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Categorization of plastic debris on sixty-six beaches of the Laurentian Great Lakes, North America

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Supplementary material for this article is available online

Abstract

The Laurentian Great Lakes system is a major global sink for plastic debris. An area of 10 m^2 on each of sixty-six Great Lakes beaches was sampled for large micro-, meso- and macroplastic items. A total of 21 592 plastic items were collected and categorized. Pre-production plastic pellets were the most abundant debris type, accounting for 58.3% of the total count. The remaining 42.7% of the debris items are the focus of this study. Detailed, multi-step characterization was performed with the plastics being categorized using physical identification, known usage, and Fourier transform infrared spectroscopy (FTIR). Values of 805.5 items m^{-2} at Baxter Beach in Sarnia, Ontario, Canada, and 688.1 items m^{-2} at Bronte Beach in Oakville, Ontario, Canada are the highest of all sampling locations. Sampling sites on only three beaches contained no plastic debris: Bay City in Michigan, U.S.A., Presque Isle State Park in Erie, Pennsylvania, U.S.A. and Pebble Beach in Marathon, Ontario, Canada. The plastic items sampled were mainly large microplastics (68.4% of total) with a total of 1477.5 items m^{-2}, followed by mesoplastics (27.3% of total) with 598.8 items m^{-2}, and macroplastics (4.3% of total) with 91.9 items m^{-2}. By mass, the microplastic fraction accounted for 25.61 g m^2 (14.1%), the mesoplastic for 47.06 g m^2 (25.9%), and macroplastic for 109.3 g m^2 (60.1%). A total of 3004 items were determined as specific polymers based on physical properties, known polymer usage, Resin Identification Code, and FTIR. A total of 1227 plastic items (40.8% of total) were identified as expanded polystyrene. The 49 most common items, excluding pellets, were scored using a matrix scoring technique to determine their potential general origin. It was determined that these items mostly originated from shoreline and urban sources, whereas pellets originated from the plastics industry.

1. Introduction

During the last 10 years, many studies have investigated the sources of plastic pollution, plastic transport pathways, and plastic in the environment. Sources of plastic pollution are diverse and include everything from microfibres from textiles discharged as washing machine effluent (Napper and Thompson 2016) to road dust (Abbasi *et al* 2019). Pathways that enable transport from land to aquatic ecosystems include tributaries (Mani *et al* 2015, Baldwin *et al* 2016, Corcoran *et al* 2020a), wastewater treatment plant (WWTP) discharges (Mason *et al* 2016, Murphy *et al* 2016), stormwater runoff (Grbić *et al* 2020), and atmospheric deposition (Allen *et al* 2019, Brahney *et al* 2020). Once in the aquatic environment, depositional sinks include benthic sediment (Ballent *et al* 2016, Dean *et al* 2018), shorelines (Zbyszewski *et al* 2014, Hoellein *et al* 2015), and deep-sea sediment (Kane *et al* 2020).

Shoreline plastic debris has been identified globally along ocean and lake margins and the banks of tributaries. Large microplastic, meso- and macroplastic debris, defined as 1–5, 5–25 and 25–1000 mm in size (Lippiatt *et al* 2013) are preferentially studied in these locations because they are readily visible and therefore easily collected within time constraints. The majority of shoreline investigations focus on the abundance and distribution of large micro-, meso- and macroplastic debris, but very few involve multi-lake comparisons within a single freshwater system. The North American Laurentian Great Lakes (LGL) provide an excellent opportunity to investigate the sources and transport pathways of plastic debris in a complex freshwater system. The LGL contain approximately 21% of the world's fresh surface water and provide clean drinking water to tens of millions of Americans and Canadians. They are a vital resource for the economies of eight states and two provinces, which depend on the waterways for shipping, fishing, tourism, water withdrawal, and conservation (US DHS 2014). Pollution of this system with plastic is therefore of major concern for both countries.

This paper provides insight into levels of large microplastic, meso- and macroplastic items, excluding pellets on 66 beaches in the LGL and is a complementary study to Corcoran *et al* (2020b), which focused solely on the factors controlling the distribution of plastic pellets on the same beaches. Analysis of visible plastic items along shorelines provides an opportunity to characterize plastics before they degrade into secondary microplastics, which are often impossible to attribute to sources. This study is the first to quantify visible plastic items on beaches throughout the LGL using standardized sampling and characterization techniques. The major objectives were to: (a) determine and compare the abundances of plastic based on size, morphology, polymer composition, and item use, and (b) to determine the potential sources of debris using a matrix scoring technique (MST).

2. Materials and methods

The LGL are composed of lakes Superior, Michigan, Huron, Erie, and Ontario, in addition to interconnecting rivers and Lake St. Clair (figure 1). The lakes face a number of stressors, including emerging contaminants such as plastics (ECCC & USEPA (United States Environmental Protection Agency) 2019). Each lake has its own characteristic circulation patterns, which vary by season and to some degree between years (Beletsky *et al* 1999). Over 30 million people live in the Great Lakes Basin—about 10% of the US population and just under a third of the Canadian population. Academic research of plastic pollution in the LGL began approximately one decade ago when Zbyszewski and Corcoran (2011) published data from a 2008 sampling campaign that involved plastic debris on Lake Huron beaches. Since that time, three review papers have been written concerning plastic pollution in the LGL (Driedger *et al* 2015, Earn *et al* 2021, Helm 2020).

2.1. Field sampling & sample processing

Large micro- (1–5 mm), meso- (5–25 mm) and macroplastic (>25 mm) debris items were collected from 66 beaches between the period 7 October to 21 October 2018 in order to constrain the sampling to 2 weeks during one season (figure 1). No extreme storm events took place within this period. These beaches were selected according to public accessibility while trying to keep the spacing between each location as regular as possible. The number of beaches sampled on each lake was chosen according to lake size. For example, Lake Superior (largest lake) had 18 sampling locations and Lake Ontario (smallest lake) had 9. Two individuals surveyed the beaches of lakes Michigan, Erie, Ontario, Huron and 15 beaches of Lake Superior. Three individuals surveyed the remaining three beaches of Lake Superior. Strandlines—the high-water mark where organic and other floating debris preferentially accumulate—were used for the collection of stratified random samples. A 10 m measuring tape was stretched along the strandline and all large micro-, meso- and macroplastic debris (including pellets) were collected from within a 1 m wide swath. The plastic debris in the top 5 cm of the beach surface was collected by hand digging (if the sediment was wet) or digging with metal sieves (if sediment was dry and fine enough to pass through 2.5 × 3 mm openings). Where sieving was used, any items that fell through the openings but were >1 mm in size were handpicked. Very large items (>100 mm) were generally not collected and were instead recorded in field notes. All items were brushed by hand to remove loosely adhered organic matter and sediment. Plastic films and bags with large amounts of adhered debris were brushed and rinsed with reverse osmosis water, as were bottle caps and other items filled with sediment.

Samples were then placed on an aluminum foil tray and dried in an oven set at 70 °C for at least 24 h, until dry. Cleaned and dried samples were sieved for 5 min at 50 Hz. The samples were separated into size fractions (1–5 mm, 5–25 mm, and 25–1000 mm) in accordance with size fractions listed in table 2.2 of GESAMP (2019). Prior to weighing, care was taken to remove all non-polymeric debris that had co-accumulated with the collected plastics, including organic matter (e.g. roots, aquatic vegetation, bugs), coal fragments, shells, slag, sea glass, and tar. Each size fraction was weighed separately for each beach location using a Mettler-Toledo XS204 analytical balance. Detailed pellet data are provided in Corcoran *et al* (2020b). Pellet size was represented as area in mm^2 (length × width), and therefore it was assumed for this study that any pellet with an area of <19.6 mm^2 fell into the 1–5 mm fraction, whereas pellets >19.6 mm^2 were assigned to the 5–25 mm fraction. A random sample of pellets from each lake was weighed and the average pellet mass was determined

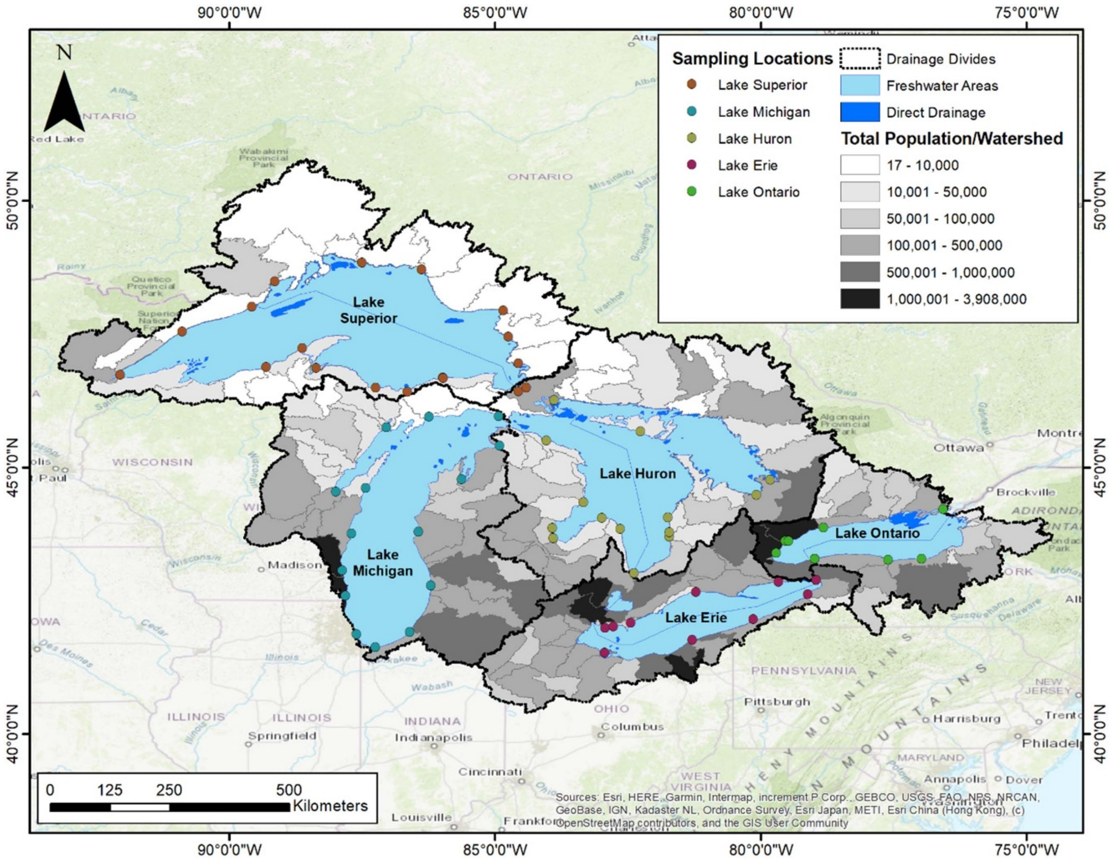

Figure 1. Sampling locations on 66 Great Lakes beaches where plastic items were collected in October 2018. Reprinted from Corcoran *et al* (2020b), Copyright (2020), with permission from Elsevier. CC BY-NC-ND 4.0.

to be 0.021 g. This mass was applied to the total number of pellets on each beach and lake.

2.2. Visual identification & characterization

All large micro-, meso- and macroplastic items were characterized according to: (a) size, (b) morphology (following GESAMP 2019), (c) colour, and (d) general item description. Table 1 lists the characterization categories and variables applicable to this study. A complete item database is provided in table S1 (available online at stacks.iop.org/ERL/17/045008/mmedia). Item use was generally obvious (e.g. bottle caps, cigarette butts), but if unknown, internet searches were utilized, and product catalogues and guides were referenced when applicable. If an item was not identifiable, crowdsourcing was used by posting photos and item descriptions to the site www.reddit.com/r/whatisthisthing/. Polymer types were determined through combinations of four methods: (a) physical properties for polymers that displayed unique visual and/or tactile characteristics (e.g. expanded polystyrene (EPS) foams and polypropylene bottle caps), (b) known polymer usage (e.g. cigarette butts = cellulose acetate (CA)), (c) consumer goods with resin identification codes 1–6 (ASTM International 2020), and (d) manufactured products with stamped/injection moulded polymer names (e.g. a car part with >TPO< indicating thermoplastic polyolefin). Other associated materials were recorded, such as adhesive on clear tape or an aluminum layer on packaging. Polymer identification was not attempted for multilayered packaging, which can be composed of up to 12 layers of varying polymers and metals (Daley 2020). Brand names, product names, and product text were noted, where possible, in order to determine parent companies. Items identified by use were categorized according to three different lists: (a) the Convention for the Protection of the Marine Environment of the North-East Atlantic (OSPAR) Commission, (b) the National Oceanic and Atmospheric Administration (NOAA), and (c) Break Free from Plastic (BFFP). Each of these categorization lists provide a different, but useful way to group plastic debris items for future analyses. Using a multi-pronged approach to visual identification and characterization (physical characteristics, item use, polymer type, product name, parent company name) leads to more reliable results than if one method is used alone. In addition, this approach enables clear reporting of sources of plastic debris.

2.3. Chemical composition

Certain plastic items were selected for Fourier transform infrared spectroscopy (FTIR) to: (a) provide evidence for use/source/origin of suspected items (e.g. plasticized vinyl, foam insulation board), (b)

Table 1. Categories assigned to plastic debris items, as applicable. See table S1 for complete sample database. Adapted from Arturo (2021). CC BY 4.0.

	Category	Variables
All Samples	Size Fraction	• 1–5 mm (large micro) • 5–25 mm (meso) • 25–1000 mm (macro)
	Intact	• Yes • No • N/A
	Morphology	• Pellet (P) • Fragment (FR) • Foam (FO) • Fibre (FB) • Textile (TX) • Film (FL) • Rubber (RB) • Non-plastic (NP) • Multiple morphologies
When Applicable	Colour	Numerous
	General item description	Numerous
	Item use	Numerous
	Polymer	Numerous
	Polymer identification method	• Physical identification (PID)/known usage (KU) • Resin identification code (RIC)/> stamped< • FTIR
	Other materials	Numerous (e.g. paper on cigarettes, aluminum on packaging)
	Brand name/Product name/ Product text	Numerous
	(Parent) Company name	Numerous
	Year	Numerous (Based on copyright, manufacturing date, etc)
Based on Item Use	OSPAR commission 100 m category	See OSPAR Commission 2010
	NOAA category	See Opfer *et al* (2012)
	BFFP category	See BFFP (2019b)
	General Waste Regulatory category	• Municipal Solid Waste (MSW) • Medical Waste • Household Hazardous Waste (HHW) • Special Waste • e-Waste

determine the polymers used in common items (e.g. artificial flower petals and leaves), or (c) confirm polymer type determined by visual identification or known item use. Informative FTIR was conducted on 63 samples at Surface Science Western, Western University, using a Bruker Tensor II FTIR Spectrometer (Bruker Optik GmbH, Ettlingen, Germany) with the Platinum attenuated total reflectance (Pt-ATR) diamond attachment in the main box. This setup allowed for a 2×2 mm $\times$ 0.6–5 μm depth area of analysis on each sample. Spectra were collected between 4000 and 400 cm^{-1}, with a 4 cm^{-1} resolution, 16 scans per sample and were detected using a RT-DLaTGS mid-infrared detector.

2.4. Parent company

A parent company audit, modeled after BFFP (Break Free From Plastic) (2018) and BFFP (2019a), was performed by examining all plastic items for any logos, product names, trademarked product designs, ZIP/Postal Codes, or other evidence of company ownership. Internet searches were performed, including searches of the Open Food Facts Database (https://world.openfoodfacts.org/) and company 'About Us' pages to determine brand ownership and ultimately parent companies (table S1). Generic trademarks were not used in the database to avoid false attribution. For example, flashspun HDPE was used instead of 'Tyvek' and polytetrafluoroethylene (PTFE) tape was used instead of 'Teflon tape'. If a company was acquired, dissolved, merged, or spun-off, the current company that assumed liabilities and assets was named in the database. For defunct companies without clear acquisitions, 'defunct' was noted. Brands identified were either manufacturers of the original plastic items (e.g. Caplugs end caps), provided parts to a finished product (e.g. CSI bottle caps for drink bottles), were contracted companies (e.g. Independent Coke Bottlers), or were licenced/advertised items (e.g. Disney adhesive bandage, pen with resort advertising). Certain identified items could have multiple companies assigned to them, and therefore all companies were listed.

2.5. Spatial & statistical analyses

Data cleanup and aggregation were performed in Microsoft Excel 16.0 and spatial analysis and mapping were done using ArcGIS 10.8.1. Methods for the human population by watershed analysis can be found in Corcoran *et al* (2020b). Exploratory plots, final plots, and statistical analyses were performed using either MATLAB R2020b (9.9.0), JMP 15.0, or Microsoft Excel 16.0. A variety of statistical analyses were performed in MATLAB R2020b and ProUCL 5.1. Outlier removal was not performed because it was determined that outliers, if present, were indicative of the nature of the respective locations and were not the product of inadvertent bias. When possible, data that were not normally distributed were transformed to meet the requirements to run parametric tests. For continuous, bivariate data, Student's t-tests or Wilcoxon–Mann–Whitney Tests were performed depending on distribution. For continuous, multivariate data, ANOVAs or Kruskal–Wallis Tests were performed depending on whether data were parametric or nonparametric. Post hoc tests and correlation analyses were performed as needed.

2.6. Matrix scoring technique (MST)

An MST modified from Tudor and Williams (2004) was utilized for this study to determine the probable origin of plastic items from three general sources—shoreline recreation, sewage related debris (urban sources), and agriculture. An MST allows for the apportionment of individual plastic items to multiple sources based on likelihood of origin, through a multi-step process. Unlike an indicator item characterization technique in which items are assigned to a singular source, an MST has the advantage of considering the relative likelihoods that plastic items can originate from multiple source areas. For example, cotton bud sticks could originate from shoreline recreation (such as beach users) or from urban areas (such as combined sewer overflows). The first step was to describe the items by material. This study utilized the 49 most common items (excludes pellets) in the 'item use' column in table S1. Next, an elimination list was formed based on factors including the nature of sampled beaches, co-accumulation of certain items, and proximity to potential sources. The elimination list facilitates the elimination of certain items by considering variables including function of items, item mix, and beach location (Earll *et al* 1999). For example, balloon strings could be eliminated from consideration for originating from agriculture, but would be expected to originate from shoreline recreation or urban sources. Items were then allocated to source sectors (e.g. shoreline recreation, urban areas, agriculture). Qualitative Likelihoods were assigned numerical scores: Very likely (LL) = 16, Likely (L) = 2, Possible (P) = 1, Unlikely (U) = 0.5, Very Unlikely (UU) = 0.125, Not Considered (NC) = 0 (Earll *et al* 1999, Tudor and Williams 2004). Although some studies have applied the elimination list separately to each sampling location, it was not feasible to do so in this study with so many sampling locations and a lack of in-depth knowledge for each location. Once all item categories considered were assigned numerical scores based on their relative probabilities of originating from respective sources, this weighted average was multiplied by the number of items per category. The final scores are a percentage for which each source contributes to the plastic items in the study area.

3. Results

A total of 21 592 large microplastic (1–5 mm), mesoplastic and macroplastic items, weighing 1 819 g, were collected from 66 beaches. Three sampling locations contained no visible debris: H-BaC-SL (Bay City), E-PIP-SL (Presque Isle State Park), and S-Ma-SL (Marathon) (figure 2). When considering all sampling locations, no outliers were identified at 1% significance for log-transformed data using a Rosner's Test for Outliers for count per m^2 and mass per m^2. Using a non-parametric one-way ANOVA (Kruskal–Wallis Test), there were no significant differences for item count ($p = 0.425$) or mass ($p = 0.837$) between the lakes at $\alpha = 0.05$. A two-sided Wilcoxon-Mann-Whitney Test was performed at $\alpha = 0.05$ to compare locations in the US to those in Canada. Beaches from the US and Canada are from the same population with regard to both item count ($p = 0.886$) and mass ($p = 0.974$).

3.1. Morphology, colour & size

The top three morphologies (pellets, fragments, and foams) account for 95.0% of all plastic items by count (figure 3). For future reproducibility and to facilitate comparison between studies, the categories of films, fibres, rubbers, and textiles were also considered, as per the GESAMP (2019) definitions. These categories collectively contained 1005 items. Additionally, 72 items were classified as multiple morphologies (e.g. adhesive bandages).

A total of 8986 items were classified by colour (colour was not recorded for 11 items; colour data for pellets are in Corcoran *et al* (2020b)). Colour categorization is important because some colours are favored by certain organisms (Santos *et al* 2016). In addition, future attempts to correlate secondary microplastics with visible debris items will probably use colour as a correlating factor. Moreover, certain products can be readily identified by colours that are unique to the product design. Colour interpretation

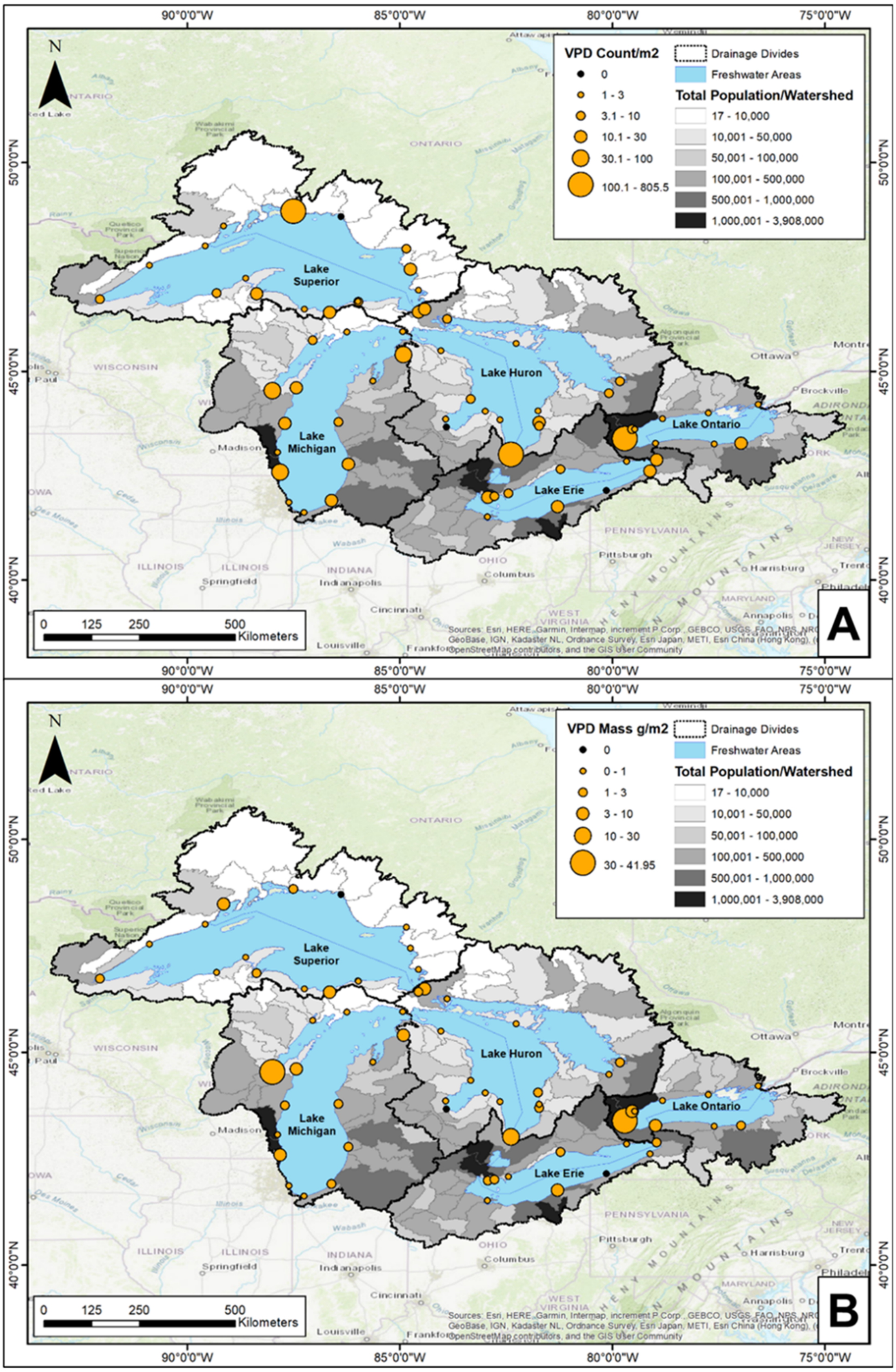

Figure 2. Abundances of visible plastic debris from sampled quadrats on each lake in: (a) count per m² and (b) mass per m². Pellet data and population by watershed are from Corcoran *et al* (2020b). Reproduced from Arturo (2021). CC BY 4.0.

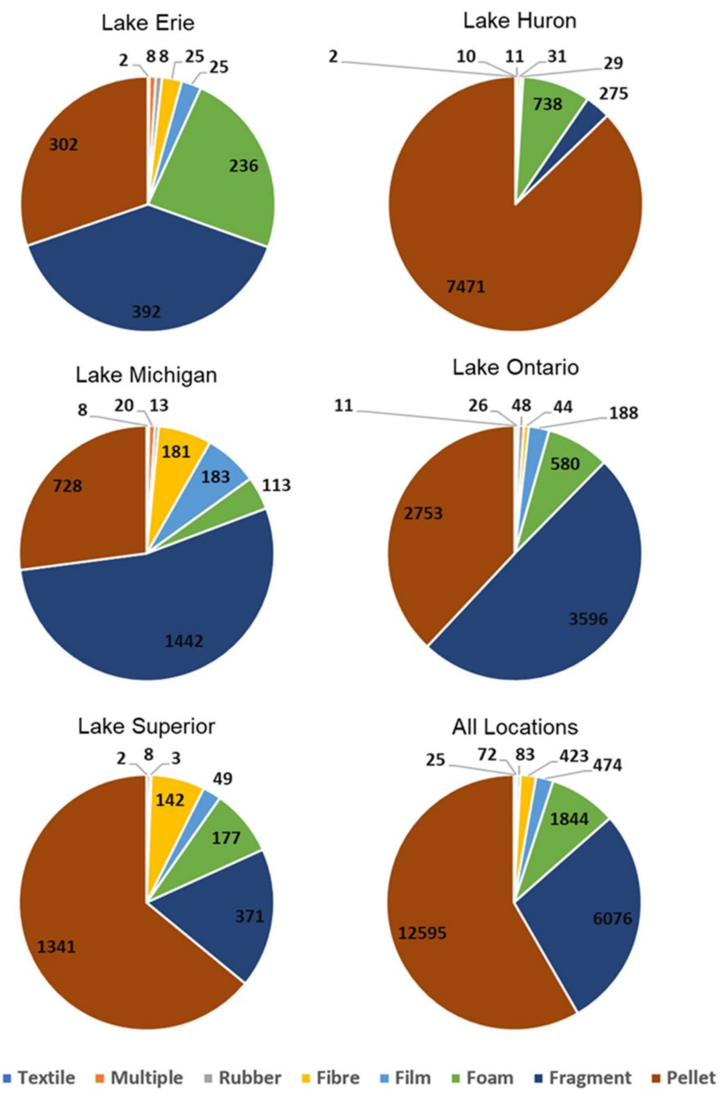

Figure 3. Pie charts of total plastic item count by morphology and lake. Morphology definitions from GESAMP (2019). Adapted from Arturo (2021). CC BY 4.0.

is a relatively subjective metric, and therefore colours were assigned to colour groupings (table S1). White/off-white coloured items were the most common, accounting for 46.4% (4170 items). The 21 592 items, including pellets, are represented by 14 775 (68.4%) large microplastics, 5898 (27.3%) mesoplastics, and 919 (4.3%) macroplastics (figure 4). By mass, the large microplastic fraction accounted for 256.1 g (14.1%), the mesoplastic for 470.6 g (25.9%), and macroplastic for 1926 g (60.1%) (figure 5 and table S2).

3.2. Polymer type

Polymers were assigned through multiple visual methods whenever possible. A total of 9025 plastic items were able to be categorized. One third of the items (3004) were assigned a polymer type through a variety of methods (figure 6) whereas the balance were flagged as 'Unknown' in the database (table S1). The most common method employed was identification by physical properties (PID) or known polymer usage (KU). Forty-three polymers were determined based on RICs or stamped/injection moulded polymer names (>stamped<). Informative FTIR spectrometry was performed to positively identify additional polymers. Although EPS, extruded polystyrene (XPS), and rigid PS are all the same polymer, they are listed separately because differentiation aids in determining sources and item use. In addition, the different properties of EPS, XPS, and rigid PS can affect their environmental fate, transport, and preferential ingestion by certain organisms. Polymers flagged with an asterisk (*) indicate undifferentiated polymers, which are considered polymers that can be narrowed to a polymer grouping but not a specific polymer type. For PE, when a low density or high density differentiation could not be made, the items were assigned PE*. For items identified as nylon (such as clothing tag fasteners and zip ties), differentiation between nylon 6 and nylon 6,6 could not be made visually. Polyurethanes (PU) are a group of structurally similar polymers,

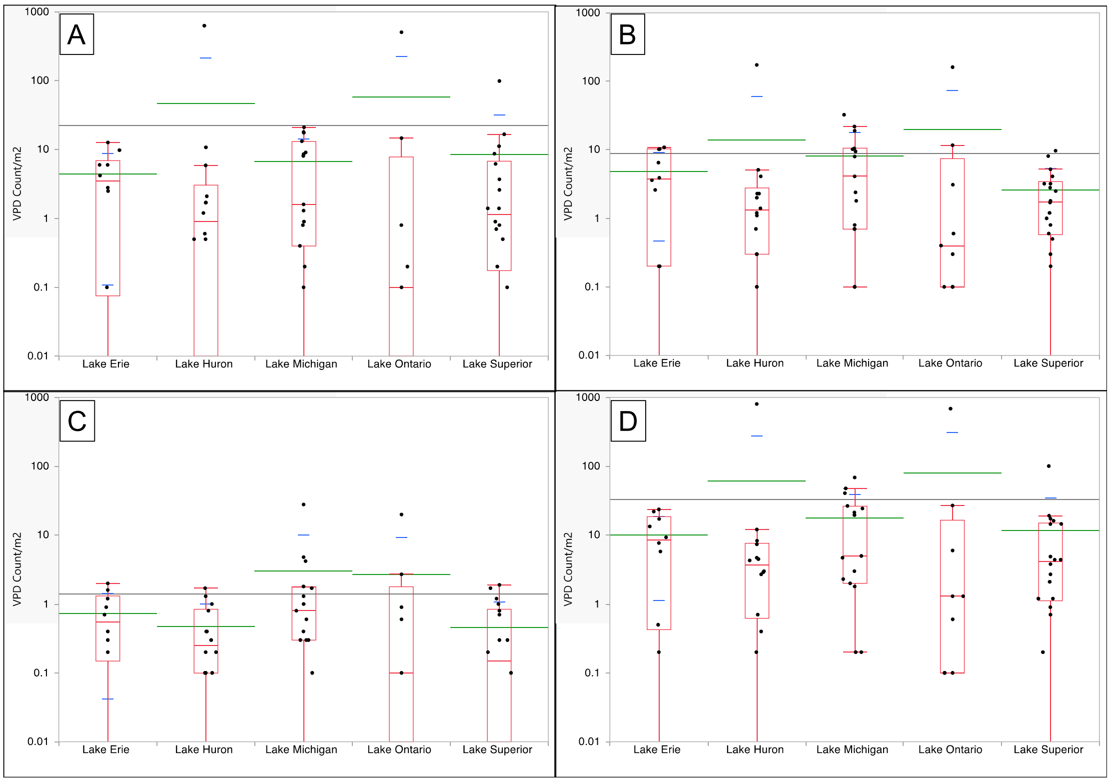

Figure 4. Box plots of plastic item count by lake for: (a) 1.0–5.0 mm, (b) 5.0–25 mm, (c) >25 mm, and (d) all size fractions. Green bars are means; blue bars are standard deviation. Adapted from Arturo (2021). CC BY 4.0.

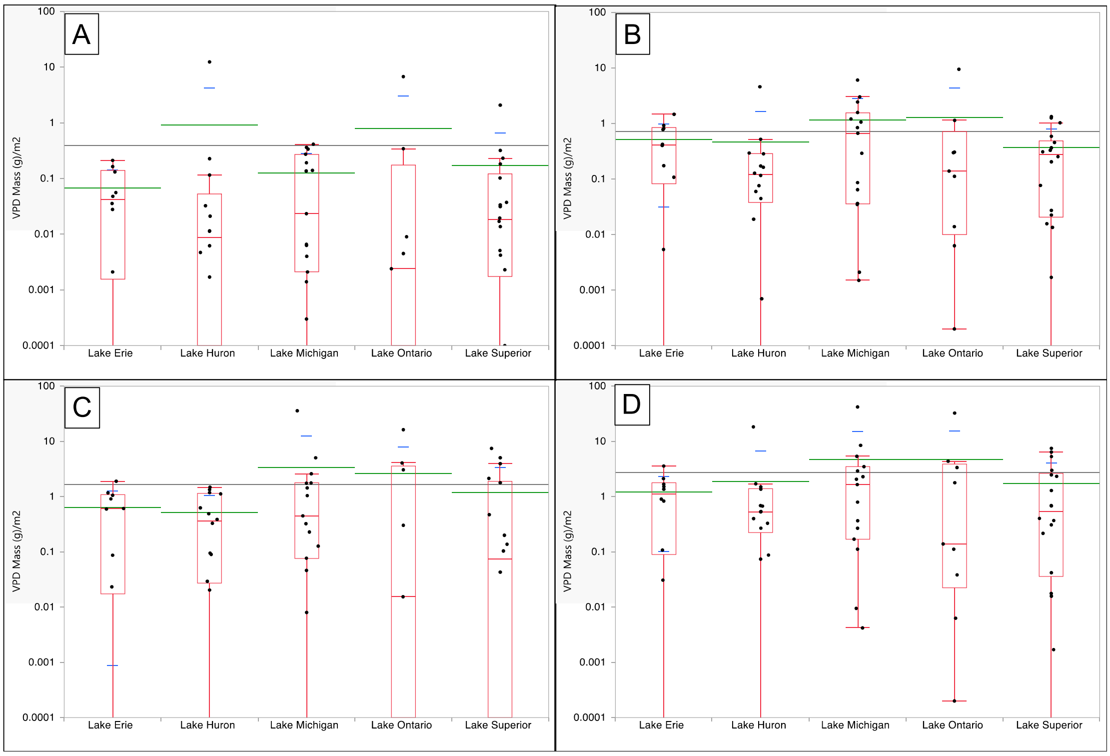

Figure 5. Box plots of plastic item mass by lake for: (a) 1.0–5.0 mm, (b) 5.0–25 mm, (c) >25 mm, and (d) all size fractions. Green bars are means; blue bars are standard deviation. Adapted from Arturo (2021). CC BY 4.0.

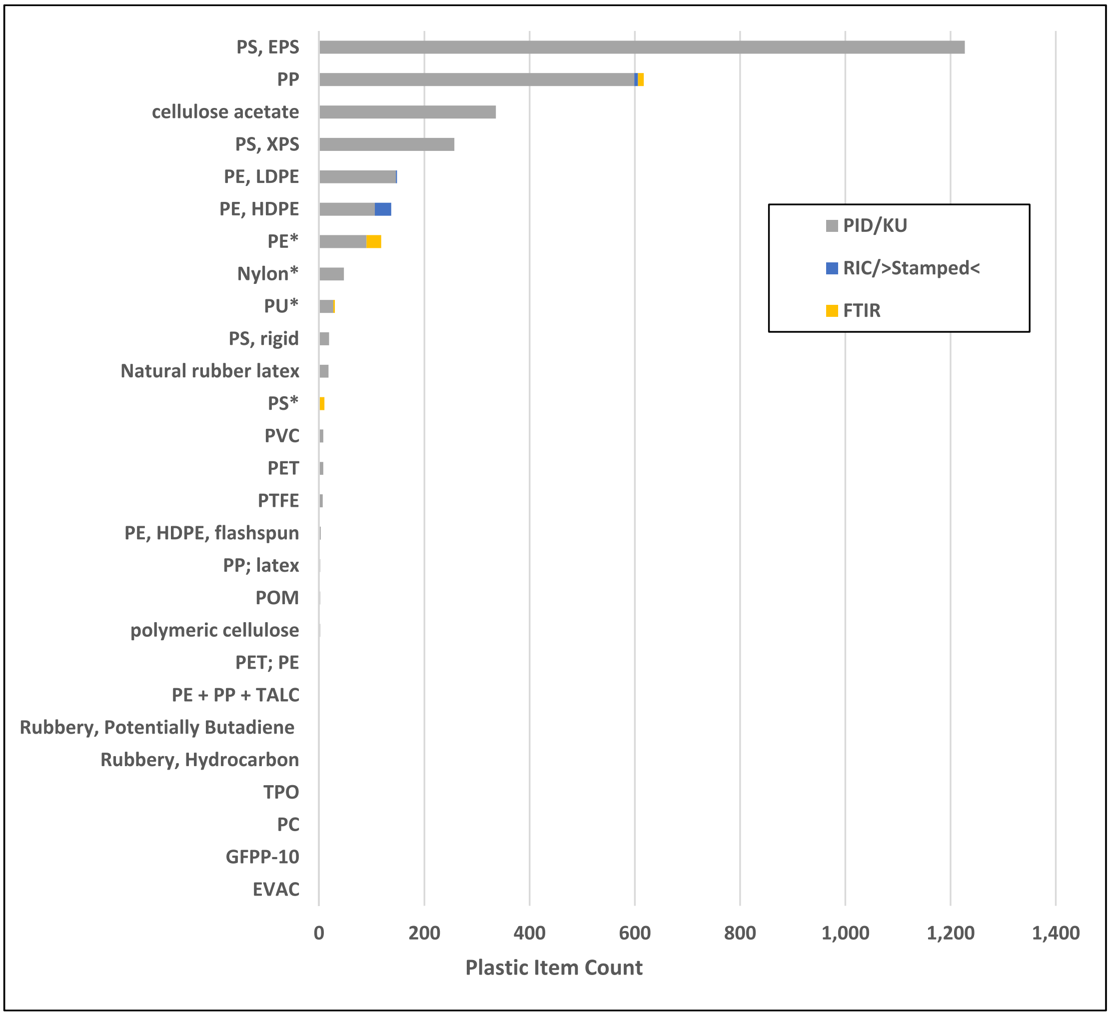

Figure 6. Plastic item count by identified polymer grouping and determination method. Excludes pellets; *Undifferentiated. PID: physical properties, KU: known polymer usage, RIC: resin identification code. Adapted from Arturo (2021).

which again, could not be differentiated using visual methods. See figure 6 for results.

Of the 3004 items identified, 40.8% ($n = 1227$) were composed of EPS, about double that of PP (20.5%; $n = 617$). All EPS items were initially identified through PID and/or KU because of the unique visual and tactile nature of EPS foams, whereas select PS foams were confirmed by FTIR. CA items comprised 11.2% ($n = 336$) of identified polymers. All CA items were cigarette butts. Two hundred fifty-seven items (8.6%) were composed of XPS, 148 (4.9%) of LDPE, 137 (4.6%) of HDPE, 118 (3.9%) of undifferentiated PE, 47 (1.6%) of undifferentiated nylon, 30 (1.0%) of undifferentiated PU, 19 (0.6%) of rigid PS, 18 (0.6%) of natural rubber latex, 10 (0.3%) of undifferentiated PS, 8 (0.3%) of PVC, and 8 (0.3%) of polyethylene terephthalate (PET) (figure 6). The remaining items combined represent under 1% of items identified by polymer: PTFE, flashspun HDPE, an item containing PP and latex, polyoxymethylene, polymeric cellulose (celluloid), an item containing both PET and PE, PE + PP + talc, rubbery hydrocarbon, rubbery substance (potentially butadiene), thermoplastic polyolefin (TPO), polycarbonate (PC), glass fibre reinforced PP, 10% (GFPP-10), and ethylene-vinyl acetate copolymer (EVAC).

3.3. Item use & parent company

Of the 21 592 plastic items, the top 25 identified by known usage accounted for 72.1% of the total items collected ($n = 15\,559$). Preproduction plastic pellets accounted for over half of the total items ($n = 12\,595$; 58.3%). The top 50 visible debris types by item use (including 'unidentified' items) account for 97.8% of the total ($n = 21\,115$) (figure 7). By ranking the top 25 items by use for each size fraction, the first and second ranked items for the large microplastic and mesoplastic size fractions are pellets and unidentified fragments respectively (table S1). The most common macroplastic items were unidentified fragments followed by plastic straws. Of the 279 items that were identified by brand name, product name, or product text, 264 items (94.6%) were positively attributed to 83 different parent companies. The top 12 parent companies by number of items identified are listed in table 2. Note that no microplastics were

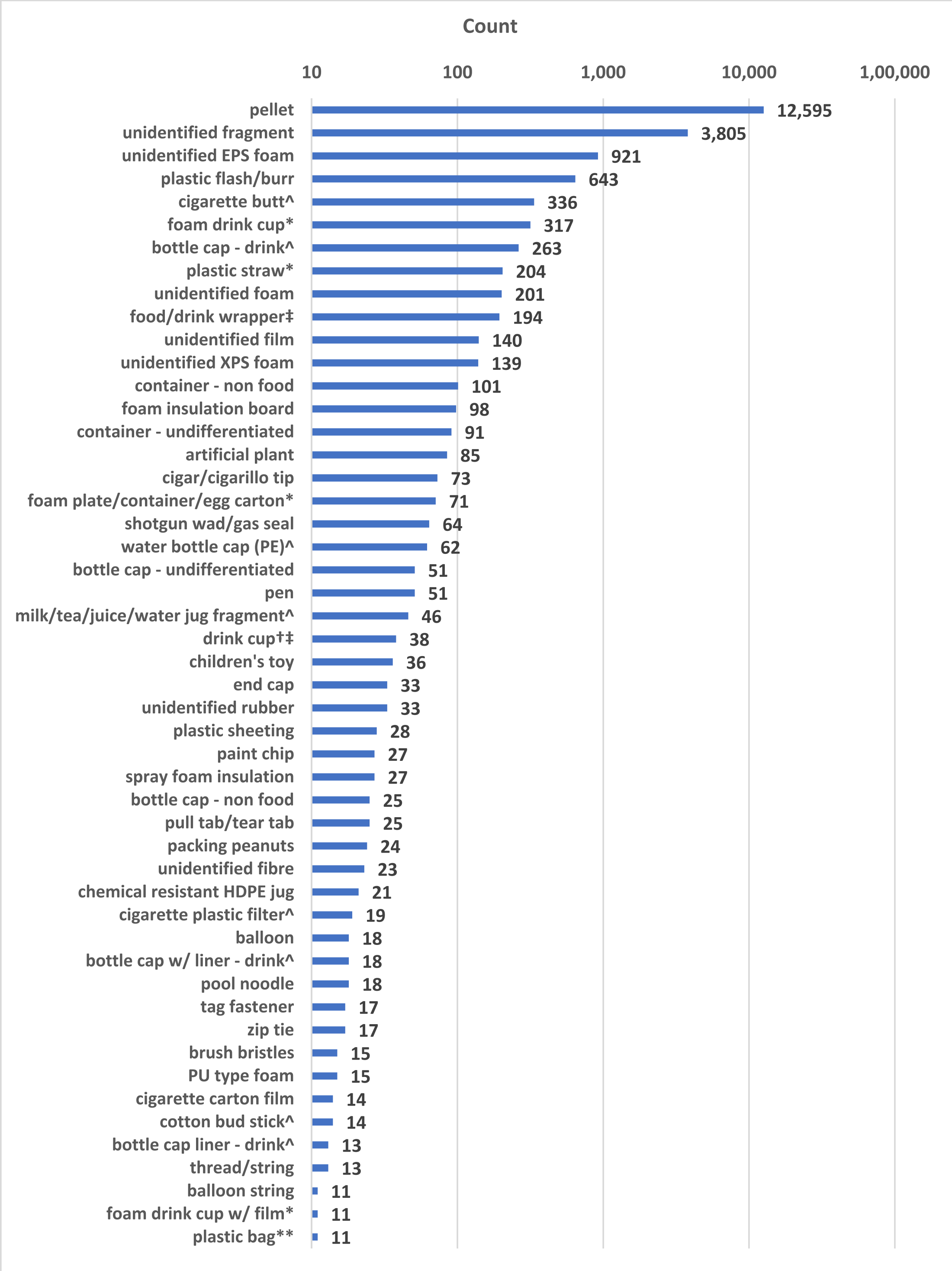

Figure 7. Bar chart in log scale showing the top 50 items by use. *GC proposed ban, **2020 NYS ban & GC proposed ban, †GC proposed reusable products/systems incentive, ‡GC proposed material specifications, ^GC proposed EPR/collection/recycling requirements. Reproduced from Arturo (2021). CC BY 4.0.

linked to parent companies due to their often fragmented and oxidized nature. Single-use plastics are the most common items for every company on the top 12 list.

3.4. Results of MST

Pellets were excluded from the MST because their general source sector is already known (the plastics industry). By applying a weighted percentage based on total item count and probability phraseology, it was determined that an estimated 41.8% of plastic items considered in the MST originated from urban sources (sewage related debris), followed by 39.1% from shoreline recreation, and 19.1% from agriculture (table S3). Note that sewage related debris can include industrial (and thereby the plastics

Table 2. Most frequently identified parent companies from branded items. Adapted from Arturo (2021). CC BY 4.0.

Rank	Parent company	Most abundant item	Item count		
			5.0–25 mm	25–1000 mm	Sum
1	CORVAGLIA MOULD AG (cr-cap brand)	Bottle caps	—	30	30
2	R.J. Reynolds Tobacco Company	Cigarettes	23	—	23
3	Altria Group, Inc. (p.k.a. Philip Morris Companies, Inc.)	Cigarettes	18	4	22
4	PepsiCo, Inc.	Bottle caps	5	12	17
5	Cerberus Capital Management, L.P. (CSI brand)	Bottle caps	4	12	16
6	The Coca-Cola Company	Bottle caps	7	6	13
7	BERICAP Holding GmbH	bottle caps	—	11	11
7	British American Tobacco	Cigarettes	11	—	11
9	Kraft Heinz Company	Food wrappers	1	7	8
9	Mars, Incorporated	Food wrappers	1	7	8
11	Grand River Enterprises	Cigarettes	6	—	6
12	Philip Morris International Inc.	Cigarettes	5	—	5

industry) because industrial land use is often located in urban areas.

4. Discussion

The widespread presence of plastic debris items on Great Lakes beaches is yet another indicator that we are in the proposed Anthropocene epoch; a geological time during which human activity has impacted Earth's natural systems. Large microplastic, mesoplastic and macroplastic debris were found within sampling quadrats of 63 of the 66 beaches. Item counts ranged from 0 to 805.5 items m^{-2} and item mass from 0.139 to 42.0 g m^{-2}. All of the 66 beaches in this study were sampled for plastic debris along the strandline where organic matter and plastic materials co-accumulate. Once deposited on the beach, plastic items are prone to degradation chemically and mechanically, which results in discolouration, crazing, and breakage (Corcoran 2021). The plastic items in the present study show typical evidence of degradation by sunlight and reworking, but also exhibited other degradation features such as bites and burning (figures 8(a) and (b)). Molten plastics that form around sediment, rocks, shells, wood, and other natural debris can solidify into plastiglomerate (Corcoran *et al* 2014), examples of which were found in this study. Some metal items with a polymer component were observed to be pedocemented (cycles of iron leaching and precipitation in terrestrial soils) (figure 8(c)). Plastics can also be degraded into flash/burrs by either plastics machining operations or other causes, including household activities (figure 8(d)).

4.1. Polymer identification method

This study represents the first of its kind to use multiple visual identification methods with FTIR to identify polymers, and expands on existing methods from Blettler *et al* (2017). Only 14.9% of 174 publications on marine debris identified by Serra-Gonçalves *et al* (2019) determined polymers. Unlike small microplastics that are more easily characterized by colour, shape, morphology, and size, visible plastic items can be categorized in many different ways. Large microplastic, mesoplastic and macroplastic debris have the advantage of being large enough to be seen in three dimensions, which allows item use to be recognized, and RICs and stamped/injection moulded polymer names to be identified. Visual identification methods employed in this study can also be utilized in low- and middle-income countries, where access to spectroscopic analytical equipment might be more limited or by non-governmental organizations with limited funding.

Polymer determination was attempted for every visible item in this study, and thus those that were more easily identifiable are overrepresented, whereas those that were more difficult to identify are underrepresented. For example, white foam spheres were confidently identified as EPS. In addition, lower density (and therefore positively buoyant) materials such as foamed polymers are also overrepresented in this study, as they preferentially float on the water surface and are cast onto shorelines. However, this study was not designed to submit a random, representative sampling of plastic debris for spectroscopic analysis. Instead, the study functions more as a proof-of-concept for multiple methods of visual identification, which can be expanded upon and replicated in future studies. An open-source polymers database by item use/type can be developed and expanded from this study and applied to the US and Canada.

4.2. Parent company

The vast majority of plastic items were not tied to companies because of their weathered nature. In addition, certain brands were overrepresented because they were readily identifiable. Remington Peters shotgun wads, marked with 'REM PET

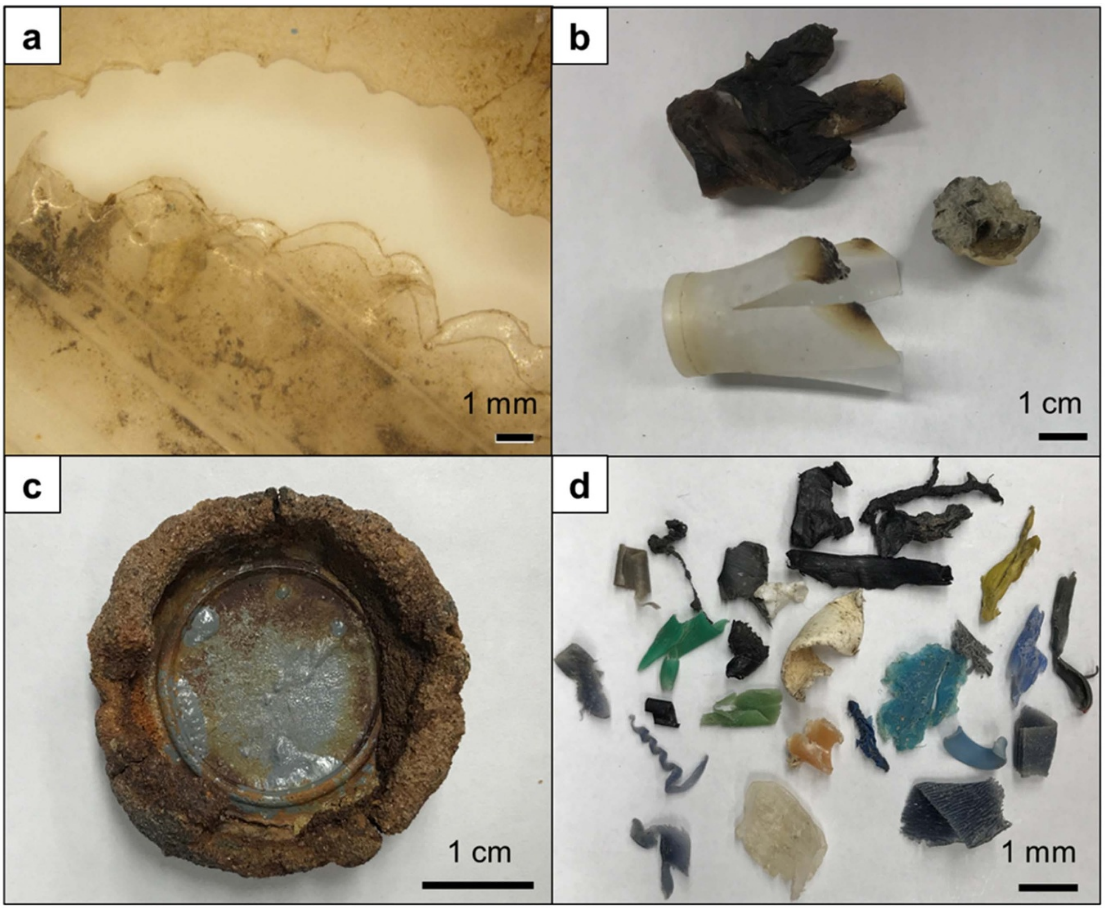

Figure 8. Examples of unconventionally degraded debris: (a) PE zip bag fragment and unidentified plastic fragment with possible fish bites, (b) partially burned shotgun wads, (c) a pedocemented steel bottle cap with bubbled polymer liner, and (d) a diverse assortment of flash/burrs. Adapted from Arturo (2021). CC BY 4.0.

PATENTED', were easily identified, whereas no other shotgun wads were identified by brand. As a result, only 2 of 64 shotgun wads/gas seals were tied back to manufacturers. Some branded items are manufactured and sold in multiple countries by different companies and therefore these were assigned to the respective company from the country of the beach on which they were sampled. For instance, Marlboro cigarette butts collected from Canadian locations were attributed to Philip Morris International Inc. whereas Marlboro cigarette butts from US beaches were attributed to Altria Group, Inc. All parent companies listed in this study have been determined to the best of our knowledge. At the time of writing, there are no legal precedents for determining corporate responsibility or regulatory enforcement for often diffuse, consumer littered items, especially when multiple industrial sectors are involved.

There are a number of companies that were in the top 12 most identified by item count in this study and also in the top 12 in 2018 and 2019 BFFP brand audits for the US and Canada. Philip Morris, PepsiCo, Inc., The Coca-Cola Company, Kraft Heinz Company, and Mars, Incorporated were in the top 12 in this study and at least one of the BFFP studies in the US or Canada. Throughout the three BFFP subgrouping lists, there is only one cigarette company (Philip Morris) and no closure (e.g. cap, lid) brands. At first this might seem surprising, given that cigarette butts and bottle caps were the 1st and 4th most commonly collected litter types in the 2019 Great Canadian Shoreline Cleanup (GCSC 2019). In fact, cigarette butts are considered the most littered item on Earth, with a significant portion of the ~6.25 trillion cigarettes consumed in 2012 ending up in the environment (WHO 2017). However, cigarette butts are often weathered enough that no paper remains, making company attribution all but impossible. Bottle caps and other closures are usually marked with the manufacturer/brand name/symbol (e.g. 'CSI', cr-cap') on the cap liner in a non-descript way that would not be noticed by the average beach cleanup volunteer/-citizen scientist conducting a brand audit. This study provides an important framework for company attribution of items which appear to be frequently missed and therefore undercounted in such audits. Institutions that conduct global brand audits, such as BFFP, should consider including a country-specific photo guide of company symbols which appear on seemingly unmarked items, while the organization could also collect subsamples of plastic debris for detailed inspection of items <25 mm.

4.3. Non-categorized items

A variety of items identified by use did not fit into typical OSPAR Commission categories, and instead were placed in the categories of 'other'. In total, 108 item uses were identified that fell into one of the OSPAR 'other' categories. Although some of these items were fairly obscure, others were frequently detected, such as artificial plants and end caps. Plastic flash/burrs were the fourth most frequently identified item ($n = 643$) in this study (figure 8(d)). Despite their ubiquity, there is very little in the literature about the presence of these items in the environment. Either flash/burrs were over represented on the beaches sampled, or other studies have grouped these items with unidentified fragments. Of the 643 flash/burrs, 555 (86.3%) were from Bronte Beach, Lake Ontario. Although some of the particles were diverse, suggesting multiple places of origin, some of the flash/burrs had visual similarities that could suggest a common origin. The ubiquity of certain items which were sparse in the literature and other categorization schemes points towards the need for a Great Lakes-specific categorization guide.

4.4. General sources

To the best of our knowledge, this is the first study of plastic debris in a lacustrine system to utilize an MST to attribute plastic items to sources. Results from the MST suggest that the majority of non-pellet items collected in this study originate from either urban areas (41.8%) or shoreline recreation (39.1%). Although other sectors were considered for the MST, ultimately only three were considered. This is because indicator items suggesting influence from shipping, commercial fishing, illegal dumping, and other sectors were not present. This should not be interpreted to mean that none of the items collected were from those sectors, but rather that applying them to the MST introduced too much noise into the matrix and resulted in erroneous values. The results from the three source sectors are consistent with other studies that have examined sourcing of plastic debris items to broad sectors. Multiple Great Lakes beach studies have found that consumer goods are the dominant type of litter (Hoellein *et al* 2014, 2015, Vincent *et al* 2017). Total anthropogenic, smoking related, and food related litter were all significantly correlated to adjacent land use for beaches analyzed by Vincent *et al* (2017).

4.5. Implications

An estimated 40% reduction in plastic loading to the environment from 2016 levels is expected by 2040 if all feasible solutions to reduce plastic pollution are implemented (Lau *et al* 2020). An encountered paradox is the increase in reuse/recycling at the expense of increasing the dispersion and loading of certain chemicals of concern (COCs). Examples include the use of biosolids to supplement traditional fertilizers (increasing COCs and microplastics in agricultural soils), PU foams reused for carpet backing (resulting in PBDEs in indoor air/dust), and crumb rubber from shredded tires used in artificial turf fields (increasing VOC emissions and rubber microparticles entering waterways). Certain plastic items actually help to conserve resources. For example, duct tape, electrical tape, and zip ties are often utilized to fix, patch, or mend damaged items, thereby extending the time of usage. Ironically, zip ties are also frequently used in packaging. Plastic manufacturers and companies that use final plastic products have a unique opportunity to change the way their products are designed. The 32nd ranked plastic item by use in this study was pull tabs/tear tabs. Companies may consider designing products that do not incorporate pull tabs/tear tabs that are fully removable. Additionally, companies can view this dataset to determine their relative contribution to plastic pollution on LGL beaches. This study, which is the first of its kind to look at company attribution of all plastics sampled (including hard to identify items), should provide companies with basic information to proactively address future regulations and act as good corporate citizens.

Both industry and government have an opportunity to work together to enact change to incorporate plastics into a circular economy. Governments can also take an active approach similar to how the Government of Canada is doing in tandem with the Canadian Council of Ministers of the Environment (CCME) through their Canada-Wide Action Plan on Zero Plastic Waste (CCME 2019, CCME 2020). Citizen science initiatives and/or volunteer cleanups have been used with success to advance knowledge and will be important in the coming years. Moving towards a more circular economy will require humanity to rethink the way we go about doing business, move away from mindless consumerism, and collaborate with all stakeholders. Although plastic pollution is one of a handful of global environmental crises, it is also one that has captured public attention and has rapidly increased focus in the scientific community.

5. Conclusions

Spatially, this study is the largest investigation of plastic debris items on beaches in one lacustrine system. The results provide an important snapshot of the nature of plastic debris on shorelines of the LGL system and will help inform future studies of debris types, abundances, and sources. Pellets, which are industry-related, were the most abundant plastic items, both by item use and by morphology. This investigation represents the first attempt to apply an MST to a lacustrine system. Excluding pellets, the results of the technique indicate that urban and shoreline recreation sources dominated over agricultural sources. The governing bodies of the states and provinces bordering the Great Lakes should consider creating an anthropogenic debris characterization system and photo guide because current photo guides are not region-specific. In addition, an open-source polymer database based on debris item use/type could be developed from this study.

Data availability statement

The data that support the findings of this study are available upon reasonable request from the authors.

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