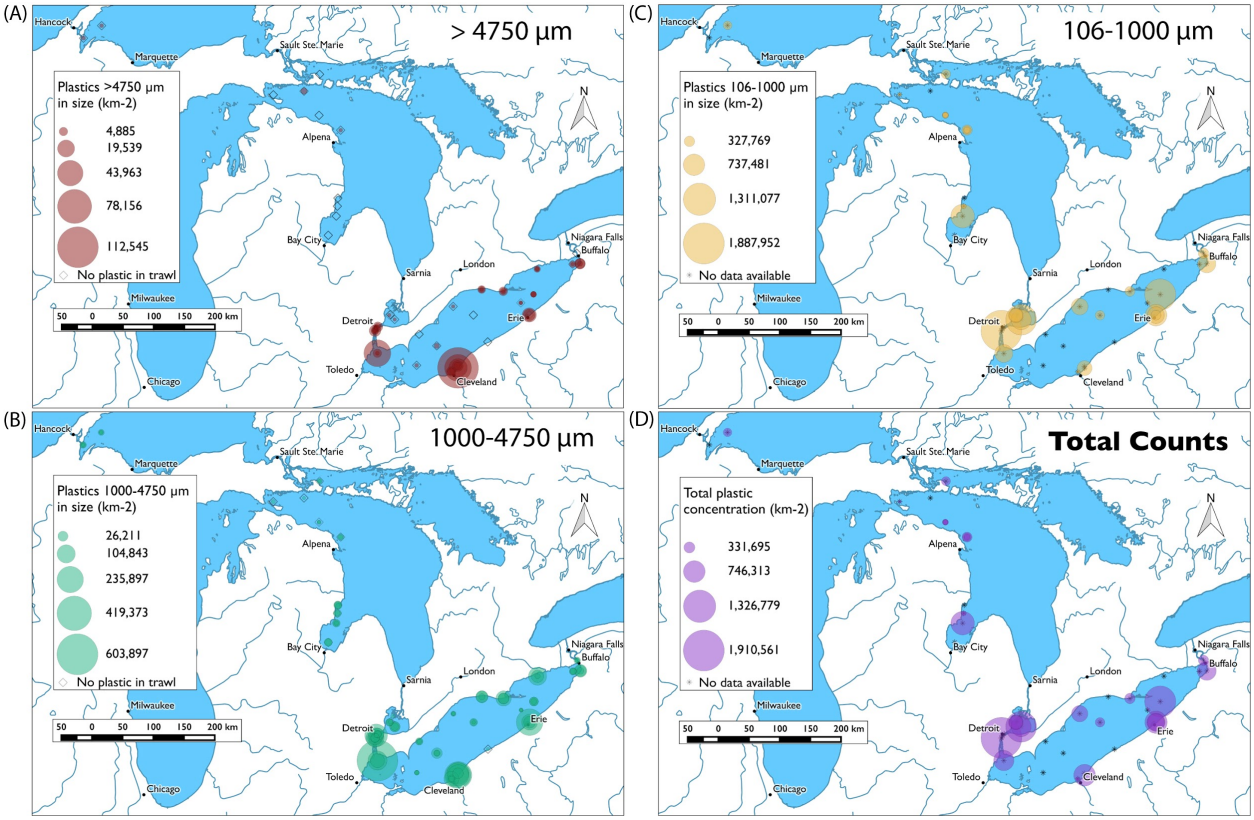

Figure 3. Maps of plastic concentrations across the lakes sampled; magnitude of concentration is depicted by size of circle around trawl location. Note, fiber counts are not included in these figures, as their quantification is error prone. **(A)** Mapped counts of plastic litter >4,750 µm. **(B)** Mapped counts of plastic litter 1,000–4,750 µm. **(C)** Mapped counts of plastic litter 106–1,000 µm. **(D)** Total mapped counts for the stations where all three size classes were quantified.

Across our Great Lakes study and in nearly all studies to date, the smallest plastics dominate. The vast majority of plastic counted was <1 mm in size (Figure 4A), regardless of water body or types of stations sampled. Smaller plastic particles stay at the water surface longer than larger particles of the same composition and shape [20,21] and are more readily consumed by smaller organisms in aquatic food webs [22]. The larger surface area to volume ratios of these small plastics increases their potential to deliver toxic chemicals (discussed below) to the organisms that consume them [1,23]. Given this trend, **it is essential that future studies document sub-millimeter (nanoscale) plastics and develop innovative high-throughput solutions to capture and quantify nanoscale plastics. The ecosystem risks of nanoscale plastics may be highest** due to subcellular effects [24]—but, due to technical limitations, they have yet to be identified or quantified in natural systems. We have begun addressing this issue (see section on Organismal Impacts, below).

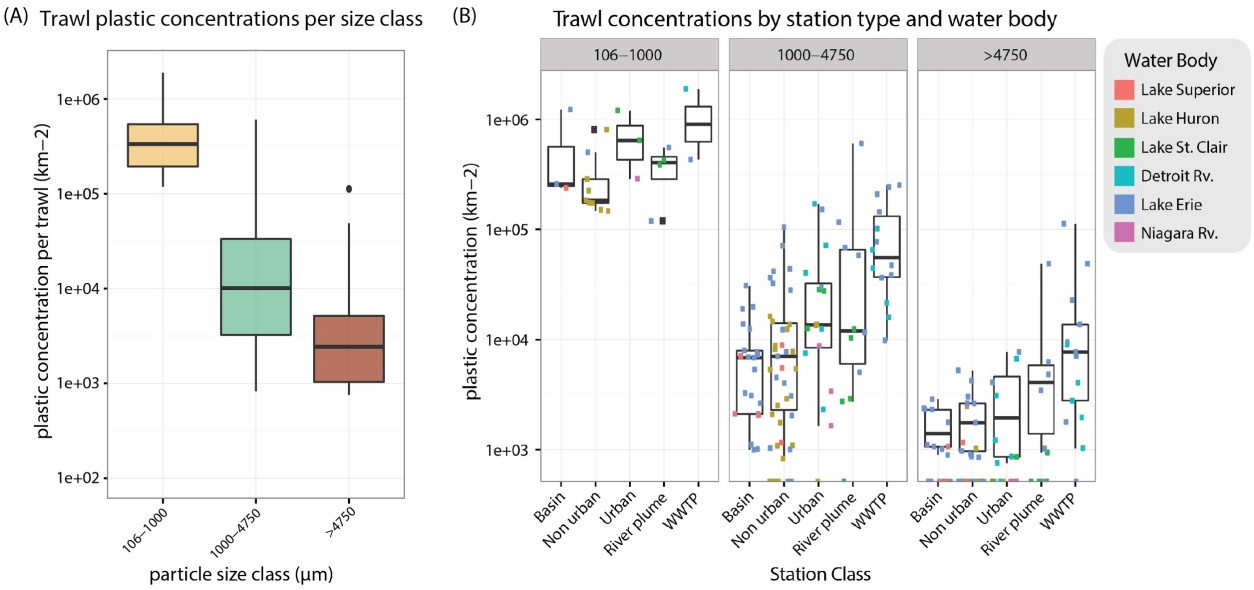

Figure 4. (A) Boxplots depicting the means and spreads of plastic counts by particle size class (from smallest to largest, left to right). **(B)** Boxplots depicting the means and spreads of plastic counts by size class, station type, and water body: Lake Superior, Lake Huron , Lake St. Clair, the Detroit River, Lake Erie, and the Niagara River.

The **highest concentrations of plastic were found near populated urban cities, in river plumes, directly at the effluent of wastewater treatment plants** (Figures 3-4), **and following storm events**. The Cleveland, OH, sample was collected at a WWTP effluent site immediately following a massive rainstorm (Figure 1, right panel; Figure 3A). We suspect we captured a combined sewage overflow event, whereby plastic in runoff that bypassed the treatment plant was delivered to the lake with no treatment.

Overall, these findings support previous reports of a correlation between plastic concentrations and proximity to urban centers in the Great Lakes [25]. Attributes that are likely to contribute to

elevated plastic concentrations in urban vs. non-urban locales include higher population densities [2], increased atmospheric inputs (including plastic; [26]), and increased areas of impervious substrate [25]. Increasing the degree of *pervious* substrate in watersheds, such as the **implementation of green infrastructure catchments, should be explored as an effective measure to capture plastic debris** in runoff and to reduce loads to waterways. As the number of storm events is expected to increase with a changing climate [27], such innovations are timely to buffer preventatively our freshwater systems from being inundated with stormwater-delivered debris.

Most Great Lakes plastic appears to be "secondary microplastics" broken down from larger pieces of debris (Fig. 5). This counters the first report of plastic from the Great Lakes that reported the majority to be in the form of spherical plastic microbeads [18], which have since been banned from rinse-off cosmetics [28].

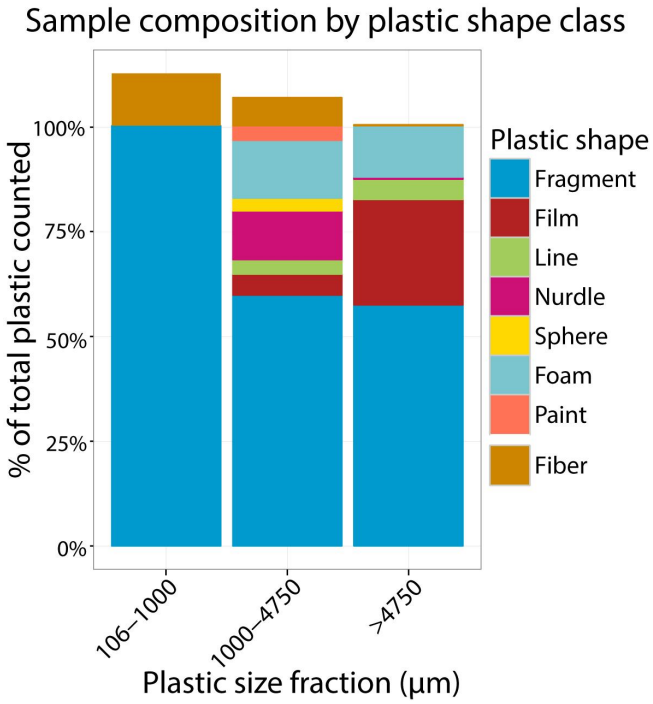

Figure 5. Stacked barplot depicting the relative abundances of different shape classes amongst plastic from each size class. The bar to 100% for each size class represent the relative abundance of different shape classes when fibers were not included in the total counts; the portion above 100% represents the relative abundance of fibers in the total counts.

Our Great Lakes study was the first survey of freshwater plastic litter to address variability in counts by conducting replicate trawls at each of 38 stations. With this replication, we were able to determine that the accuracy of a single trawl at one station was quite low. Repeated trawls at the same location can vary in precision by up to 3-fold. Evidence suggests that this variability is due to undersampling. In other words, to get reliable data, we must sample multiple times at each site and each sample must be larger.

Yet, across this field of research, replication is nearly never performed due to the massive investment that would be needed for data collection. Currently the most common method for quantification of plastic depends near-exclusively on visual sorting and counting.

Analytical approaches have been employed that rely on spectroscopic techniques (e.g., fourier transform infrared spectroscopys—FTIR, Raman spectroscopy) to confirm whether particles are known synthetic polymers. But as of yet, these approaches are low-throughput and are limited by our inability to identify complex (often proprietary) mixtures of polymers and dyes outside the standard known polymer classes.

The **development of analytical techniques for high throughput, high confidence plastic counts is critically needed**. Such advancements will pave the way for accelerated data collection, down to nano-sized particle classes, and will drastically improve the reliability and value of future data generated.

B. Modeled Transport

In the absence of an inexpensive, rapid, and accurate method to quantify plastic debris on large temporal and spatial scales, hydrodynamic models were applied to predict the plastic distribution and transport of plastic in one of the Great Lakes, Lake Erie (D. Beletsky, R. Beletsky; U-M Cooperative Institute for Great Lakes Research; NOAA Great Lakes Environmental Research Labs; Ann Arbor, MI).

Our plastic transport model predicted habitats along the southern coast of Lake Erie to be most affected by plastic pollution (Figure 6) [15].

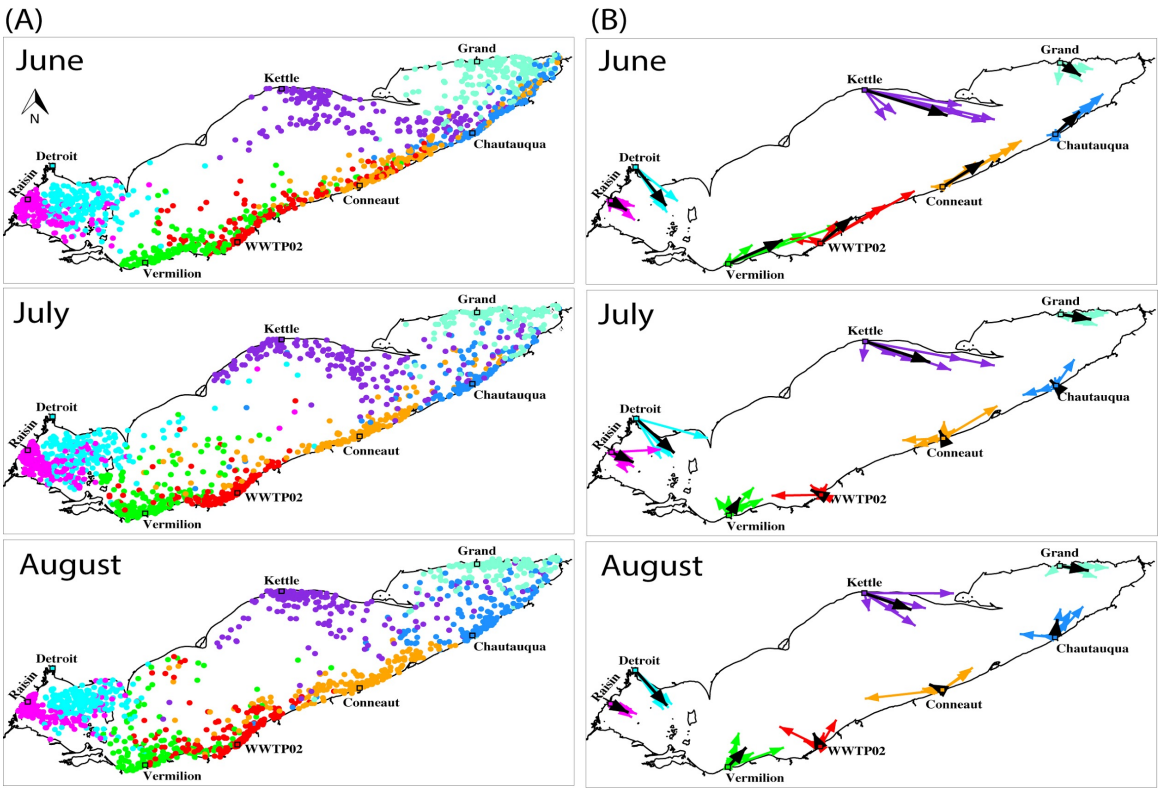

Figure 6. (A) The modeled distribution of neutrally buoyant particles in Lake Erie at the end of month-long simulated transport in June, July, and August for 6 years. For visual simplicity, 8 of the 29 sources (influents) are depicted: the Raisin Rv. (magenta), Detroit Rv. (cyan), Kettle Rv. (purple), Grand Rv. (turquoise), Chautauqua Rv. (blue), Conneaut Rv. (orange), Cleveland WWTP (red), and Vermillion Rv. (green). **(B)** Mean transport vectors summarizing the positions of all particles at the end of month at each of the same eight representative sources (similarly colored coded). The six vectors per source represent mean transport for each of the 6 years. The 6-year mean vector is shown in black at each input.

In most months, rather than moving offshore, the model predicted longshore transport from coastal sources (Figure 6A). This **model indicates that future plastic pollution mitigation and management efforts in Lake Erie should focus on its southern shore and downstream of urbanized areas**. Extending this plastic transport model to the other four Great Lakes will similarly inform future efforts across this critical watershed.

C. Plastic-adsorbed Toxins

Plastic floating in water serve as veritable sponges of toxic persistent organic pollutants (POPs). Plastic additives leach from plastics as they degrade (e.g., phthalates, BPA), induce toxic effects in aquatic organisms [29], and bioaccumulate in plastic-ingesting organisms [4,7] with unknown consequences.

Two carcinogens, polyaromatic hydrocarbons (PAHs) and polychlorinated bisphenyls (PCBs), were detected on plastic samples collected from Lake St Clair, the Detroit River plume, and

Cleveland WWTP effluent. PAHs were detected on plastics at concentrations ranging from 3500-17,000 ng/g; PCBs ranged from 4-99 ng/g (L Rios Mendoza; U-W Superior). The levels of PAHs measured on individual pieces of surface-floating plastic are 10 to 100 times higher than concentrations considered hazardous to sediment-dwelling organisms (6-150 ng/g[b]). Concentrations of PCBs measured on plastic are on the order measured in plankton in the Great Lakes [Hornbuckle 2006]. Both PAHs and PCBs bioaccumulate with the potential to biomagnify, meaning that due to their persistence in the environment and the inability of some organisms to metabolize the compounds, toxins can be passed to consumers in prey. Biomagnification happens across the food web for PCBs and only in low levels (algae and lower invertebrates) for PAHs. This results in concentrations of PCBs in apex predators at the top of the food chain higher than would be expected based on transfer from water alone.

Beyond the suite of POP toxins most plastic researchers screen for, researchers at the University of Michigan conducted the first survey of non-target toxins on plastics in the Great Lakes. **Antibiotics, herbicides, fungicides, and insecticides were identified on plastic in Lake Erie** (K Wigginton; U-M Civil and Environmental Engineering). The implications of these of findings have not yet been explored.

D. Organismal and Food Web Impacts

In a study of fish and mussels collected from the Great lakes, roughly one-quarter of all Great Lakes fishes and one-third of bivalves examined contained plastic fibers in their stomach contents (Larissa Sano, University of Michigan). Of the particles documented in the fishes, 100% were fibers. **A systematic survey of the incidence and population-level impacts of consumption of micro- and nanoplastics across the Great Lakes biota is needed**.

In collaboration with the Banaszak Holl Lab at the University of Michigan and the San Francisco Estuary Institute, with funds from the Gordon and Betty Moore Foundation and NSF-REU program, we have **developed and applied a new method to identify nanoscale plastic pollution** (Figure 7). This method combines atomic force microscopy (AFM) with infrared spectroscopy (IR) create infrared spectra of individual micro- and nanoplastics at the individual particle-level .

[b] http://www.ukmarinesac.org.uk/activities/water-quality/wq8_40.htm

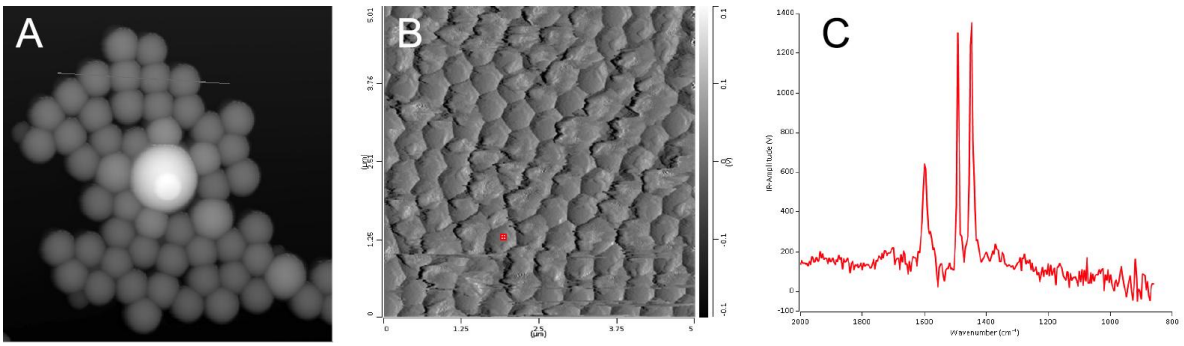

Figure 7. (A) A monolayer of beads visualized using AFM. (B) Red dot indicates polystyrene bead from which spectrum generated in panel C was obtained. (C) IR spectrum indicating characteristic peaks of polystyrene at 1452 cm^{-1} and 1492 cm^{-1}. Data generated by Rachel Merzel, Banaszak Holl Lab (University of Michigan).

We have confirmed the uptake of nanoplastics by Great Lakes filter feeders, a first step in defining the impact of their consumption on the Great Lakes food web.

Quagga mussels collected from Lake Michigan were fed fluorescently dyed nanoplastics the same size and at roughly the same concentration as their algal food source (0.01 and 0.1 picomolar; Figure 8). The mussels ingested the nanoplastic in a manner analogous to food consumption. The patterns observed in the gill tissue (Figure 8C) follow those of normal food accumulation, moving from the gills to the intestines. Mussels have internal mechanisms to reject particles they do not intend to digest. These data suggest the **nanoplastics are not rejected by Lake Michigan Quagga mussels, but rather are mistaken for food**. When smaller beads were used (200 nm), they also were observed in the gills and digestive tract. The Banaszak Holl lab will confirm whether such small beads are able to pass directly across cell membranes, which would pose a more lethal threat.

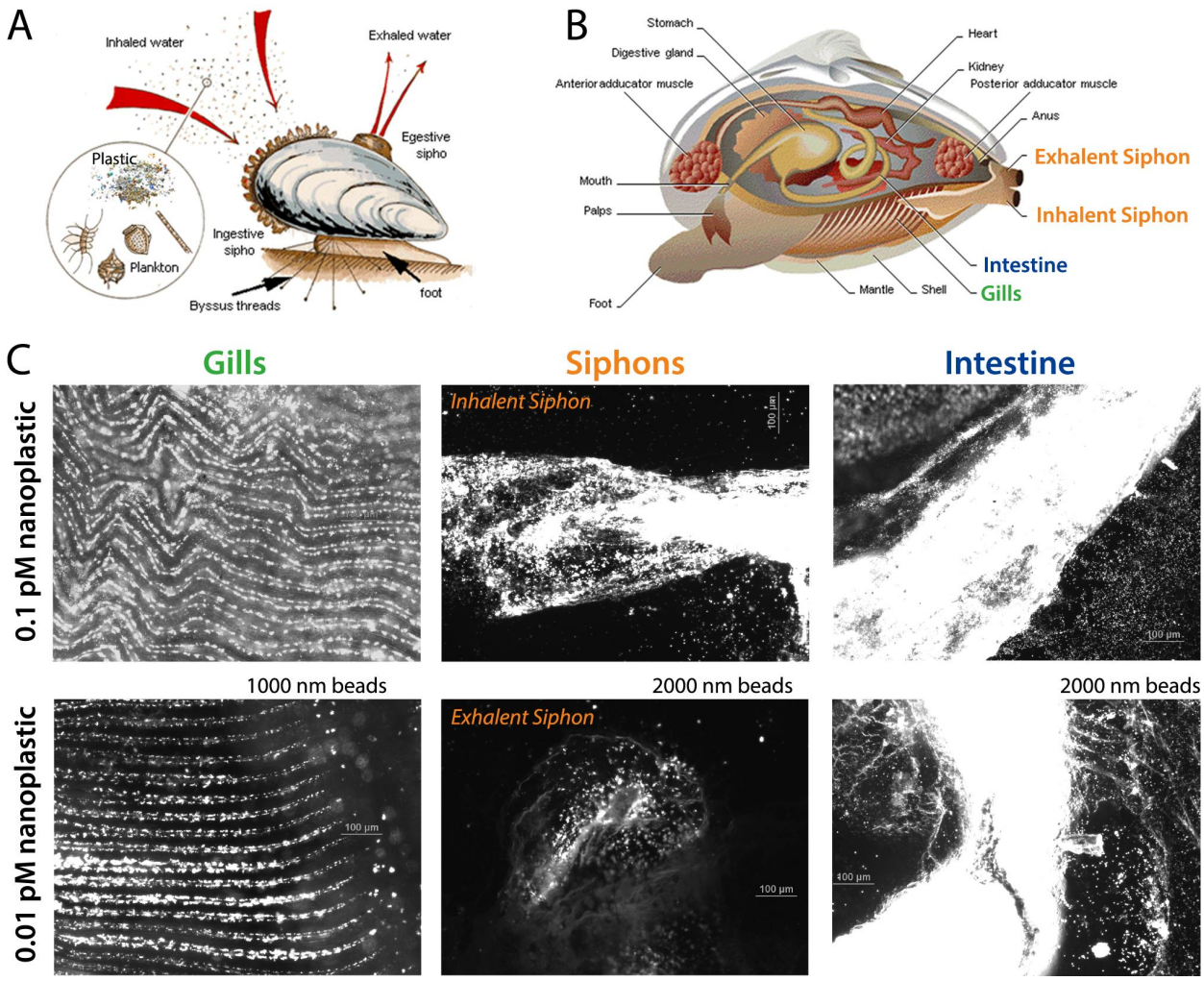

Figure 8. (A) Image[c] of mussel filter feeding. Plastic and food (plankton) enter the mussel in inhaled water, waste exits in exhaled water. (B) Diagram[d] of mussel anatomy. Note gills, inhalent and exhalent siphons, and intestines. (C) Microscopy images of internal structures of Lake Michigan Quagga mussels after being fed their algal food source along with 0.1 picomolar (top) and 0.01 picomolar (bottom) fluorescent plastic spheres. Plastic particles are the bright white elements of the image. Images from Lauren Purser, Banaszak Holl Lab (University of Michigan, NSF-REU). Recently collected data from currently unpublished work.

Benthic **Chironomid worms that live in the Lake Michigan sediment with the mussels also ingest the 200 nm and 2000 nm nanoplastics** and at concentrations greater than those observed in the mussels (Figure 9).

[c] http://www.molluscs.at/bivalvia/index.html?/bivalvia/main.html
[d] https://7salemanimalkingdom.wikispaces.com/Mollusks

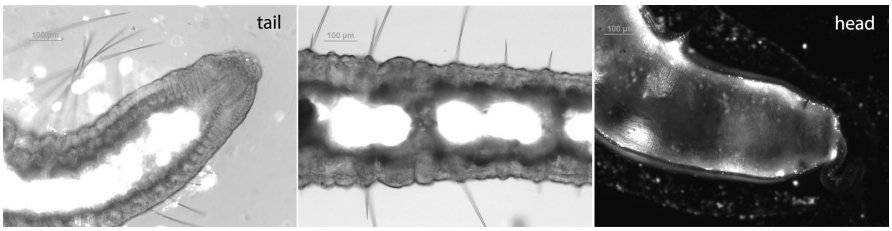

Figure 9. (A) Microscopy images of Lake Michigan Chironomid worms in tank with Quagga mussels exposed to fluorescent plastic spheres. Plastic particles are the bright white elements of the image. Images from Lauren Purser, Banaszak Holl Lab (University of Michigan, NSF-REU). Recently collected data from currently unpublished work.

Chironomids, as well as Quagga mussels, are central to the Lake Michigan food web. They are consumed by all foraging fish that live in the lake (Figure 10)—and, in fact, most of the Great Lakes. Trophic transfer of consumer plastic has been confirmed [31]. As such, owing to their resistance to degradation, **nanoplastics consumed by these Great Lakes mussels and worms have the potential to move up the Great Lakes food web to the high value piscivorian fishes ("fish-eating fishes"), such as salmon, trout, bass, and walleye** (Figure 10).

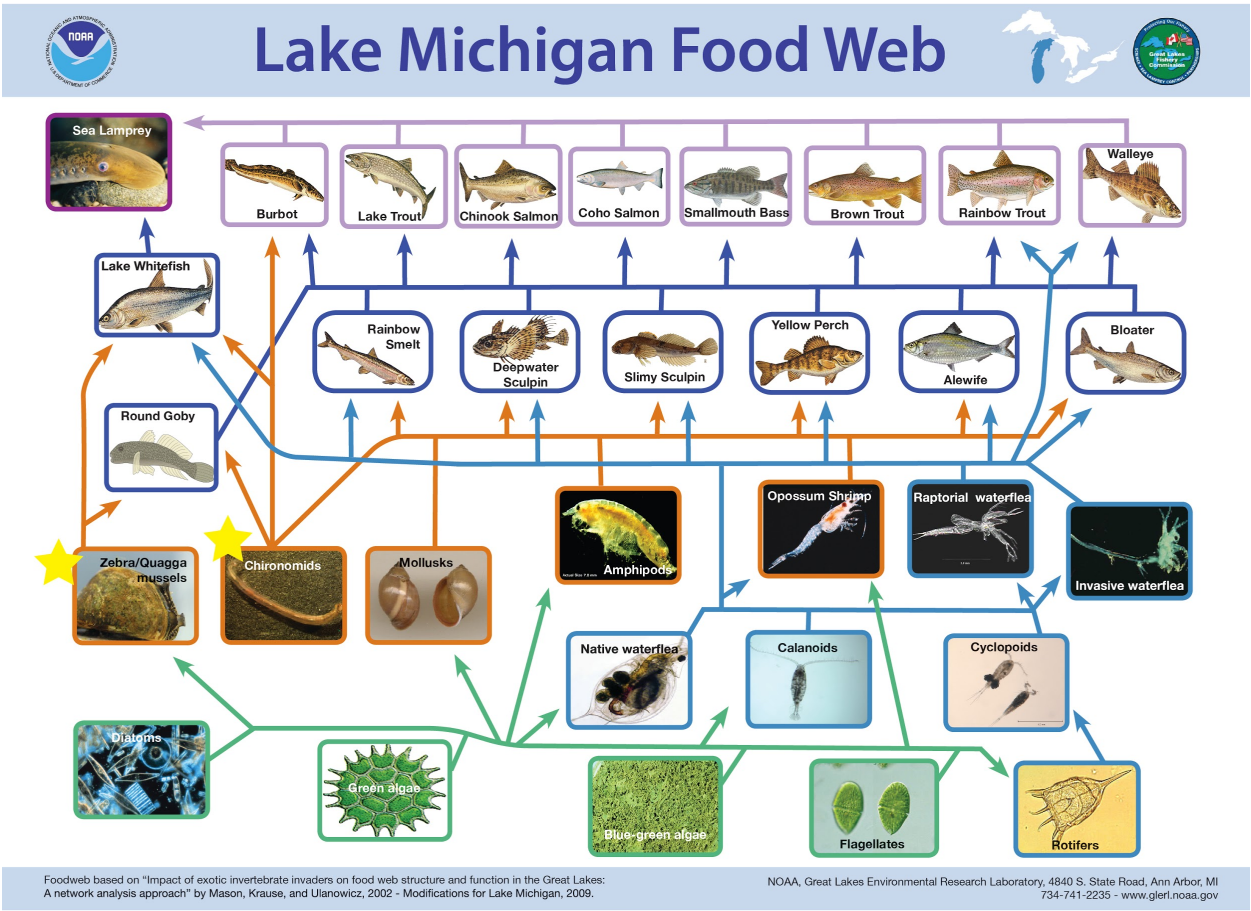

Figure 10. Lake Michigan food web. Prepared by NOAA Great Lakes Environmental Research Laboratory[e]. Note the yellow stars indicating the Quagga mussels and Chironomid worms highlighted in the research shared above.

Other researchers have confirmed that **ingesting plastic in place of food results in reduced biomass**; plastic lacks nutrients for growth [7]. Ingested plastic nanoparticles have led to **changed foraging behavior and organ function in fish** [32]. It is yet to be confirmed what the effects of plastic consumption are on the population-level fitness of Great Lakes fishes. This work is needed to determine the economic and public health impacts of plastic pollution in the Great Lakes.

III. CONCLUSION

As the largest freshwater system on the planet, the Great Lakes hold 20% of the world's surface freshwater. With this study, plastic pollution has now been documented down to the smallest size class reported to date. This led to the discovery of plastic concentrations up to **2 million particles per square kilometer, the highest reported levels in the Great Lakes and possibly any surface water ecosystem**. These high numbers can be attributed to high nearshore population densities, a feature unique to inland waterways that does not similarly influence remote ocean basins, and the long hydraulic residence time of some of the Great Lakes (3–100s of years, depending on the lake). Given this time and the recalcitrance of plastic to degradation, **fragments of some of the first plastic ever produced for the consumer market are certainly present in the Great Lakes still today**. This scenario is likely representative of lakes worldwide, which account for 87% of the planet's surface freshwater and have an average residence time of 50–100 years[f]—indeed spanning the introduction of plastics to the consumer market.

We know plastic is there in our critical freshwater. What is next? "Although we are all responsible for microplastics in the environment, getting the entire world to rethink the way it uses synthetic polymers would be a long, arduous process requiring compelling evidence of severe environmental risks (D. Sedlak [33], included with this report)." Critical to this process and the advancement of this research field are (1) the development of analytical techniques for high-throughput, accurate detection and quantification of micro- and nano-plastic, (2) development of hydrodynamic models to guide (3) targeted research surveys and experiments, to develop (4) a global plastic mass balance transport model (*"Where does it comes from? Where does it go?"*), (5) determination of food web impacts, and ultimately (6) the risk to humans. These research outputs will define further the ecosystem and public health risks plastic pollution pose to our vital freshwater systems and inform the needed policy, mitigation, and prevention initiatives of the future.

[e] https://www.glerl.noaa.gov/res/projects/food_web/food_web.html
[f] http://journal.frontiersin.org/article/10.3389/fenvs.2017.00045/full#note4

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ADDENDUM 1: Recent editorial on "lessons learned from plastic reearch" by David Sedlak, Editor-in-Chief of Environmental Science and Technology, the premier environmental science journal with focus on emerging contaminants of concern. Sedlak, D. Three Lessons for the Microplastics Voyage. *Environ Sci Technol* **51,** 7747-7748 (2017).

ADDENDUM 2: Publication from University of Michigan that culminated from the bulk of the results discussed in above report. Cable, R., Beletsky, D., Beletsky, R., Locke, B. W., *et al.* Distribution and modeled transport of plastic pollution in the Great Lakes, the worlds largest freshwater resource. *Frontiers in Environmental Science* **5,** 40 (2017).

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Three Lessons for the Microplastics Voyage

Whether it is DDT, perchlorate, perfluoroalkyl substances, or pharmaceuticals, the process through which a contaminant emerges follows a predictable pattern. First, researchers stumble upon a previously unknown contaminant or observe effects on the health of humans or wildlife that they cannot readily explain. Driven by curiosity and a desire to protect the environment, the researchers, operating on a shoestring budget, publish a paper documenting their initial findings. The attention that their research receives results in a wave of papers on detection, occurrence and toxicology of a now-emerging contaminant.

About a decade after the first wave of papers appears the emerging contaminant reaches a crossroads. If the research does not seem to justify action, the funding tide ebbs and the community moves onto other issues. But if there is sufficient ground for concern, a second wave of research starts, with an expansion into policy-relevant questions related to establishing regulatory standards, implementing treatment technologies, and reformulating products to minimize future releases.

Microplastics are our newest emerging contaminant. Although scientists have expressed concerns about the impacts of plastic pollution for over four decades, microplastics did not become emerging contaminants until 2007. The issue gained momentum about five years later, when researchers reported the presence of microbeads from consumer products in wastewater effluent-receiving waters. Facing negative publicity for a nonessential ingredient, leading manufacturers voluntarily eliminated microbeads and accepted the decision to ban them in the United States in 2015. Now that we are into the second wave of research that will determine whether or not the remaining sources of microplastics will be controlled, it is worth considering lessons learned from other emerging contaminants.

The first lesson is that occurrence data and laboratory toxicology studies alone are not enough to bring about action when the effects being studied do not involve humans. When it comes to wildlife, adverse effects must be documented in the field. In the case of DDT, the direct link between tissue levels and reproductive failure of bald eagles and brown pelicans turned the tide on a product that was considered essential to farmers. In contrast, the widespread occurrence of polybrominated diphenyl ethers (PBDEs) and perfluoroalkyl substances in polar bears garnered plenty of media attention, but without field evidence of adverse effects, regulatory actions were hard to justify. For microplastics, the public might not be as motivated if the adverse effects are limited to decreased feeding by microscopic creatures living near the bottom of the food web. Furthermore, waterways with the highest concentrations of microplastics are also subject to other pollutant stresses that could make it difficult to attribute compromised wildlife health to microplastics. To prove adverse effects of microplastics under realistic conditions, dosing of entire lakes, using methods similar than those used to document the effects of ethinyl estradiol on fish populations, might be needed. Because the addition of microplastics to pristine ocean waters would be impractical, such large-scale manipulations would require researchers to devise clever ways of removing microplastics from already contaminated marine waters.

Turning our attention to people, the second lesson is that contaminants are more likely to emerge if there is a reasonable possibility that their use is endangering human health. For example, when PBDEs were reported in human serum and breast milk, regulators took action before health effects were documented. As long as we consider human health as our top environmental priority, occurrence data and toxicology studies suggesting that contaminant concentrations are approaching a level of concern can bring about action. In the case of microplastics, human health risks have been posited, but the complexities associated with microplastic uptake as well as the simultaneous exposure of people to a myriad of other particles are going to challenge researchers seeking to assess the health risks of microplastics. Furthermore, one of the human health concerns that is frequently discussed—namely that microplastics expose people to lipophilic chemicals—is likely to be seen as an issue that is best handled by controlling the lipophilic chemicals rather than the media that increase their uptake.

The third lesson is that the likelihood that society will control an emerging contaminant is inversely proportional to the cost of solving the problem as well as the degree to which blame can be affixed on a small number of companies. The first part of this lesson is intuitive: expensive regulatory action requires a high threshold of evidence. Replacing microbeads in facial scrubs is a lot easier than rethinking the thousands of uses of plastics in the economy. The second part is less obvious but just as relevant: product bans and requirements to clean up contamination are more likely when only a few companies manufacture and use the chemical. For example, Monsanto, Westinghouse, and General Electric spent over $10 billion cleaning up PCB-contaminated sites. In contrast, the hundreds of companies that mine and use copper in construction materials, electronics and brake pads have not funded upgrades to sewage treatment plants or the installation of stormwater treatment systems in places where waterways are contaminated with the metal.

If it turns out that a specific use of plastic accounts for a disproportionate share of the microplastics detected in the environment, action is more likely. As long as researchers focus on a suite of sources that would be nearly impossible to eliminate, control options implemented in the near term are likely to be restricted to relatively inexpensive practices (e.g., litter control campaigns, marketing of biodegradable plastics to eco-friendly consumers) that might ultimately have little impact. Although we are all responsible for microplastics in the environment, getting the entire world to rethink the way it uses synthetic polymers would be a long, arduous process requiring compelling evidence of severe environmental risks.

The science and engineering of microplastics will be different from that of the chemical contaminants that preceded them. Nevertheless, we should learn our emerging contaminant history lessons. As we embark on our second decade of microplastics

Published: July 10, 2017

DOI: 10.1021/acs.est.7b03340
Environ. Sci. Technol. 2017, 51, 7747−7748

research, we need to set our sights on how best to provide society with the information needed to decide what to do about our newest emerging contaminant.

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Notes

Views expressed in this editorial are those of the author and not necessarily the views of the ACS.

The author declares no competing financial interest.

DOI: 10.1021/acs.est.7b03340
Environ. Sci. Technol. 2017, 51, 7747−7748

frontiers in Environmental Science

ORIGINAL RESEARCH
published: 19 July 2017
doi: 10.3389/fenvs.2017.00045

Distribution and Modeled Transport of Plastic Pollution in the Great Lakes, the World's Largest Freshwater Resource

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Most plastic pollution originates on land. As such, freshwater bodies serve as conduits for the transport of plastic litter to the ocean. Understanding the concentrations and fluxes of plastic litter in freshwater ecosystems is critical to our understanding of the global plastic litter budget and underpins the success of future management strategies. We conducted a replicated field survey of surface plastic concentrations in four lakes in the North American Great Lakes system, the largest contiguous freshwater system on the planet. We then modeled plastic transport to resolve spatial and temporal variability of plastic distribution in one of the Great Lakes, Lake Erie. Triplicate surface samples were collected at 38 stations in mid-summer of 2014. Plastic particles >106 μm in size were quantified. Concentrations were highest near populated urban areas and their water infrastructure. In the highest concentration trawl, nearly 2 million fragments km^{-2} were found in the Detroit River—dwarfing previous reports of Great Lakes plastic abundances by over 4-fold. Yet, the accuracy of single trawl counts was challenged: within-station plastic abundances varied 0- to 3-fold between replicate trawls. In the smallest size class (106–1,000 μm), false positive rates of 12–24% were determined analytically for plastic vs. non-plastic, while false negative rates averaged ∼18%. Though predicted to form in summer by the existing Lake Erie circulation model, our transport model did not predict a permanent surface "Lake Erie Garbage Patch" in its central basin—a trend supported by field survey data. Rather, general eastward transport with recirculation in the major basins was predicted. Further, modeled plastic residence times were drastically influenced by plastic buoyancy. Neutrally buoyant plastics—those with the same density as the ambient water—were flushed several times slower than plastics floating at the water's surface and exceeded the hydraulic residence time of the lake. It is likely that the ecosystem impacts of plastic litter persist in the Great Lakes longer than assumed based on lake flushing rates. This study furthers our understanding of plastic pollution in the Great Lakes, a model freshwater system to study the movement of plastic from anthropogenic sources to environmental sinks.

Keywords: plastic debris, Great Lakes, freshwater pollution, transport model

OPEN ACCESS

Edited by:
Julia Reisser, The Ocean Cleanup, Netherlands

Reviewed by:
Mario Barletta, Federal University of Pernambuco, Brazil
Francisco José A. Cysneiros, Federal University of Pernambuco, Brazil
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Specialty section:
This article was submitted to Marine Pollution, a section of the journal Frontiers in Environmental Science

Received: *05 December 2016*
Accepted: *03 July 2017*
Published: *19 July 2017*

Citation:
Cable RN, Beletsky D, Beletsky R, Wigginton K, Locke BW and Duhaime MB (2017) Distribution and Modeled Transport of Plastic Pollution in the Great Lakes, the World's Largest Freshwater Resource. Front. Environ. Sci. 5:45. doi: 10.3389/fenvs.2017.00045

INTRODUCTION

In recent years, anthropogenic litter in the form of plastic debris has been documented in widespread and diverse marine (Law et al., 2010, 2014; Cózar et al., 2014; Fischer et al., 2015; van Sebille et al., 2015; Law, 2016), freshwater (Eriksen et al., 2013; Free et al., 2014; Mani et al., 2015; Baldwin et al., 2016; Mason et al., 2016), and even aeolian (Dris et al., 2015) biomes. It is estimated that 4.8–12.7 million tons of plastic enters the ocean in a single year (Jambeck et al., 2015), with a quarter of a million tons currently floating in the world's oceans (Eriksen et al., 2014). It is estimated that 70–80% of marine litter (most of which is plastic) originates from inland sources via rivers (GESAMP, 2010). In the absence of mechanisms to incentivize improved waste management and behavior change, this number will continue to rise, reflecting the exponentially increasing global production of plastic goods (PlasticsEurope: Association of Plastics Manufacturers, 2015). Studies have shown that aquatic organisms ingest plastic pollutants (Boerger et al., 2010; Foekema et al., 2013). Consumption results in energetic and fitness costs (Besseling et al., 2012; Wright et al., 2013) and other morbid impacts (Rochman et al., 2013). There is a high likelihood that humans are consuming plastic derived from fish and shellfish (Van Cauwenberghe and Janssen, 2014; Rochman et al., 2015b), with as of yet unknown health consequences. In the wake of these discoveries, the United Nations has declared plastic pollution among the most critical emerging environmental issues of our time (UNEP, 2016). The scientific consensus is that plastic pollution must be reduced to avoid the risk of irreversible ecosystem harm (Rochman et al., 2016). Yet, an incomplete understanding of the global plastic litter budget hinders the strategic development of mitigation and prevention policy. To effectively target major sources and pathways, the question remains: what drives the concentration and flux of plastic debris across environmental reservoirs?

Plastic pollution first was reported in the ocean over 40 years ago (Carpenter and Smith, 1972; Colton et al., 1974; Wong et al., 1974) and has continued to be a focus of extensive research efforts (Moore et al., 2001; Thompson et al., 2004; Law et al., 2010, 2014; Cózar et al., 2014). Recently, there has been a call to bring similar focus to freshwater (Wagner et al., 2014; Dris et al., 2015; Eerkes-Medrano et al., 2015). Concentrations of microplastics—plastics <5 mm in the largest dimension—in lakes and rivers have been reported as high, or higher, than in central oceans gyres (Eriksen et al., 2013; Castañeda et al., 2014; Free et al., 2014; Lechner et al., 2014; Yonkos et al., 2014; Corcoran et al., 2015; Mani et al., 2015; Baldwin et al., 2016; Mason et al., 2016). Freshwater ecosystems play a critical role in the global water cycle and human health. They connect the inland watersheds that provide drinking water for most of the global population. It is essential to understand the nature and impacts of emergent contaminants, such as, plastic litter, their associated persistent organic toxins (Koelmans et al., 2016; O'Connor et al., 2016), and properties of plastic-toxin interactions (Hankett et al., 2016) to effectively preserve this resource.

The North American Great Lakes system contains one-fifth of the world's freshwater and is arguably one of the continent's most valuable natural resources. Field surveys have confirmed the presence of microplastics in Great Lakes surface water (Eriksen et al., 2013; Mason et al., 2016), sediment (Corcoran et al., 2015; Ballent et al., 2016), and beaches (Zbyszewski and Corcoran, 2011; Hoellein et al., 2014; Zbyszewski et al., 2014; Driedger et al., 2015), as well as the rivers (Baldwin et al., 2016) and wastewater treatment plant (WWTP) effluents (Michielssen et al., 2016) that directly feed into the Great Lakes. Yet, these data are sparse. There is currently insufficient knowledge of spatial and temporal resolution of plastic debris in the Great Lakes to efficiently focus strategic mitigation and management.

The study of plastic in the environment is a rapidly growing field of research. Studies from many sectors have employed diverse analytical methods for the isolation, identification, and quantification of plastic particles in environmental samples. While studies continue to resolve the limits of the myriad new methods used, it remains difficult to obtain, with meaningful throughput and accuracy, a seemingly simple data type: plastic counts. For instance, in the absence of replicate sampling, we do not know how representative single samples are of the environments from which they are collected. Further, most studies rely on visual inspection of samples to identify and count plastic particles. Yet, visual identification can incur error rates from 20 (Eriksen et al., 2013) to 70% (Hidalgo Ruz et al., 2012), with nearly 99% misidentification for sediment samples (Löder and Gerdts, 2015). These challenges hinder future research efforts, as well as our ability to leverage existing data describing environmental plastic.

In this study, we addressed five objectives and sought to answer: (i) What is the spatial distribution of plastic litter across three of the Great Lakes (Lakes Superior, Huron, and Erie) and one connecting lake (Lake St. Clair) down to the smallest particle size yet explored (106 μm)? We hypothesized that plastic concentrations would correlate with proximity to urban areas and that the concentrations observed would dwarf those reported using a larger size cut-off (333 μm; Eriksen et al., 2013). (ii) How is the distribution and the residence time of plastic litter influenced by physical properties of the plastic particles (e.g., buoyancy)? We hypothesized that neutrally buoyant particles, which move three dimensionally through the water column, would have a longer residence time than floating particles that experience surface drift only. (iii) Do permanent features of high plastic pollution exist (e.g., a "Lake Erie Garbage Patch") where mitigation could be focused? Based on existing hydrodynamic models of Lake Erie that predict summer convergence (Beletsky et al., 2013), we hypothesized that permanent features of high plastic pollution would exist in surface drift models and field survey data in anticyclonic anomalies. To inform method development and data interpretation in this study and across the field, we sought to answer (iv) how variable are plastic concentrations among triplicate trawls sampled consecutively at the same location? We hypothesized that within-station variability in count data would not be even across sites, but rather could depend on weather and sampling conditions. Finally, we asked (v) what is the false-positive rate for discerning plastic from non-plastic particles based on visual inspection? As dozens of previous studies have relied on visual inspection alone, we hypothesized that false-positive rates would be <50%, implying

that this method was not prohibitively error-prone. Collectively, these efforts lead to a better understanding of the drivers of freshwater plastic pollution in the Great Lakes and around the globe.

MATERIALS AND METHODS

Lake Sampling

To assess the spatial distribution of plastics across three Great Lakes and Lake St. Clair (objective i) surface water samples were collected at 38 stations (**Figure 1**) throughout the summer (May–August) of 2014 using a rectangular manta trawl 16 cm high by 61 cm wide towing a 100 μm Nitex mesh net 3 m long (Wildco) with a 100 μm Nitex mesh cod-end 28 cm long by 15.5 cm in diameter and a flowmeter (General Oceanics Model 2030R Mechanical Standard Rotor). The net was towed outside the wake of the boat at ∼2 knots for 20 min. For precision comparison at each station (objective iv), consecutive triplicate trawls were performed over the same transect. The difference in flow meter readings was multiplied by the manufacturer rotor constant and the width of the net mouth to calculate the area of water sampled. In order to standardize and compare plastics concentrations with previous studies (Eriksen et al., 2013; Mason et al., 2016), counts were divided by respective trawl area to achieve concentrations of plastics km^{-2}.

Stations were categorized as basin (>12 km from coast, $n = 7$), non-urban (<12 km from coast with <5,000 inhabitants km^{-2}, $n = 15$), urban (<12 km from coast with >5,000 inhabitants km^{-2}, $n = 6$), river plume ($n = 5$), and WWTP (sampled from environment near where final effluent is released, $n = 5$; **Figure 1**). Environmental data describing conditions at the start of each trawl, including wind speed, cloud cover, water temperature, air temperature, wave height, eastward surface water velocity, northward surface water velocity, wave direction, and wave period, were collected from the Great Lakes Observing System Point Query Tool of the Great Lakes Coastal Forecasting System[1]. Hourly data (or 3-h data, in the case of water temperature) for before and after the start time of each trawl were pulled, and the average was weighted by the number of minutes between data points. Descriptors of all trawls are available (Supplementary Data Sheet 1) where data interpolation was possible (e.g., no data existed for stations in Lake St. Clair or rivers).

Samples were recovered by rinsing the contents of the cod-end into a series of brass-framed sieves (Humboldt Mfg. Co.; Elgin, IL, USA) with stainless steel mesh sizes 4.75 mm, 212 μm, and 45 μm (**Figure 2A**; Humboldt Mfg. Co.). Each fraction was rinsed into a plastic bottle (HDPE bottle, PP screw top, Fisher Scientific 03-313-6C, 03-313-6B) with 70% ethanol for

[1]http://data.glos.us/glcfs/

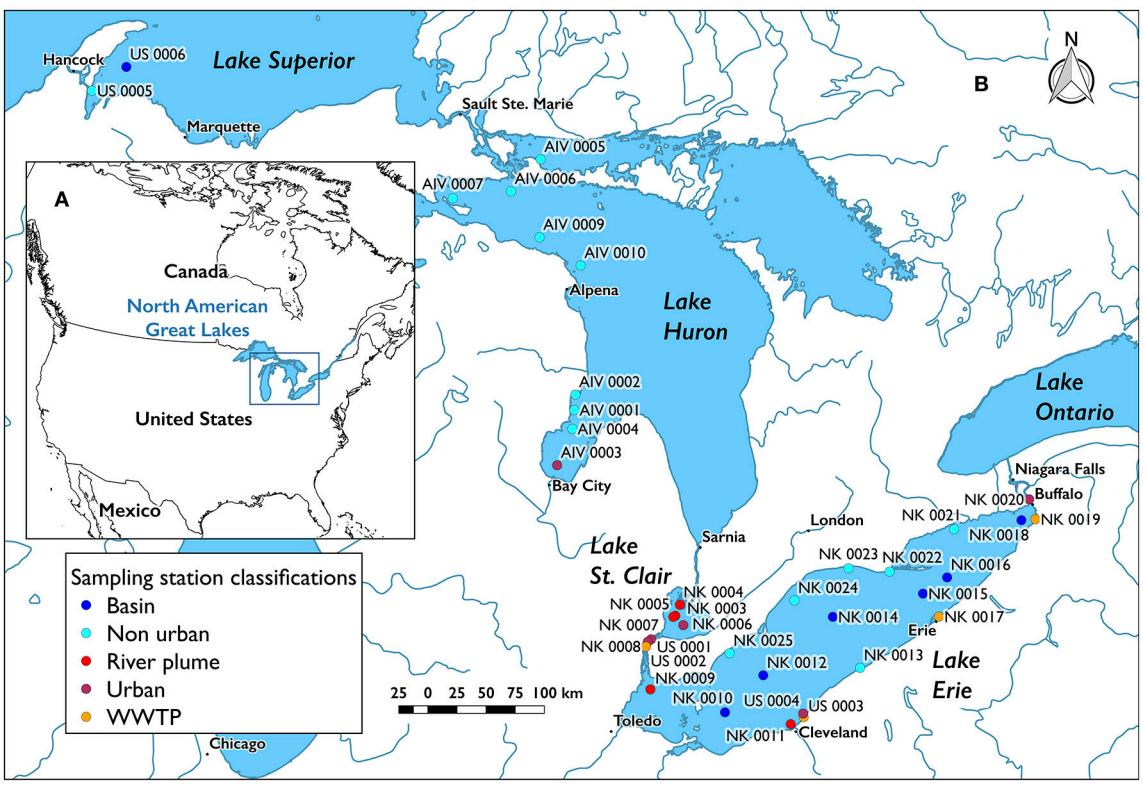

FIGURE 1 | Maps of all stations sampled. Each station contained replicate trawls. **(A)** Overview and location of the North American Great Lakes on the North American continent. **(B)** Station locations and classifications in Lake Superior, Lake Huron, Lake St. Clair, and Lake Erie.

FIGURE 2 | Photographs of samples at various stages of processing, as well as examples of different shape classes. Arrows indicate plastic amidst co-sampled nonplastic organic matter; blue: fragment, dark red: line, yellow: nurdles, cyan: sphere/bead, brown: fiber. **(A)** Bulk sample directly upon retrieval from Manta on a stack of a series of sieves. This sample contained an abundance of algal biomass. **(B)** Bulk sample drying on a 53 μm mesh net. **(C)** Sample after enzymatic processing, which included an incubation in hydrogen peroxide that bleached much of the non-plastic organic matter. This bleaching aided in differentiating plastic (tended to retain color) from non-plastic (prone to bleaching) particles. **(D)** Examples of plastic of sphere class; zoomed in subset of sample in **(B)**. **(E)** Smallest size fraction (106–1,000 μm) after WPO. Note colored plastic fibers (brown arrows) enmeshed in mass of natural fibers bleached white from hydrogen peroxide treatment. **(F–H)** Examples of plastic of fragment, film and line shape classes, respectively; ruler markings are in cm units. **(J,I)** Examples of plastic of paint chip and fiber shape classes, respectively; grid squares are in 5 mm units.

preservation. Sampled items that were too large to fit in a bottle were stored in Ziploc XL bags for later examination. All liquids used directly on the samples were filtered through 100 or 20 μm Nitex mesh in the field.

Sample Processing and Counting

Field-collected samples were spread over 53 μm Nitex mesh (**Figure 2B**), weighed for wet mass, dried at 60°C, and subsequently weighed for dry mass. Large pieces of organic material (e.g., sticks, leaves, etc.) were removed manually. The sample was mixed at a 1:1 ratio with 10% sodium dodecyl sulfate (Acros Organics 226140025) and incubated at 50°C, rotating at 80 rpm for least 24 h. Samples were then size-fractionated through a series of brass and stainless steel sieves (Humboldt Mfg. Co.; Elgin, IL, USA) with mesh sizes 4.75 mm, 1,000 μm, and 106 μm.

The 106–1,000 μm fraction of each sample was digested to remove non-plastic labile organic matter. The first digestion method used consecutive incubations with proteinase, cellulase, and chitinase, followed by incubation in 30% H_2O_2 for 24 h [*sensu* (Lorenz, 2014); **Figure 2C**: sample images]. Following the release of a NOAA Marine Debris Technical Memorandum providing guidelines on the analysis of microplastics in the marine environment (Masura et al., 2015), all previously processed samples were re-processed, and all subsequent samples were processed using only the wet peroxide oxidation (WPO) protocol recommended therein (2015). After oxidation, the remaining material was filtered over 104 μm stainless steel filters (TWP Inc., 150 Mesh T304 Stainless 0.0026; Berkeley, CA), and transferred to a glass petri-dish with 70% ethanol and dried.

Plastic pieces were manually pulled from the <4.75 mm fraction. The raw 1.00–4.75 mm and digested 106–1,000 μm fractions were visually sorted with the aid of a stereo dissecting microscope (10–80x; Zeiss SteREO Discovery.V8; Oberkochen, Germany). Each plastic piece in the two larger size classes was categorized by shape (**Figures 2E–I**): fragment (secondary plastic broken down from larger debris), film (e.g., thin plastic from bags and wrappers), fiber (e.g., individual filaments of textile threads, very thin and frequently curled), line (e.g., fishing line, straighter, and thicker than fiber), nurdle (preproduction plastic pellet), sphere, foam, or paint (consistent with multiple studies that consider paint a plastic or confirm it is composed of, e.g., alkyds and (poly)acrylate/styrene; Lima et al., 2014; Kang et al., 2015; Neves et al., 2015; Song et al., 2015; Imhof et al., 2016; Nizzetto et al., 2016). Such detailed categorization was not possible for the smallest size class (106–1,000 μm), so the smallest particles were classified as either fragment or fiber.

Substantial effort was invested in gaining experience and establishing confidence in visually and tactilely distinguishing plastic from non-plastic particles, especially in the smallest (106–1,000 μm) size class. A collection of characteristics was established to distinguish plastic from non-plastic and to categorize plastics into morphological types. Physical features (color, hardness, fragility, shape) were considered. Features that frequently indicated plastic fragments included: malleability (not brittle), defined jagged shape, shiny surface, and presence of artificial dyes. Dye-free plastic particles were identified by their opaque and white nature. Features that often indicated an inorganic particle included: brittleness or unresponsiveness to force applied by tweezers, audible scratching noise when scraped, transparency, and well-defined crystalline structures and right-angle fractures.

Precaution was taken to minimize risk of sample contamination from handling and the laboratory environment. All liquid that came in contact with the samples (water for sieving, ethanol for storing) was filtered to remove particles >10 μm, glassware for storage was blasted with high-pressure air before use. Thin Teflon sheets (0.005 "Natural Virgin PTFE Roll Stock 12" Wide, Ridout Plastics Co. Inc.) were inserted between storage glassware and their plastic screw tops, as Teflon is rare among environmental plastics and its diagnostic fluoride ion could be detected analytically downstream if contamination did occur. Samples were processed in a laminar-flow or fume hood and remained covered otherwise. Cotton laboratory coats were worn by all individuals. Blank samples consisting of one 1,500 ml and two 500 ml aliquots of 10 μm-filtered MilliQ were processed and counted alongside field samples to account for environmental plastics incorporated during the sampling process that would lead to false positive plastic counts.

All data treatment and statistics were performed using the R statistical environment (version R-3.3.1; Team, 2014). All R code generated to create figures and perform calculations is freely available[2][3] Maps of trawl locations and counts were generated with Quantum GIS (v. 2.18; QGIS Development Team, 2016).

Scanning Electron Microscopy Energy-Dispersive X-Ray Spectroscopy (SEM-EDS)

To assess human error and determine our false positive vs. false negative rates in the assignment of the smallest particles as plastic (objective v), a subset of particles from the smallest size class were randomly chosen from each of the suspected plastic ($n = 10$) and suspected non-plastic particle ($n = 10$) pools across 10 trawls. These particles were characterized analytically (described below). In addition, we prepared a library of 35 known standards to inform our ability to differentiate plastic, mineral, and non-synthetic organic matter and identify potential contamination of our samples from plastic in the processing environment. Standard items included virgin polymers, plasticware, and instruments used for sample collection, processing, and storage, paint from a sampling vessel (R/V *Nancy K*), fibers from lab coats, hair from sample processors, phytoplankton carcasses, and mineral particles.

SEM-EDS was performed to acquire an atomic signature for the 260 particles and standards assessed. Particles were mounted on an SEM peg (0.5 in. diameter; Electron Microscopy Sciences, Cat. 75160; PA, USA) with a piece of double-sided carbon tape (Electron Microscopy Sciences, Cat. 77816; PA, USA). A thin layer (∼40 nm) of gold was applied to the sample using a gold sputter coating machine (120 s, Denton Vacuum Inc., Desk II, Cherry Hill, N.J.). Each particle was imaged using a JEOL JSM-7800F SEM at an accelerating voltage of 15 keV and an acquisition time of 20 s. A rectangular well-focused central area on each particle was excited via EDS. The resulting spectra were analyzed with Oxford AZtec 3.1 EDS software. The auto-ID function using default parameters verified the presence of elements on the surface of each particle. Following data acquisition, particles were assigned to each of three classes based on peak elements and surface texture: inorganic/mineral (IO), non-plastic (NP) organic matter, and plastic (P). Some gradation was allowed between discrete classes resulting in 5 different categories: P, P-NP, NP, NP-IO, IO.

Lake Erie Plastic Transport Model

It is not feasible to perform the high-resolution spatial and temporal sampling required to understand the lake-wide distribution and movements of plastic pieces. Thus, a Lagrangian particle transport model previously used in Lake Erie (Michalak

[2]https://github.com/DuhaimeLab/Frontiers_2017_GreatLakesPlasticDistrib
[3]http://www-personal.umich.edu/~duhaimem/Rpubs_code/GreatLakes_Plastic_Pollution_Study_Cable_etal_2017.html

et al., 2013; Fraker et al., 2015; Beletsky et al., 2017) was applied to simulate transport of microplastics over a variety of timescales and plastic properties (e.g., its buoyancy; objectives ii and iii). In this model, particle trajectories were calculated with the hydrodynamic model velocity recorded at regular time intervals (e.g., hourly). For each particle, the gridded velocities were interpolated to its location and the particle was moved to a new location based on the interpolated velocity and the time step of the particle transport model (Lynch et al., 2014). The three-dimensional particle trajectory code is based on the second order accurate horizontal trajectory code, as described in Bennett and Clites (1987), with the addition of vertical position tracking. Plastic "particles" in the model are neutrally buoyant (i.e., have the same density as the ambient water), passive (i.e., they follow local three-dimensional currents), and biochemically inert. If collision with model boundaries occurs, particles remain in the nearshore zone. The model includes horizontal and vertical diffusion, as introduced by Smagorinsky (i.e., with a non-dimensional coefficient of 0.005 in the horizontal diffusion parameterization; Smagorinsky, 1963) and random-walk approaches, respectively. Vertical diffusion was set at 5×10^{-4} m^2 s^{-1}. Because the size of most particles in this study is <1 mm, they are considered fully submerged and therefore windage is zero. In experiments that examine the effect of plastic buoyancy on residence time and transport, floating particles were driven by surface currents only, which were obtained from the top layer of the 3D hydrodynamic model.

Advection fields used by the particle model were produced by the three-dimensional finite-difference hydrodynamic model based on the Princeton Ocean Model (Blumberg and Mellor, 1987), driven by the wind, heat flux, and tributary flow from 22 major rivers and two outflows (listed in Schwab et al., 2009). The hydrodynamic model used a uniform 2 km horizontal grid with 21 vertical levels. Six years of hourly current data (2004–2005, 2007, and 2009–2011) obtained from previous applications (Beletsky et al., 2013) were used to model microplastic transport in summer months (including the month of June, the month of Lake Erie field sampling). In addition, year-long simulations were conducted when particles were continuously released throughout each year. To calculate residence times, the sequence of years was looped because longer time periods were required to flush the vast majority of particles from the lake.

In each model simulation, virtual particles were released daily to Lake Erie surface water at 29 tributaries (Supplementary Table 2) and two WWTPs in the Cleveland area. Particles left the lake through Niagara River and Welland Canal (easternmost edge of Lake Erie). For residence time calculations, particles were released during the first year (2004) and then tracked until the percentage of particles remaining in the lake dropped to 1% (eight years for neutrally buoyant particles).

RESULTS AND DISCUSSION

This dataset represents the largest single-season effort of plastic quantification in the Great Lakes to date. Plastic was counted in 108 surface trawl samples, which spanned 38 stations across Lakes Superior, Huron, St. Clair, and Erie. Plastic was found at every station sampled (**Figure 3**). The trawl with the highest total concentration of plastic contained 4-fold higher plastic than yet reported in the surface of the Great Lakes (Eriksen et al., 2013; Mason et al., 2016). The vast majority of plastic counted was <1 mm in size (**Figure 4A**).

Concentrations and Distributions of Great Lakes Plastic

Plastic Concentrations Were Highest at Urban Centers

Total plastic abundances per surface trawl spanned an order of magnitude. They ranged from 1,910,562 particles km^{-2} in the Detroit River plume (NK0008-3) to 126,933 particles km^{-2} in the Straits of Mackinac in Lake Huron (NK 0007-1; **Figure 3D**; Supplementary Data Sheet 1). Notably, these total concentrations and all that follow do not include counts of fibers, as during sample processing it became evident that fibers could not be quantified with equally high confidence across size fractions, an issue which is discussed at length below. Fiber concentrations were analyzed separately to explore patterns in the data.

The highest concentrations of plastic were found in samples collected within 12 km of the coast of populated urban cities, in river plumes, or directly at the effluent of WWTPs (**Figures 3**, **4B**). All of the most concentrated samples but one were collected in Lake Erie or the urban river and estuary-like lake directly feeding it (Detroit River and Lake St Clair; **Figure 2**). Our empirical data support recent model predictions that the loads of Lake Erie plastic inputs are 4- and 80-fold higher than Lakes Huron and Superior, respectively (Hoffman and Hittinger, 2017). Notably, the plastic input loads for this model were scaled to census-derived population density of the coastlines (Hoffman and Hittinger, 2017)—an underlying presumed correlation our field data support. The lowest counts were collected at non-urban coastal stations and offshore basin stations, with the exception of the deepest point of the Eastern Basin of Lake Erie (**Figures 3**, **4B**). These findings support previous reports of a correlation between plastic concentrations and proximity to urban centers in the Great Lakes (Baldwin et al., 2016), as well as other enclosed and semi-enclosed aquatic environments across the world, such as, tributaries to the Chesapeake Bay, USA (Yonkos et al., 2014), the Bay of Brest in France (Frère et al., 2017), the Xiangzi Bay upstream of the three Gorges Dam (Zhang et al., 2017), inland lakes around Wuhan, China (Wang et al., 2017), and estuaries in and around Durban, South Africa (Naidoo et al., 2015). Attributes that are likely to contribute to elevated plastic concentrations in urban vs. non-urban locales include higher population densities (Jambeck et al., 2015), increased particulate aeolian inputs (including plastic; Dris et al., 2015), and increased areas of impervious substrate. The percent of a watershed comprised of impervious substrate is positively correlated with higher plastic concentrations in the Great Lakes watershed (Baldwin et al., 2016), likely due to greater volume and higher velocity runoff during storm and snow melt events. The higher concentrations in river plumes

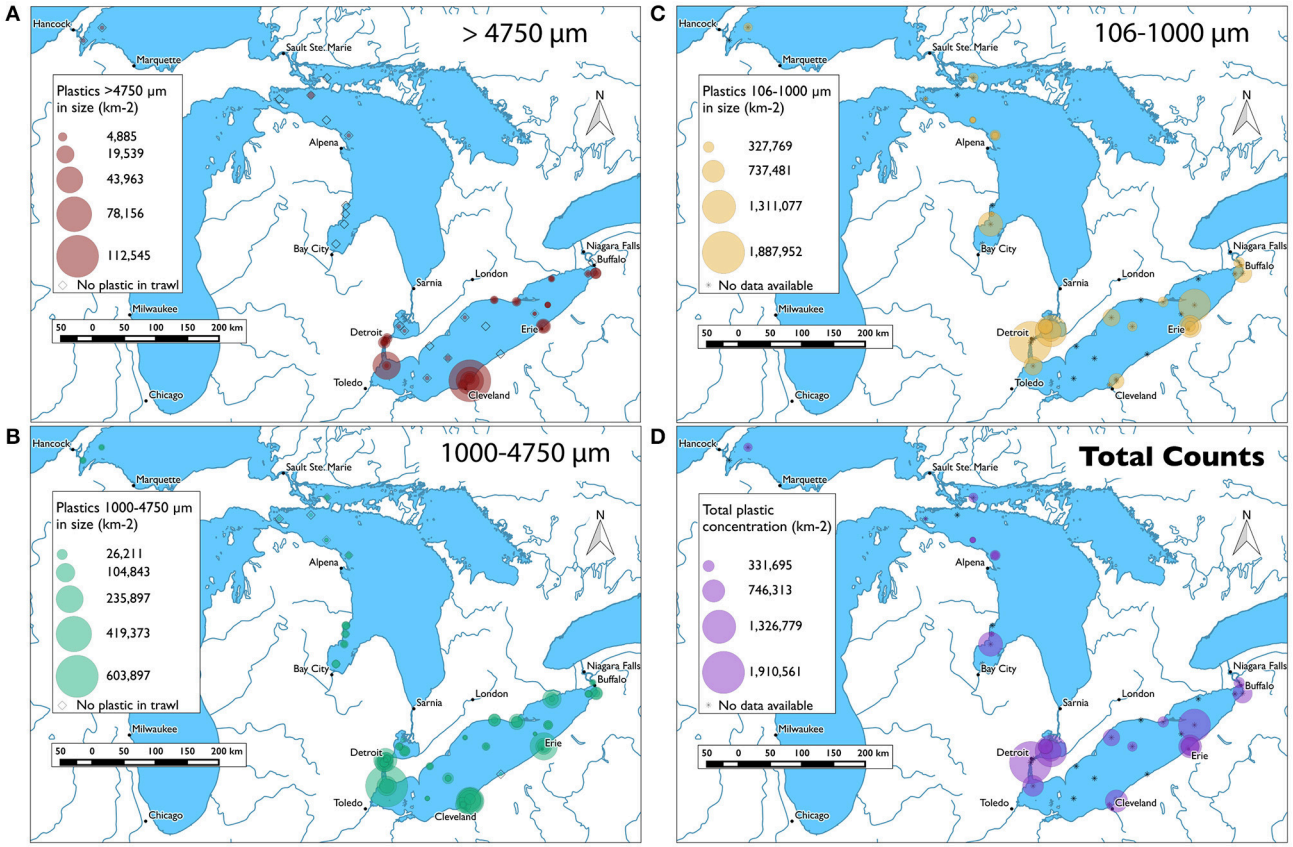

FIGURE 3 | Maps of plastic concentrations across the lakes sampled, where magnitude of concentration is depicted by size of circle around trawl location. Note, fiber counts are not included in these figures, as discussed below. **(A)** Mapped counts of plastic litter >4,750 µm. **(B)** Mapped counts of plastic litter 1,000–4,750 µm. **(C)** Mapped counts of plastic litter 106–1,000 µm. **(D)** Total mapped counts for the stations where all three size classes were quantified.

and near WWTP effluents than in coastal areas (**Figure 4B**) suggest these inputs to be sources (McCormick et al., 2014) and that plastic debris enters this system from inland waterways and human activity. Increasing the degree of pervious substrate in watersheds, such as, the implementation of green infrastructure catchments, should be explored as an effective measure to capture plastic debris in runoff and reduce loads ultimately reaching waterways. As the number of storm events is expected to increase with a changing climate (IPCC, 2012), such innovations are timely to preventatively buffer our freshwater systems from being inundated with stormwater-delivered debris.

Single Trawls Are Imprecise: Within-Station Variability Can Vary 3-Fold

This is the first survey of freshwater plastic litter to address variability in counts by conducting replicate trawls at each of 38 stations. The distributions of all trawl concentrations, total concentrations, and station concentrations deviated significantly from normal distribution (Shapiro Wilks test, $p << 0.01$) with high skewness (1.9–6.62) and kurtosis (3.5–49.25; Supplementary Figure 1). As a result, non-parametric tests were used (e.g., Spearman's rank correlation) and metrics that do not represent strongly skewed data (e.g., standard deviation) were not used to describe and interpret the results. Rather, to assess factors that influence within-station variability, we calculated a metric we termed the mean-normalized range (MNR) by dividing the count range (max–min) of each station by the mean of the station.

The vast majority of trawl concentrations from the same station varied more than 100%, as depicted by a mean normalized range (MNR) >1 (**Figure 5**; Supplementary Data Sheet 3). In other words, the accuracy of a single trawl at one station is quite low and repeated trawls at the same location can vary in precision by up to 3-fold. We suspect that the magnitude of MNR at certain stations is due to undersampling. Precision increases as the plastic concentration sampled increases, as MNR is significantly negatively correlated with total trawl concentration (Spearman's $rho = -0.629$, $p = 0.000$; **Figure 5**). MNR is <1 for all counts in the smallest size class, which have the largest concentrations ($M = 0.09$) and most frequently >1 in the largest size class ($M = 1.94$; **Figure 5**, Supplementary Figure 2), which have relatively lower concentrations. While dependent on plastic concentration, the MNR was not significantly influenced by air velocity ($rho = -0.093$, $p = 0.245$), east-west surface current velocity ($rho = -0.072$, $p = 0.364$), wave period ($rho = -0.078$, $p = 0.330$), or wave height ($rho = -0.093$, $p = 0.242$)—all local conditions that could influence the distribution of plastics at the water surface

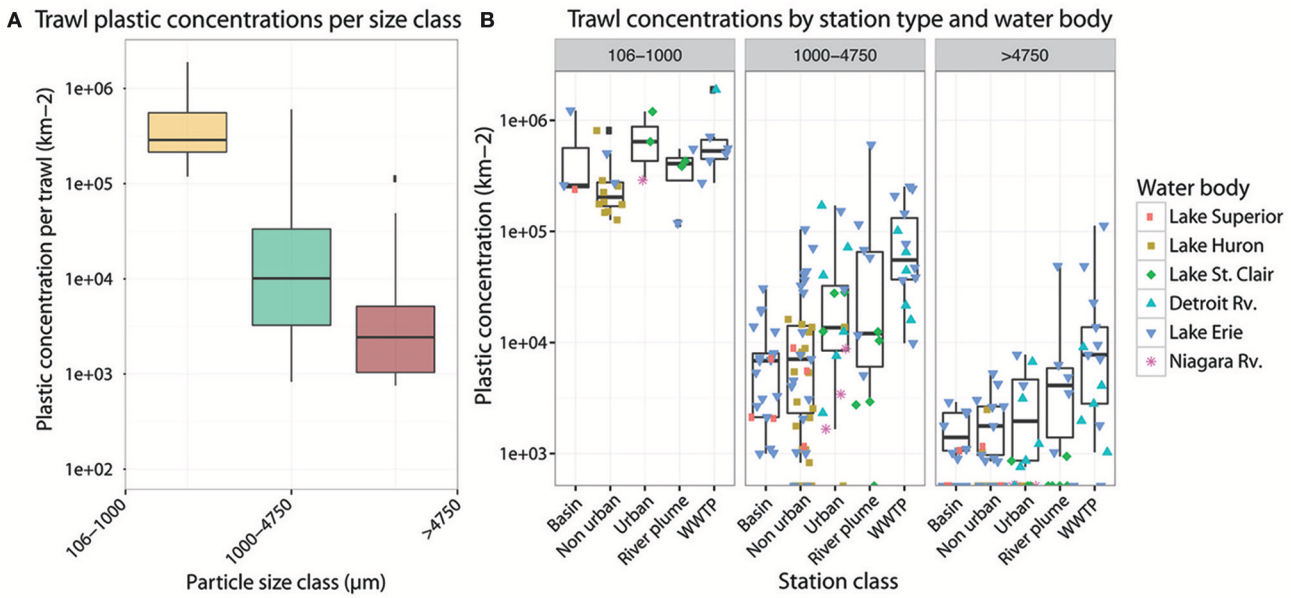

FIGURE 4 | (A) Boxplots depicting the means and spreads of all individual trawl counts by particle size fraction. **(B)** Boxplots depicting the means and spreads of individual trawl counts by size class, station type, and water body: Lake Superior (pink square), Lake Huron (brown square), Lake St. Clair (green diamond), the Detroit Rv. (blue upright triangle), Lake Erie (dark blue upside-down triangle), and the Niagara Rv. (purple star).

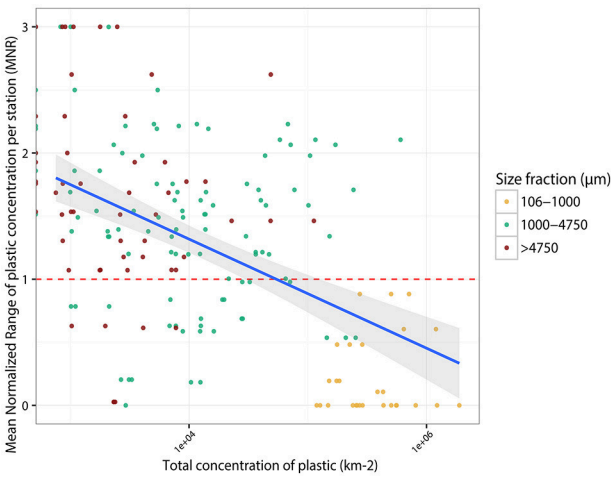

FIGURE 5 | Plot of trawl mean normalized ranges (MNR) across stations vs. the log of plastic concentration in each individual trawl. Regression line of the linear model used to predict MNR from plastic concentration is shown (R^2 = 0.442, slope = −0.357, intercept = 2.737, p < 2.2e-16). The gray band depicts the 95% confidence level interval for predictions from the linear model.

between trawls. However, longitudinal surface current velocity positively varied with MNR ($rho = 0.166$, $p = 0.037$); an increase in north-south current velocity was correlated with a decrease in precision between trawls. As currents in the lake are mostly wind-driven and winds on Lake Erie predominantly blow west to east, increases in north-south current velocity may indicate a local weather anomaly, such as, a squall or storm. These features are known to build up and die down quickly; it was not uncommon to experience a short burst in weather change over the course of the 1–2 h spent sampling at a single station. Such dynamic local conditions could increase the variability between trawl counts within a single station and decrease the accuracy of a trawl. To maximize reliability of surface plastic counts, we suggest samples not be taken around wind-related weather anomalies.

A similar survey of marine plastic debris assessed variability with replicate sample quantification in the North Pacific Gyre (Goldstein et al., 2013). This study found a mean within-station coefficient of variation (CoV; calculated as the station standard deviation divided by the station mean) of 51.4% for net-collected samples. CoV depends on the station standard deviation, which we deemed an inappropriate representation of data as heavily positively skewed as ours (Supplementary Figure 1). Yet, for purposes of comparison, we determined the CoV across the stations in this study and found they ranged from 1.5 to 173% (Supplementary Figure 3). The CoV of the smallest size class was less than that of the North Pacific study, whereas the CoV of larger size classes was greater (Supplementary Data Sheet 3). In the power analysis performed by Goldstein et al. (2013), statistical power increased when number of samples increased. In the case of our data, within-station variability appeared more influenced by the plastic count in each sample than the number of samples counted (as $n = 28$ for the smallest size class, and $n = 108$ for the two larger size classes). In order to reduce the within-station variability of the larger two size classes at stations with low overall plastic concentrations, greater counts are needed per trawl, thus sampling should occur over a larger area. We suggest a minimum MNR of <1 and ideally lower. As field survey data is time consuming and costly, recognition of this count-dependent variability and the importance of replication is critical

TABLE 1 | Mean and standard deviations of plastic type concentrations (km^{-2}) across all trawls and size classes quantified.

Size (μm)	Fragment	Film	Line	Nurdle	Sphere	Foam	Paint	Total Plastic	*n*
106–1,000	465,606 ± 403,378	NA	NA	NA	NA	NA	NA	465,606 ± 403,378	28
1,000–4,750	19,237 ± 42,995	1,607 ± 3,195	1,109 ± 2,040	3,742 ± 19,500	966 ± 3,343	4,443 ± 12,953	1,115 ± 2,475	32,219 ± 73,576	108
>4,750	2,009 ± 8,500	880 ± 2,883	168 ± 460	19 ± 138	0 ± 0	427 ± 1,865	0 ± 0	3,503 ± 12,766	108

for maximizing the value of such datasets, especially as future field survey studies are designed and implemented.

Plastic Less than 1 mm Dominated the Dataset

The mean concentration of plastic in the smallest size class (106–1,000 μm) was 15-fold higher than the middle size class (1,000–4,750 μm) and 130-fold higher than the largest size class (>4,750 μm; **Figure 4A**, **Table 1**). A similar pattern was maintained in all trawls, regardless of water body or types of stations sampled (**Figure 4B**). These findings are consistent with surveys of other lakes, such as, lakes near Wuhan, China where more than 80% of the plastics found were 2 mm and smaller (Wang et al., 2017). However, plastics 1–5 mm in size were most abundant in sections of the Xiangxi River, perhaps due to a shorter residence time and less weathering while in the river (Zhang et al., 2017). Previous surveys of Great Lakes plastic have found a 40- and 6-fold difference between the smallest and largest size classes (Eriksen et al., 2013; Mason et al., 2016). It is likely that the order of magnitude increase in the relative abundance of the smallest size class between previous Great Lakes surveys and the overall maximum abundance in our study can be attributed to our use of a 106 μm size mesh collection net, as opposed to the 333 μm mesh used previously in the Great Lakes and their tributaries (Eriksen et al., 2013; Baldwin et al., 2016; Mason et al., 2016) and in most aquatic plastic debris surveys to date (Hidalgo Ruz et al., 2012; Law, 2016). As a result, our data more comprehensively capture the "micro" plastic range in the Great Lakes, knowledge of which is critical to our assessments of environmental risk. Smaller plastic particles stay at the water surface longer than larger particles of the same composition and shape (Khatmullina and Isachenko, 2016; Kowalski et al., 2016) and are more readily consumed by smaller organisms in aquatic food webs, increasing the chances of biomagnified effects due to predation (Wagner et al., 2014). Further, the larger surface area to volume ratios of these small particles increases their potential as vectors of adsorbing contaminants (Barnes et al., 2009; Teuten et al., 2009). Future studies should continue to probe this small size class, as well as develop innovative high-throughput solutions to capture and quantify particles below 106 μm and into the nanoscale, where risk may be highest due to subcellular effects (Syberg et al., 2015).

Secondary Plastics (Fragments) Were the Most Common Plastic Type

Fragments were the most abundant plastic shape class across the dataset (**Figure 6**). This finding is consistent with other recent studies that used comparable analytical methods, including a survey of 59 stations in Lake Michigan (79% fragments, 14% fibers; Mason et al., 2016), and even a study in remote Lake Hovsgol, Mongolia (40% fragments, 20% fibers and lines; Free et al., 2014). Rivers and urban effluent (e.g., WWTP) are thought to be major contributors of plastic to freshwater water systems. Notably, studies of sources of plastic to the Great Lakes have documented fibers to dominate, not fragments. An analysis of 29 Great Lakes tributaries (Baldwin et al., 2016) found total debris comprised of 71% fibers and 17% fragments. Similarly, anthropogenic litter in the effluent of a high capacity wastewater treatment plant that discharges directly to the Great Lakes was found to be 61% fibers and 33% fragments (Michielssen et al., 2016).

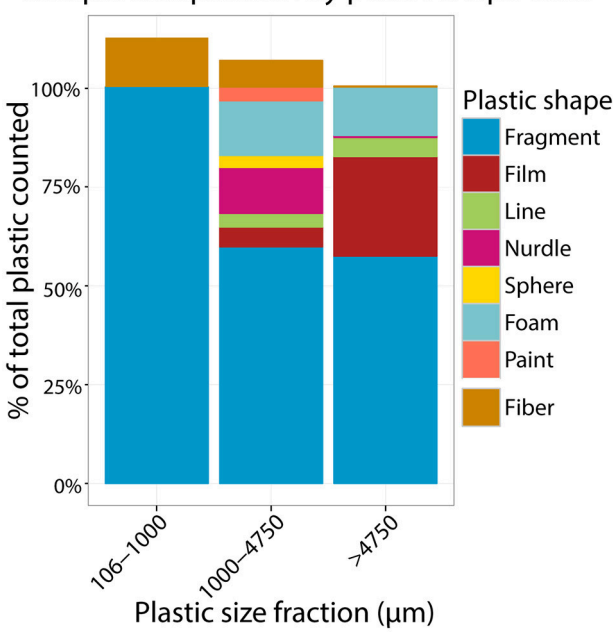

FIGURE 6 | Stacked barplot depicting the relative abundance of different shape classes amongst plastic from each size fraction. The bar to 100% for each size class represent the relative abundance of different shape classes when fibers were not included in the total counts and the portion above 100% represents the relative abundance of fibers in the total counts.

This difference may be due to the fact that typically fibers are comprised of polymers that are denser than water, e.g., nylon, polyester, acrylic. As such, in a stable water body (e.g., large lakes, ocean gyres) they are expected to sink, while in the flow of turbulent mixing systems (e.g., streams, rivers, WWTP effluent, tidal inlets) these fibers may remain mixed and in the seston (Baldwin et al., 2016). Fragments are primarily secondary

plastic debris and are likely to be composed of more positively buoyant polymers (e.g., polyethylene and polypropylene, as demonstrated in a study in Lake Michigan; Mason et al., 2016) that float at the lake surface. Alternatively, fibers may be drastically underestimated in surface aquatic environments owing to difficulties collecting fiber data, as discussed below.

When station type was considered, the relative abundances of fragments, foam, and (for the largest size class) film were high in urban and river plume samples—the latter of which were all coincidentally urban, as well (Supplementary Figure 2). Similarly, this trend was observed in river samples, where "litter-related plastic" (the collective class of fragments, foam, and film) was significantly more highly represented in Great Lakes tributaries of watersheds with urban attributes (Baldwin et al., 2016). This may be attributed to proximity to land-based plastic sources, such as, recreation on populated beaches and litter in urban areas and suggests that curbing mismanaged waste in urban centers could reduce the load of plastic in waterways.

Assessing Confidence in Plastic Count Data

Though recommendations (Ryan et al., 2009) and protocols (Masura et al., 2015) have been put forth for sample collection, processing, and quantification, standardized sampling methodology, and reporting are critically lacking (Hidalgo Ruz et al., 2012; Law, 2016). The reasons for these inconsistencies are multifaceted. This is a relatively young field of research with many newly recruited researchers from broad disciplines, e.g., environmental science, biology, chemistry, engineering, physics, oceanography, ecology, bringing diverse backgrounds to a common problem. Each study contributes new insights, but also highlights the Achilles' heel of their given approach. This process is necessary to arrive ultimately at a unified approach. In the present study, the greatest uncertainty arose in the treatment of fiber count data, as well as our ability to visually and chemically discern plastic particles from non-plastic in the smallest size class.

Confidence in fiber count data depends on size class and sorting effort

Fibers were identified in all size classes, yet the degree of certainty in the fiber count data depended on the size class, oxidative treatment of sample, and effort of the sample sorter. First, it is likely that fiber counts from field samples were underestimated because the sampled material was so heterogeneous causing fibers to be missed and unaccounted for. This was especially likely in the larger two size classes (1,000–4,750 μm and >4,750 μm), where WPO treatment was impractical at the volumes needed to be effective and thus could not be used to eliminate bulk non-plastic organic matter. In these fractions, the fibers, which are much less rigid than other plastic morphologies and more prone to "stick" to other objects when wet, were deeply enmeshed in the crevices of or entwined in natural fibers of non-plastic items (e.g., leaves, sticks, bark, feather, etc.) during sieving and sorting. As a result, fibers were difficult to separate from the non-plastic organic matter co-sampled from the lake surfaces, much of which was naturally fibrous (**Figures 2B–E**). This increased difficulty in acquiring fiber counts also required greater effort and vigilance by the person visually sorting, given the enmeshed fibers would be much thinner than other items the sorter was looking for. These issues were much less apparent in the smallest size class, where most non-plastic organic matter was removed chemically and fibers were more obvious with little surrounding or overlapping material. Thus, it is difficult to compare fiber abundance across size classes, as the "sorting effort" required varied widely. Second, owing to their small width and surface area, we could not use the same sensory data that we relied upon to discern plastic fragments from non-plastic particles under the microscope (e.g., squeezing, pinching, scratching, etc.). The small size of fibers also prohibited the controlled physical manipulation needed to perform chemical analysis via SEM-EDS—though we cannot predict whether this led to an over- or underestimate of fiber counts. Notably, these issues did not influence our ability to detect and report concentrations of plastic line. Lines were more discernible and behaved very differently when manipulated owing to their greater length, thickness, and consequent rigidity (**Figures 2A,H,J**).

Finally, fibers were the plastic type most likely to contaminate a sample during processing in this study. All but one of the 126 particles introduced to the blank controls were fibers (Supplementary Table 3). For instance, the 1,000–4,750 μm fraction of a single blank control contained 33 fibers, whereas the maximum raw number of fibers counted in the same size class was 33 and the average across all trawls was 24 (Supplementary Table 3). Further contributing to the underestimate of fibers in field samples relative to sample counts was that blank samples were pristine and easy to see, whereas fibers in field samples were often complex conglomerations of suspected natural and plastic fibers (**Figure 2E**). Though anecdotal evidence derived from observations during processing suggest that the environmental samples contained more fibers than the blanks, the possibility of contamination of samples by fibers could not be ruled out. Fiber contamination during sample processing has been reported previously (Foekema et al., 2013; Dekiff et al., 2014; McCormick et al., 2014; Woodall et al., 2015). A comparison of numbers of fibers introduced using different protocols suggested fiber contamination was introduced primarily as a result of sample sieving and moving from one holding vessel to another (unpublished data; BW Locke, RN Cable). We recommend taking precautions to reduce the number of times a sample is transferred, sieved, or filtered from the beginning of sample collection, in addition to reducing the amount of time a sample is exposed to open air outside of a fume or laminar hood.

It is paramount that the field overcomes the limitations and uncertainties related to the quantification of plastic fibers. Evidence is mounting that fibers are a dominant form of plastic pollution in many aquatic ecosystems—especially fluvial (McCormick et al., 2014; Zhao et al., 2014; Dris et al., 2015; Baldwin et al., 2016), but also in marine beaches and sediment (Browne et al., 2011; Claessens et al., 2011; Woodall et al., 2014; Fischer et al., 2015; Naidoo et al., 2015; Van Cauwenberghe et al., 2015). The ecological implications of these fibers remain to be shown, but plastic fibers are increasingly found in the stomachs and tissues of aquatic wildlife, many of which are consumed by larger animals, including humans (Neves et al., 2015; Rochman et al., 2015a; Vandermeersch et al., 2015; Li et al., 2016). Direct

human health impacts have been reported, as well: when inhaled, microplastic fibers are retained the lung tissues and can become associated with malignant tumors (Pauly et al., 1998). We must develop an accurate assessment of the sources, abundances, and impacts of synthetic fibers in our environment so that informed mitigation practices can be put into place, if deemed necessary.

Visual discrimination of plastics is confirmed by analytical methods

While most studies rely on visual inspection alone (reviewed in Hidalgo Ruz et al., 2012; Law, 2016), such human sensory-based observations can be error-prone. First, misidentification can occur due to the similarities in appearances of plastic and non-plastic particles (Filella, 2015). Second, the reliability of visual identification decreases with decreasing particle size. In the smallest size class, we used SEM-EDS analysis to test and reduce our rate of incorrectly differentiating plastic from non-plastic via visual and tactile inspection alone. EDS spectra and SEM images representative of plastic, inorganic, and non-plastic organic particles were highlighted (**Figure 7**). EDS spectra are summarized in Supplementary Data Sheet 2; EDS spectra and SEM microscopic data files are included in Supplementary Image 1.

To address erroneous counts caused by misidentification while sorting, we built a diverse library of standards (described in Supplementary Data Sheet 2). This library was used to train our classification efforts prior to analyzing sample spectra. Among the qualitative anecdotes resulting from the analysis of this library, we learned that microbeads from personal care products all contained the elements C (primary peak), N, Si, and, all but one, O (Supplementary Data Sheet 2). One personal care product (PCP) bead standard had a large Si peak relative to the other

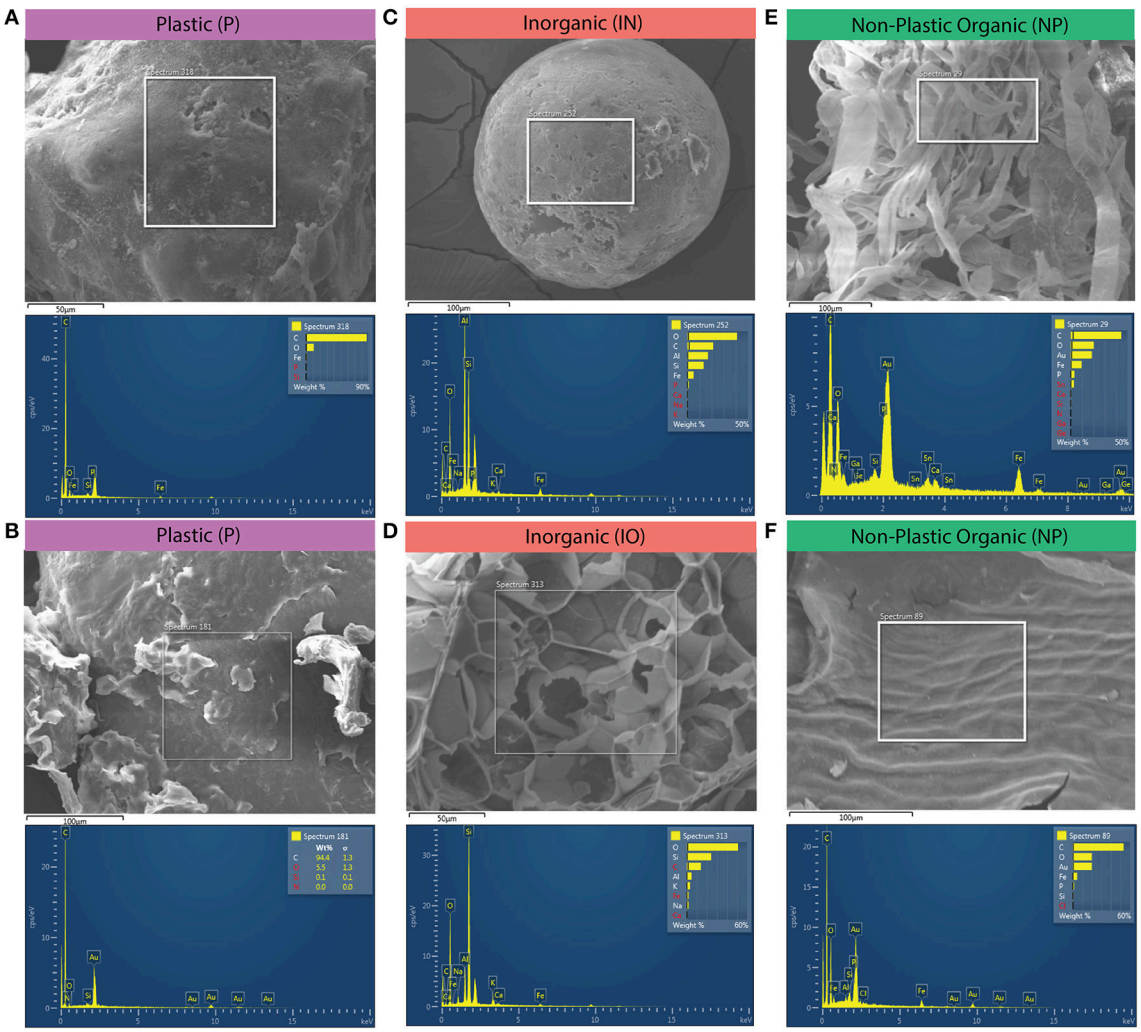

FIGURE 7 | Examples of SEM-EDS spectra and microscopic images representative of the three compositional classes: plastic (P), inorganic (IO), and non-plastic organic (O). **(A)** plastic fragment. **(B)** Flaking, jagged, "crumbly" bead from a personal care product that served as a positive control. **(C)** Coal fly ash from a Lake Erie sample. **(D)** Piece of silica foam from Lake Erie sample. **(E)** Stringy, fibrous organic matter particle from Lake Erie sample. **(F)** Non-plastic organic matter control depicting diagnostic shallow surface veining; likely a leaf or other vegetation.

elements. We attributed this composition to the particle being mica or previously having been in close association with mica. Indeed, sparkling "beads" from PCPs that crumbled upon touch were found often, which we presumed were mica particles, after finding it listed as an inactive ingredient in PCP. Further, all organic matter standards contained Fe (in the presence of O), as did the nylon mesh net that had been used to filter organic material, whereas no Fe was found in pristine virgin polymers. This pattern held until environmental samples were analyzed. As opposed to pristine standards, Fe was detected in nearly all particle types (plastic, non-plastic organic, and mineral) that had been exposed to the environment.

Physical features of the particle surface further informed our classification decisions between plastic and non-plastic organic. Plastic tended to have deep and clean fractures, and smooth surfaces with shallow flakes (e.g., **Figures 7A,B**); though this could be obscured as particles oxidized with age and appeared brittle. Particles with relatively simple elemental spectra consisting of a large primary C peak, frequently with a smaller O peak, were classified as plastic (P; **Figures 7A,B**). Inorganic (IO) particles were best characterized by the presence of a large primary peak of the element Si (**Figures 7C,D**; Supplementary Data Sheet 2). One IO particle (of 47 total) that lacked Si instead contained Ti (Supplementary Data Sheet 2). Many of the IO particles were round spheres suspected to be coal fly ash (**Figure 7C**), a positively buoyant byproduct of coal combustion that has been reported previously in Great Lakes surface waters (Eriksen et al., 2013). Some IO particles physically resembled styrofoam balls but were confirmed to be puffed silica foam, having contained prominent mineral elements (e.g., **Figure 7D**). Non-plastic organic matter (NP) was physically characterized by stringy fibers of irregular width or shallow-relief surface patterns typical of leaf veining (**Figures 7C,F**, respectively) and chemically characterized by more complex elemental signatures with several smaller peaks rather than a single dominant C peak.

To assess our tendency to accurately classify plastic from non-plastic, we compared our initial visual classifications with those based on EDS-SEM analysis (**Figure 8**; Supplementary Data Sheet 2; Supplementary Image 1; Supplementary Table 1). Of all pieces visually identified as plastic, 76% were confirmed as P, 2% were NP, 12% could not be identified as P or NP, and 10% were IO. Of all pieces visually identified as non-plastic, 46% were confirmed as NP, 35% were IO, 11% couldn't be identified as P or NP, and 7% were plastic (**Figure 8**). A chi-squared test of independence confirms that the EDS-SEM-based plastic (P) calls occur most often in the visually-determined plastic category, followed by the P-NP class, and the EDS-SEM-based non-plastic (NP) calls occur most often in the visually-determined non-plastic category, followed by the inorganic (IO), and NP-IO ($\chi^2 = 112.63$, $p = 2.003\text{e-}23$, **Table 2**). These findings provided confidence in the visual discrimination between plastic and non-plastic particles in the smallest size class, and that rates of false-positives in both categories are similar enough that there was no need for adjustments to plastic abundances.

Lake Erie Plastic Transport Model

To develop a more holistic view of plastic transport dynamics than is possible based on discrete field collections and assess

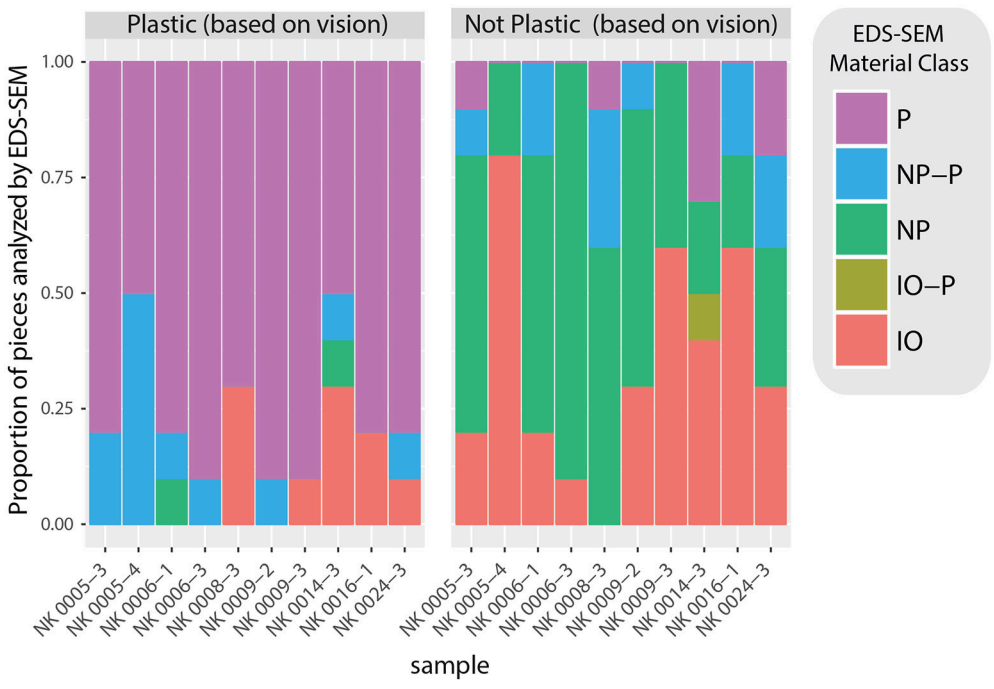

FIGURE 8 | Barplots representing the agreement between visually categorized fragments (plastic vs. non-plastic) and SEM-EDS analytically categorized fragments. SEM-EDS-based classifications fall along a spectrum of three discrete categories: plastic (P), inorganic (IO), and non-plastic organic (NP). This is a visual representation of the contingency table used in the chi-square test of independence (Supplementary Table 1).

the possibility of predicting plastic distributions, we modeled the transport of plastic and tested the effect of plastic buoyancy on the resident times in Lake Erie. Lake Erie is the smallest and shallowest of the Great Lakes, but is disproportionately surrounded by highly populated areas and used heavily for shipping and fishery industries.

No Lake Erie "Garbage Patch," but Prominent Longshore Transport Highlights at-Risk Coastal Areas

For decades, studies have described the presence of an oceanic "garbage patch" (coined in Moore et al., 2001), an amalgam of human-generated trash caught-up in the North Pacific Central Gyre that results from the convergence of floating debris in the anticyclonic eddy of the gyre's high pressure cell (Day and Shaw, 1987; Law et al., 2014). Similar anticyclonic currents form in Lake Erie in summer months (Beletsky et al., 2012) and the high concentrations of plastic in Lake Erie's eastern basin have been attributed to this feature (Eriksen et al., 2013; Driedger et al., 2015). Yet, our plastic transport model did not predict a permanent plastic "garbage patch" in Lake Erie (**Figures 9**, **10A**). This lack of a "garbage patch" may be explained by less intense convergence of surface lake currents or by the less persistent lake currents that last on the order of only weeks to months. Comparatively, stable anticyclonic circulation persists in the oceans for much longer time periods.

Results of monthly drift in summer (June, July, and August, each run over 6 years) illustrated the variability of spread due to changing current patterns (**Figure 9**). In early summer, the model generally predicted the eastward drift of neutrally buoyant particles. This was especially pronounced along both the northern and southern coasts in June, the month the majority of our field survey took place. Later in the season, the large-scale anticyclonic circulation that typically develops in mid and late summer (Beletsky et al., 2012) influenced the movement of plastics. Due to that circulation feature,

TABLE 2 | Residuals of chi-squared test of independence performed on the contingency table (Supplementary Table 3) that related the number of samples visually deemed as plastic and not plastic vs. their SEM-EDS-based classification into plastic (P), non-plastic organic (NP), and inorganic (IO) particles.

Visual-based class	SEM-EDS-based class				
	P	P-NP	NP	IO-P	IO
Plastic	5.3554386	0.147442	−4.4907312	−0.7071068	−2.6352314
Not plastic	−5.3554386	−0.147442	4.4907312	0.7071068	2.6352314

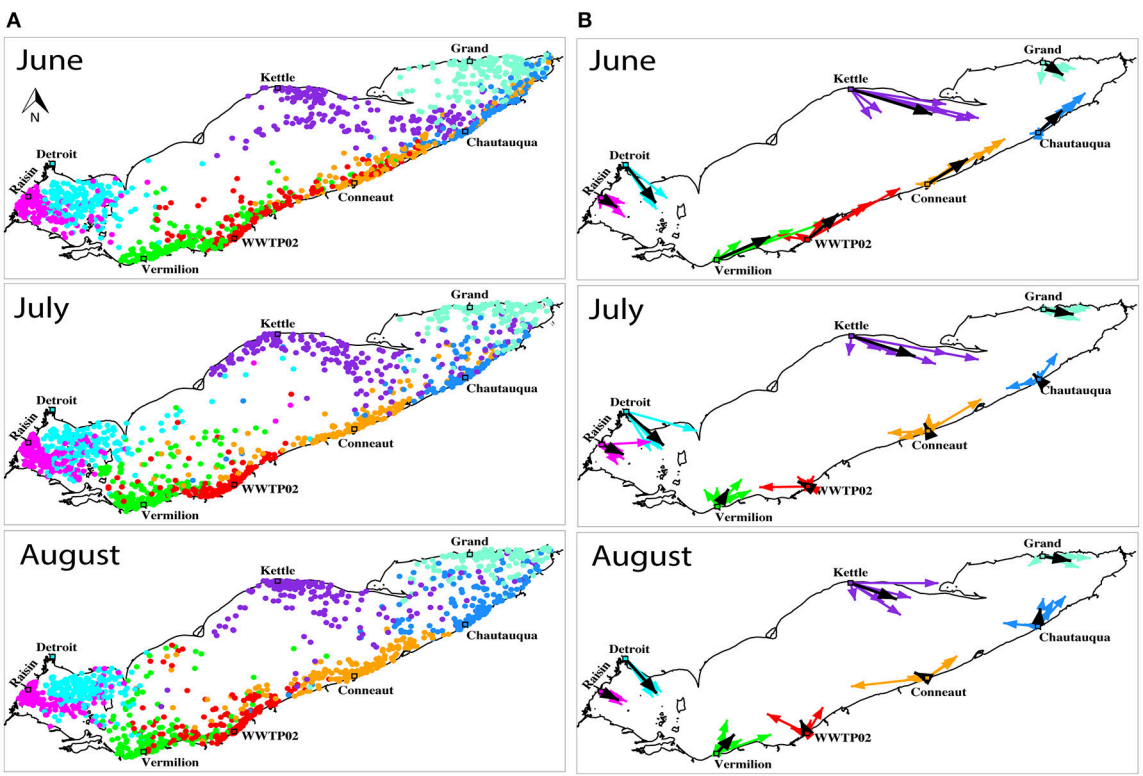

FIGURE 9 | (A) The distribution of neutrally buoyant particles at the end of month-long transport in June, July, and August for 6 years. For visual simplicity, 8 of the 29 sources (influents) are depicted: the Raisin Rv. (magenta), Detroit Rv. (cyan), Kettle Rv. (purple), Grand Rv. (turquoise), Chautauqua Rv. (blue), Conneaut Rv. (orange), Cleveland WWTP (red), and Vermillion Rv. (green). **(B)** Mean transport vectors summarizing the positions of all particles at the end of month at each of the same eight representative sources (similarly colored coded). The six vectors per source represent mean transport for each of the 6 years. The 6-year mean vector is shown in black at each input.

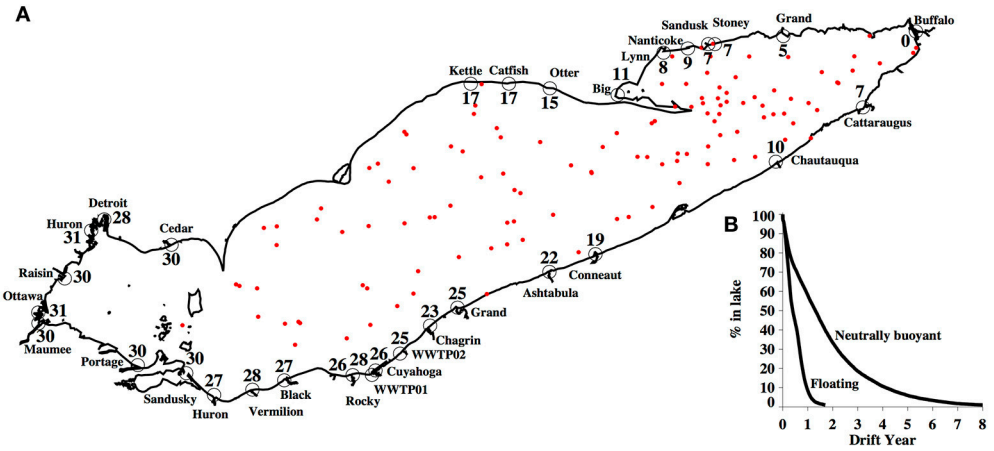

FIGURE 10 | (A) Mean residence time (months) for each source of neutrally buoyant particles. Particles were released daily for 1 year and tracked for 8 years (time for 99% of particles to leave the lake). Red dots represent the particles that remain in the lake after 8 years of drift. **(B)** Percent of particles remaining as a function of time for the neutrally buoyant (right curve) and floating (left curve) models; based on same run conditions as **(A)**.

particles released along the southern coast east of Cleveland were often transported westward (**Figure 9**). During that time, temporary patches (lasting for a few days) formed in the floating particles model. In this case, particle aggregation due to current convergence *is* expected. For example, in mid-August 2010, floating particles aggregated in a large anticyclonic gyre developed in the central basin and two smaller anticyclonic gyres in the eastern basin (Supplementary Figure 4). Overall, particles in both neutrally buoyant and floating cases exhibited general eastward drift and flushed quickly from the western basin by the Detroit River flow (**Figure 10A**). Recirculation in the central and eastern basins was especially pronounced in the summer. Neutrally buoyant particles drifted more slowly than floating particles because of reduction of current speed with depth.

Our model did not predict elevated concentrations of plastic in Lake Erie's eastern basin relative to the central basin, as seen in both our field survey (**Figure 3D**) and that of a prior study in this lake (Eriksen et al., 2013). Notably, this pattern was absent in a recent Great Lakes particle model, as well (Hoffman and Hittinger, 2017). This is despite the fact that the forcing used in the particle model presented here has superior temporal resolution (e.g., hourly vs. three-hourly) and more accurately predicts observed Lake Erie circulation patterns (Beletsky et al., 2013). For example, the winds used in the Hoffman and Hittinger particle model (NOAAs Great Lakes Coastal Forecast System model output) typically produce cyclonic circulation patterns in summer, rather than the anticyclonic patterns observed in summer (Beletsky et al., 2013). We hypothesize that model discrepancy can be either due to a temporary patch in both observational surveys or due to an elevated input near or in the eastern basin that was not accounted for in our model (e.g., Baldwin et al., 2016 documented a peak in microplastic concentration at Ashtabula, OH).

According to the neutrally buoyant particle model predictions, habitats along the southern coast of Lake Erie were predicted to be most affected by plastic pollution (**Figure 9**). The higher concentration of rivers along the southern coast led to more particles released in that area in model runs. The eastward drift of particles from upstream sources (e.g., the Detroit River and other rivers in the western basin) led to higher concentration of particles (**Figure 9**; particle release points identified by open circles and are listed in Supplementary Table 2). This interpretation is consistent with the recognition that rivers are major sources of plastics to inland water bodies (Wagner et al., 2014), including the Great Lakes (Baldwin et al., 2016). In most months, rather than moving offshore, the model predicted longshore transport from coastal sources. This model indicates that future plastic pollution mitigation and management efforts in Lake Erie should focus on its southern shore and downstream of urbanized areas. Extending this plastic transport model to the other four Great Lakes will similarly inform future efforts across this critical watershed.

Plastic Density Drastically Impacts Residence Time in the Lake

The buoyancy of modeled particles had a strong effect on residence time in the lake; floating particles flush from the lake in 1.7 years—nearly 5 times faster than neutrally buoyant particles (8.1 years; **Figure 10B**). In fact, the modeled flushing time for neutrally buoyant particles in Lake Erie substantially exceeds hydraulic residence time estimates (2.7 years; Bolsenga and Herdendorf, 1993). However, the residence time is not uniform across the lake. Average residence times of neutrally buoyant particles released at different sources show a west-east gradient (**Figure 10A**), with the shortest residence times for the particles released at the Buffalo River (less than a month) and longest for those released at the Ottawa and Huron Rivers in the western basin (over 30 months, **Figure 10A**).

Most surveys of environmental plastic pollution tend to collect samples at the water surface, capturing floating plastic only. According to this model, most of the floating plastics

sampled in the western and central basin would have been in Lake Erie for <2 years. However, while most virgin plastic used in consumer products—especially one-time use plastic (PlasticsEurope: Association of Plastics Manufacturers, 2015)—is predicted to be positively buoyant, plastic litter is readily found in sediment (Corcoran et al., 2015; Van Cauwenberghe et al., 2015; Ballent et al., 2016). This can be attributed to denser polymer types sinking, but there are other dynamic changes in the buoyant density that plastics are likely to undergo once in the environment, e.g., oxidation or biofouling. These changes are poorly described, but our results indicate the need to resolve these phenomena to effectively model the loads and fluxes of environmental plastic pollution in freshwater and marine systems alike.

CONCLUSION

This study has improved our understanding of the distribution, transport, and fate of plastics in four lakes of the Great Lakes system. As the largest freshwater system on the planet, these critical lakes hold 20% of the world's fresh water. Plastic pollution was documented down to the smallest size class yet reported, shedding light on the magnitude of plastics in a small size class (106–333 μm) that is missing from most existing reports (Hidalgo Ruz et al., 2012; Law, 2016). This led to load estimates of nearly 2 million particles km^{-2}, the highest reported levels in the Great Lakes and possibly any surface water ecosystem. These high numbers can be attributed to the high nearshore population density, a feature unique to inland waterways that does not similarly influence remote ocean basins, and the long hydraulic residence time of some of the Great Lakes (3–100s of years, depending on the lake). Given this time and the recalcitrance of plastic to degradation, fragments of some of the first plastic ever produced for the consumer market are certainly present in the Great Lakes still today. This scenario is likely representative of lakes worldwide, which account for 87% of the planet's freshwater and have an average residence time of 50–100 years[4]—indeed spanning the introduction of plastics to the consumer market.

Data describing the abundance of plastic pollution in the Great Lakes are sparse and will continue to be. Field-based quantification surveys are time-consuming, expensive, and low-throughput. As a result, there is insufficient spatial and temporal resolution of plastic debris in the Great Lakes and other aquatic ecosystems. In addition, detailed data on plastic loads (e.g., from rivers and WWTPs) are needed to determine the plastic budget and to inform future models. Integrating the modeling approach developed here will guide targeted research surveys, experiments, and technological innovation for improved understanding of the ecosystem and public health risks plastic pollution pose to freshwater systems. These are the steps needed to develop a global plastic mass balance transport model to effectively inform the policy, mitigation, and prevention initiatives needed to protect our vital freshwater resources.

Research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

[4]https://scied.ucar.edu/longcontent/water-cycle

AUTHOR CONTRIBUTIONS

MD, DB, and KW: conceived the study. MD, RC, and BL: performed field sampling, sample processing, and plastic count data collection and analyzed data. DB and RB: developed and ran the transport model and analyzed data. MD, RC, DB, and KW: wrote the manuscript.

FUNDING

This work was supported by the University of Michigan Water Center, a center of the Graham Sustainability Institute. The Water Center is supported by funds from the Fred A. and Barbara M. Erb Family Foundation and the University of Michigan. Additional funding for DB and RB was provided by NOAA GLERL. Funding was awarded to Cooperative Institute for Limnology and Ecosystems Research (CILER) through the NOAA Cooperative Agreement with the University of Michigan (NA12OAR4320071). Additional funding was provided by the NSF grant 1039043.

ACKNOWLEDGMENTS

We wish to acknowledge David Brooks for providing and operating the research vessel, Nancy *K*, for funding its operation, and for providing additional engineering and logistical support during the 2014 campaign. We thank Steve Ruberg (NOAA GLERL) and the captain and crew of R5501 for sampling in the Detroit River and the Cleveland area. Gregory Boehm, Sarah Halpern, Kat Wiles, Kaitie Benedeck, Scott Koenigbauer, Naomi Huntley, Shibhum Sidhar, Payton Watt, Andrew Wong, Emma Borjigin-Wang, Marlies Michielssen, and Elien Michielssen assisted in the massive effort of sample collection in the field and processing in the lab. Larissa Sano, Allen Burton, Jeanne Hankett, Zhan Chen, and Lorena Rios Mendoza supported project thematic development. We thank Will Bender for technical assistance with SEM-EDS. This is CIGLR 1113 and NOAA GLERL contribution 1858.

SUPPLEMENTARY MATERIAL

The Supplementary Material for this article can be found online at: http://journal.frontiersin.org/article/10.3389/fenvs.2017.00045/full#supplementary-material

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Conflict of Interest Statement: The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.