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A comprehensive investigation of industrial plastic pellets on beaches across the Laurentian Great Lakes and the factors governing their distribution

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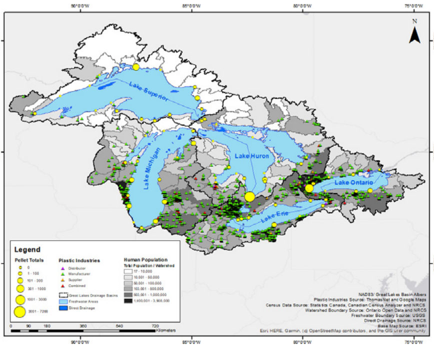
HIGHLIGHTS

- Shorelines of the Great Lakes are littered with industrial plastic pellets.
- 42 of 66 beaches contained pellets, for an average of 19.1 pellets/m^2.
- Abundance increased with number of plastic industries and proximity to river mouths.
- Variety was greatest on a beach in a watershed containing 112 plastic industries.
- Pellets were most abundant on very fine, fine, and medium sand beaches.

GRAPHICAL ABSTRACT



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ABSTRACT

Industrial, pre-consumer pellets are a major type of plastics pollution found on shorelines worldwide. This study investigates the distribution and characteristics of plastic pellets accumulated on beaches of the Laurentian Great Lakes of North America and provides a "snapshot" of pellet distribution in a lake system that accounts for 21% of the world's freshwater reserves. We sampled pellets simultaneously from 10m^2 quadrats on 66 beaches and characterized the 12,595 pellets collected (average of 19.1 pellets/m^2). Forty-two beaches contained pellets and 86% of the pellets were found on three beaches: Rossport (Lake Superior), Baxter (Lake Huron), and Bronte (Lake Ontario). The number of pellets on each beach was compared with factors hypothesized to control their accumulation. In general, positive correlations were found between pellet abundance and watershed population, number of plastic-related industries, and proximity to a river mouth, although for Lake Superior, abundance was related to a train spill that took place over 10 years ago. Beach grain size appears to be related to pellet abundance, with very fine sand, fine sand and medium sand containing the greatest number of pellets. All pellets were visually characterized based on size, color, shape, weathering, and distinguishing traits. The predominant color was white, oblate shapes were most common, and the main distinguishing trait was a dimple. Most pellets showed little evidence of weathering, with the weathered samples mainly from Lakes Erie and Ontario. Lake Ontario pellets were the most varied, with 6/7 shapes, 35/40 colors, and 21/25 distinguishing traits, indicating a wider range of pellet sources compared to the other lakes. Polymer compositions were mainly polyethylene

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(PE) and polypropylene (PP). Our results will lead to increased recognition of regional pellet pollution in the Great Lakes watershed, thereby motivating change during their production, transport and use.

1. Introduction

The global proliferation of plastic debris has been widely documented in both aquatic and terrestrial environments. The first thermosets, synthetic materials that melt when heated and solidify when cooled, were developed in the late 19th and first half of the 20th centuries. Of all thermoplastics produced, polyethylene (PE) and polypropylene (PP) are the most common (Andrady and Neal, 2009). Both PE and PP are derived through polymerization of hydrocarbon monomers in the presence of heat and a catalyst. Once formed, the plastic is cut into small particles by a pelletizer and the pellets (also known as "nurdles") are then transported to industries to be melted and extruded or molded into common plastic products. Unfortunately, numerous pellets are lost during production, transport and storage (Karlsson et al., 2018). Spilled or discarded pellets make their way into drainage systems, circulate through the surface waters of rivers, lakes, seas, and oceans, and are eventually deposited along shorelines. Documenting, quantifying, and characterizing pellets provides information that can potentially be used to change policy or industry behavior in favour of the thoughtful handling of these materials.

Plastic pellets have been reported from beaches across the globe for over 45 years (e.g. Carpenter and Smith, 1972; Gregory, 1977; Zbyszewski et al., 2014; Fernandino et al., 2015; Karlsson et al., 2018). Identification of pellets thousands of kilometres from the nearest pellet production or use facility (e.g. Corcoran et al., 2014) indicates that pellet buoyancy results in long-range transport. If a pellet is encrusted with minute biota (e.g. Carpenter et al., 1972), the potential for introduction of invasive species is increased. In addition, pellets have been shown to adsorb and release persistent organic pollutants (POPs) such as polychlorinated biphenyls (PCBs), dichloro-diphenyltrichloroethane (DDT), hexachlorocyclohexanes (HCHs), and polycyclic aromatic hydrocarbons (PAHs) (e.g. Mato et al., 2001; Rios et al., 2007; Ogata et al., 2009; Heskett et al., 2012; Koelmans et al., 2016). International Pellet Watch, initiated in 2005 by Hideshige Takada, shows that pellets have been found in over 50 countries, proving that pellet pollution is truly a global issue (http://www.pelletwatch.org/index.html). Similarly, The Great Nurdle Hunt, organized by UK charity FIDRA, aggregates citizen-collected data on pellets. As of the time of writing, over 3000 "Nurdle Hunts" have taken place, with at least one on every continent (https://www.nurdlehunt.org.uk/). Once in the environment, pellets can be ingested by aquatic wildlife such as ocean fish, squid, and seabirds (e.g. Braid et al., 2012; Van Franeker and Lavender Law, 2015; Miranda and de Carvalho-Souza, 2016). Although few studies focus on the physiological effects of pellet ingestion, numerous investigations show that microplastic (<5 mm plastic particles) ingestion can affect feeding behavior, reproduction, and growth of aquatic organisms (Chae and An, 2017).

Pollution of the Laurentian Great Lakes with plastic debris was first documented in 2011 in a study examining plastic particle distribution and degradation along the shoreline of Lake Huron (Zbyszewski and Corcoran, 2011). Subsequent investigations focused on the presence of micro- and macroplastic debris in surface waters of the Great Lakes and its tributaries (Eriksen et al., 2013; Baldwin et al., 2016; Lenaker et al., 2019), shorelines (Hoellein et al., 2014; Zbyszewski et al., 2014; Driedger et al., 2015), and benthic lake and river sediment (Corcoran et al., 2015; Ballent et al., 2016; Dean et al., 2018; Corcoran et al., 2020). Plastic pellets were components of the debris load in surface water and beach studies, with very minor amounts reported from benthic zones, as a result of their low density and high surface area. Zbyszewski and Corcoran (2011) and Corcoran et al. (2015) showed that pellets are major components of shorelines in the Great Lakes, with as many as 33 pellets/m^2 (Baxter Beach, Lake Huron) and 21 pellets/m^2 (Humber Bay), respectively.

The overall objective of this study was to record a "snapshot" of the distribution of plastic pellets along shorelines of the five Laurentian Great Lakes of North America; a lake system that may be considered a smaller-scale, freshwater proxy for the world's oceans. The Great Lakes have approximately 16,500 km of shoreline and hold 21% of the world's surface freshwater reserves. An essential freshwater resource, they are simultaneously the location of intense agricultural and industrial activity, including significant plastics manufacturing. Understanding how pellets enter and are transported and distributed through the lake system is essential in the reduction of pellet pollution.

In this study, we aim to determine the relationships between the quantity, types and distribution of pellets in each lake to ascertain: 1) the influence of plastic industries and watershed population, 2) the influence of spatial location with respect to proximity of river mouths and major highways, and location within or outside bays, and 3) the influence of depositional processes as related to beach grain size. By statistically examining these relationships, we convey the extent of pellet pollution and its possible sources. Through this work, we strive to raise awareness of the plastic pellet issue and to inspire policy development in terms of proper handling of pellets throughout the supply chain.

2. Materials and methods

2.1. Regional setting

The Laurentian Great Lakes are located in central North America, straddling the boundary between Canada and the United States (Fig. 1a). Covering an area of 244,000 km^2, the Great Lakes system accounts for approximately 21% of the world's surface fresh water and is home to >30 million people (EPA, 2019). It is composed of the five main lakes Superior, Michigan, Huron, Erie, and Ontario. Each lake has its own sub-basins (https://www.ngdc.noaa.gov/mgg/image/images/greatlakesbasin.pdf) and unique water current patterns (Beletsky et al., 1999; EPA, 2019).

2.2. Field sampling

A total of sixty-six beaches were surveyed for plastic pellets between October 7th and 21st in 2018. The number of sampling locations around each lake varied with lake size. Eighteen beaches were sampled on Lake Superior, 15 on Lake Michigan, 14 on Lake Huron, 10 on Lake Erie, and 9 on Lake Ontario (Fig. 1b). Each lake was surveyed by two people, except Lake Superior where two people surveyed 15 beaches and three other people surveyed the remaining 3 beaches. In total, thirteen samplers were involved in the fieldwork, all were given detailed instructions prior to departure, and all were familiar with the appearance of plastic pellets. At each beach, samplers were instructed to i) take photographs of the beach, strandline, and any plastic debris identified, ii) measure the grain size of the natural sediment on the beach; if this proved difficult, samplers collected sediment and brought it back to the lab for measurement, iii) collect all plastic debris along the strandline. The latter step was conducted by stretching a 10 m measuring tape perpendicular to the strandline and collecting pellets and other plastic debris within 1 m of the tape (Fig. 2a). Plastics from only the top 5 cm of the beach

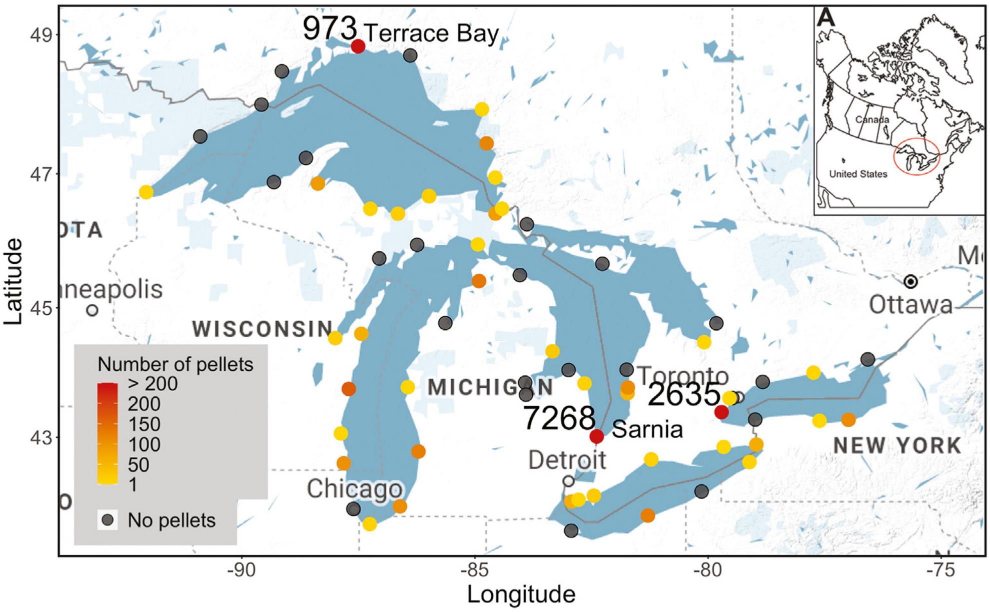

Fig. 1. Spatial and pellet abundance details. A) Location of the Great Lakes system in North America. B) Sixty-six sampling locations along Great Lakes beaches, with relative abundances of pellets indicated.

surface were collected using bare hands if the sediment was wet. Metal sieves were used if the sediment was dry and fine enough to pass through the sieve openings, thereby leaving pellets and plastic debris >2.5 × 3 mm remaining in the sieve. Pellets and other plastic debris were stored in paper bags, and each bag was labelled according to lake and beach name. The paper bags were brought back to the University of Western Ontario for pellet characterization.

2.3. Pellet characterization

A total of 508 pellets were randomly selected for chemical analysis using Fourier transform infrared spectroscopy (FTIR) at Surface Science Western, University of Western Ontario. Pellet surfaces were analyzed using platinum attenuated total reflectance (Pt-ATR) equipped with a diamond crystal in the main box (Bruker Tensor II spectrometer). This experimental setup allowed for analysis of an area of approximately 2 mm × 2 mm to a depth of 0.6–5 μm. The spectra were collected from 4000 to 400 cm^{-1} with a resolution of 4 cm^{-1}. Some of the pellets were also analyzed in cross-section. A total of 101, 99, 105, 100, and 103 pellets were analyzed from Lakes Superior, Erie, Huron, Michigan, and Ontario, respectively.

Over the course of six months, the pellets were separated from other plastic items and each pellet was characterized according to size, shape, diagnostic trait, visible weathering, and color (Fig. 2b–f). Information concerning each pellet was entered into a database for statistical analysis. The size of each pellet was measured with a ruler by finding the plane of maximum projection, then measuring the long (l) and intermediate (i) axes and multiplying them together. The short axis (thickness) of each pellet was not measured due to time constraints. The shape of each pellet was categorized as either circular (l = i and pellet has no angular edges), square (l = i and pellet has four angular edges), rectangular (l > i and has four angular edges), oblate (l > i and pellet has no angular edges), cylindrical (l > i; pellet is a circular prism), cylindrical irregular (l > i and pellet is not similar on all edges of the circular prism), or irregular (l > i and pellet has no well-defined shape). Diagnostic traits (e.g. rims, lines, dimples) were visually determined and divided into 23 categories. Color was visually determined and divided into 38 categories. Because six people were involved in pellet characterization, the categories for shape and diagnostic traits were cross-checked among examiners. Color was not cross-checked and therefore represents the greatest source of characterization error given that different individuals see color in different ways.

2.4. Spatial analysis

The number of plastic suppliers, distributors and users were determined from ThomasNet Supplier Discovery and Google Maps using the search terms "plastics" and "polymers" within a 100–300 km radius of each lake depending on the size of each lake basin. Coordinates were assigned to each facility for mapping purposes.

Mapping and spatial analysis for Fig. 7 was performed using ArcMap 10.5. Spatial analysis methods for determining population density by watershed were modified from Ballent (2016). Dissemination block area boundary files were provided from Statistics Canada (2020) and were joined to 2016 census data collected from the Canadian Census Analyzer (2014). Dissemination block data were used for Ontario. The profile variables selected from the population and dwelling counts for 2016 included previously normalized data, population density per square kilometer. United States Census Bureau block-level data were used from the 2010 US Census for each of the eight states in the Great Lakes Basin. Watersheds were mapped at the USGS HUC-8 and Ontario Tertiary levels, which are equivalent (Neff et al., 2005). Census data were clipped to watersheds and were converted to raster using 100 m cell size. Zonal statistics were used to determine the population per watershed, and watersheds were mapped using approximate quantiles. "Direct Drainage" areas, which indicate islands and land adjacent to the Great Lakes that were not incorporated into the watershed shapefiles were not considered in population analysis.

2.5. Statistical analysis

In order to determine whether pellet distribution was related to river mouth proximity, any beach within 5 km of a river was given a "true" value, with beaches >5 km from a river mouth assigned a "false" value for statistical analysis. Similarly, any beach sampled that was located in a bay was given a "true" value, as opposed to beaches outside bays, which were given "false" values. Proximity to major highways was determined using the Trans-Canada and 400-series (Canada)

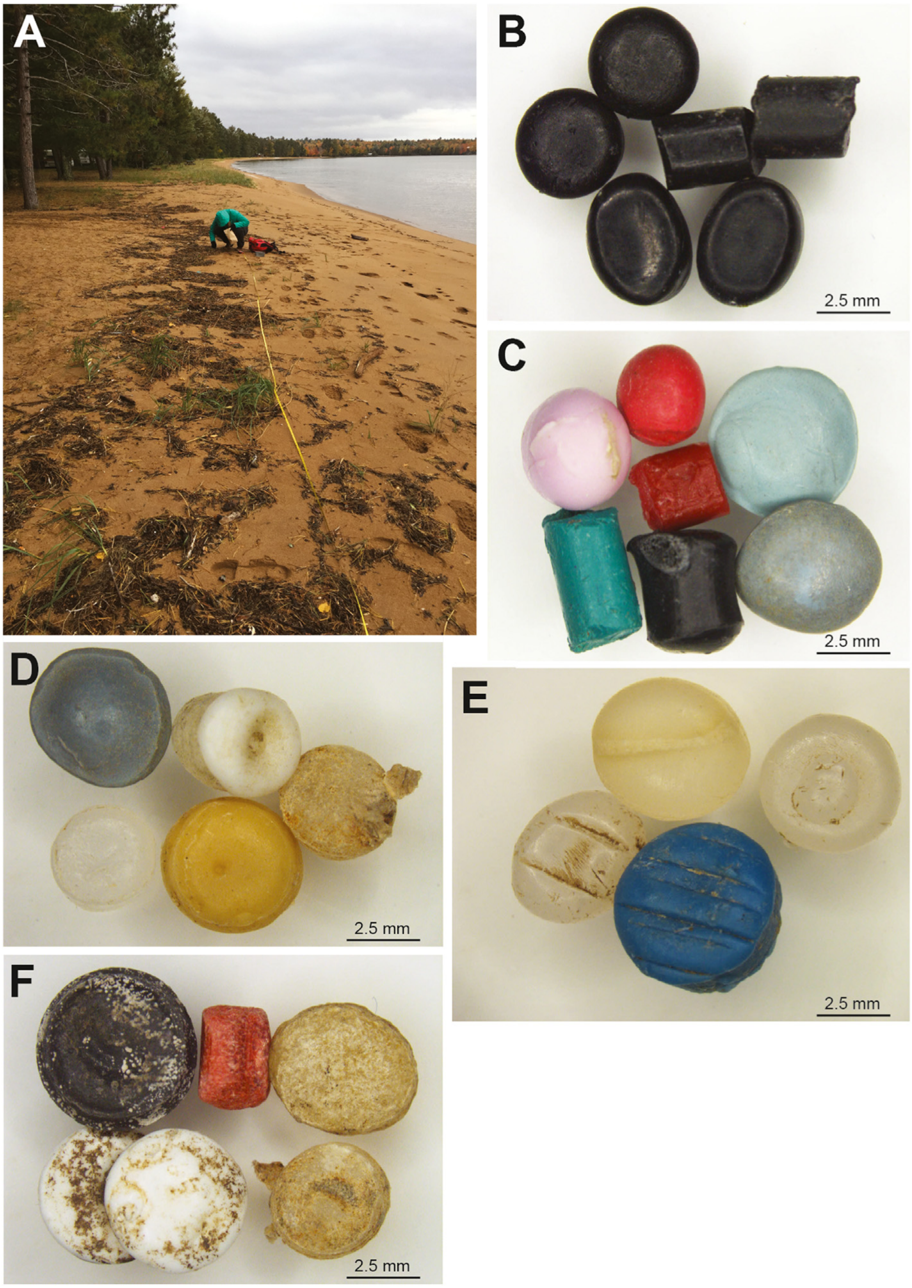

Fig. 2. Sampling site and pellet characteristics. A) Sampling pellets from a 1 × 10 m quadrat along the strandline of a Lake Superior beach. B) Examples of three distinct types of pellets. Clockwise from top left: black, oblate; black, cylindrical; black, oblate, rimmed. Note that the three types are of different sizes. C) Examples of the three most common shapes, clockwise from upper left: two circular pellets (rifle beads), two oblate, and three cylindrical. D) and E) Distinguishing traits of pellets sampled. Clockwise from top left in D: rimmed and nodule; dimple and dirty; rough and broken; rimmed and hole; rimmed. Clockwise from top left in E: groove; dimple and nodule; lines and dirty; lines and dirty. F) Weathered pellets with distinguishing traits such as dirty, rough and broken. The two lower left pellets are coalesced (joined together).

and Interstates (U.S.), and distances ranged from 1 to 270 km. Figs. 2 to 6 were created in R version 3.6.0., using the package ggplot2 version 3.2.0 (Wickham, 2016). Fig. 1 was created using the package ggmap, version 3.0.0 (Kahle and Wickham, 2013).

3. Results

The total number of pellets collected from around the Great Lakes was 12,595, for an overall average of 19.1 pellets/m^2. A total of twenty-four beach sampling quadrats contained no pellets (Fig. 1b). The total number of pellets found on Lake Superior beaches was 1341, for an average of 7.5 pellets/m^2; 67% of the beaches contained pellets. On Lake Michigan beaches, a total of 728 pellets were collected, for an average of 4.9 pellets/m^2, with 73% of the beaches containing pellets. A total of 7471 pellets were found on Lake Huron beaches with an average of 57.5 pellets/m^2, but only 43% of the beaches contained pellets. Lake Erie beaches contained 302 pellets, for an average of 3.0 pellets/m^2; 80% of the beaches contained pellets. Finally, Lake Ontario beaches contained 2753 pellets with an average of 30.6 pellets/m^2, but only 56% of the beaches contained pellets. The overall pellet counts from Lakes Huron, Ontario and Superior beaches are skewed as a result of one beach on each lake accounting for 73% (Rossport – Lake Superior), 96% (Bronte – Lake Ontario), and 97% (Baxter – Lake Huron) of the pellets (Fig. 3). Removal of these outliers indicates that the beaches sampled on Lake Michigan contained the greatest number of pellets, followed by beaches on Lakes Erie, Superior, Huron, and then Ontario (Fig. S1).

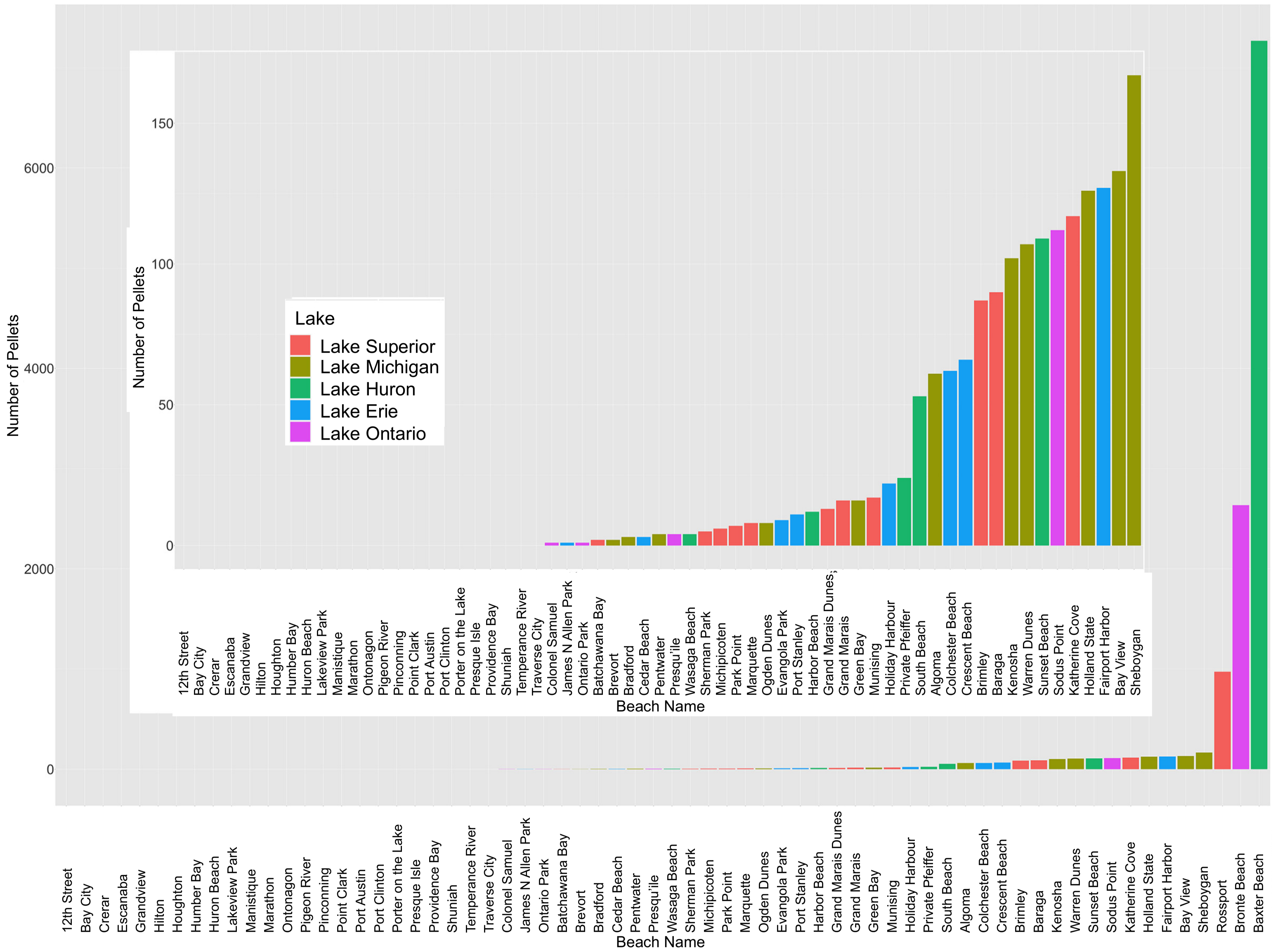

Fig. 3. Pellet abundance per sampling quadrat on each beach. Lower graph includes all beaches and inset includes only those beaches with 200 pellets or less. Twenty-four beaches contained no pellets. Bars are color-coded according to lake. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

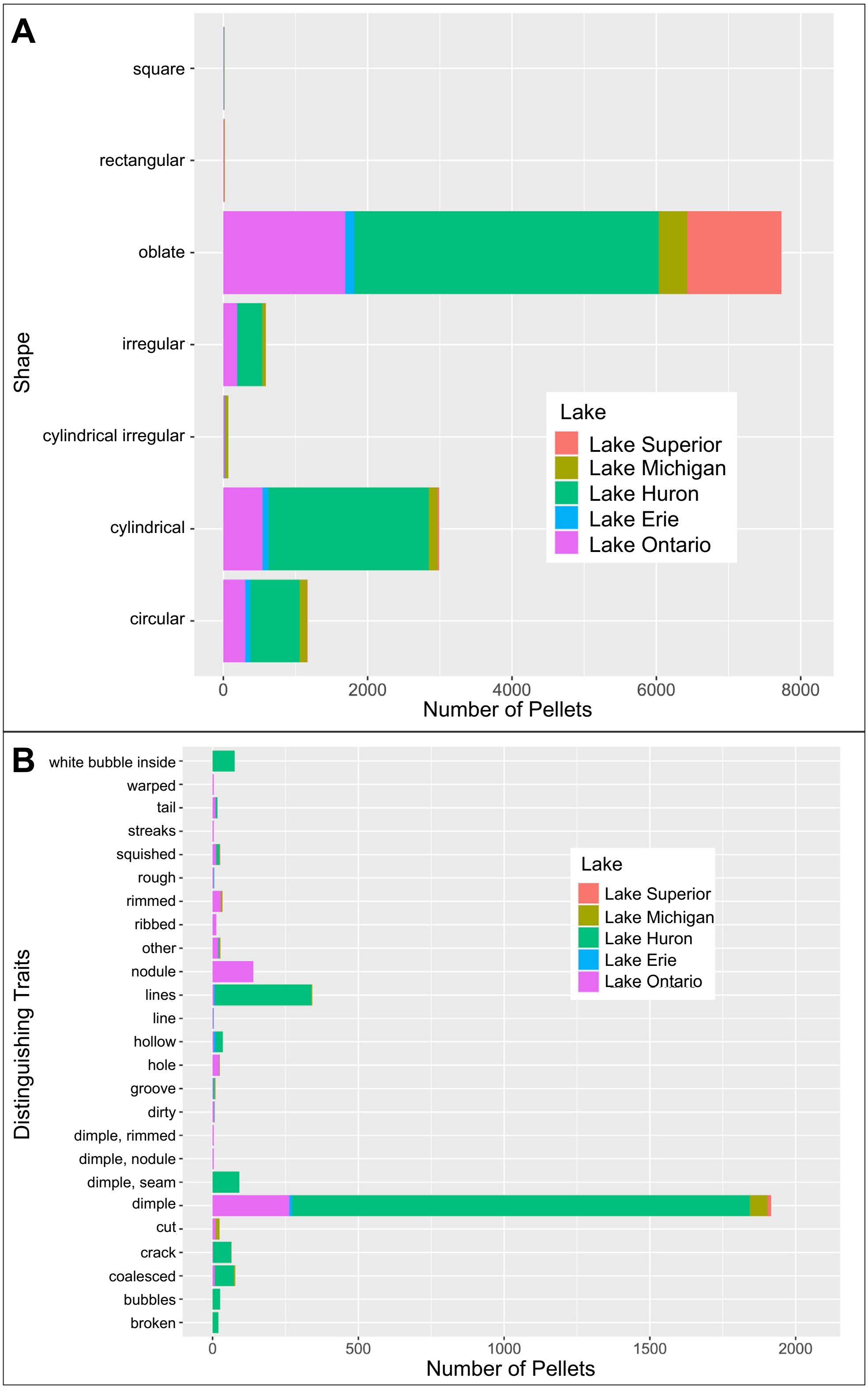

Fig. 4. Bar graphs illustrating the shapes and distinguishing traits of pellets found on the beaches of each lake. A) The most common shape found on all lakes was oblate, followed by cylindrical, then circular. B) The most common distinguishing trait found on all lakes was a dimple, although 74% of the pellets contained no distinguishing trait at all.

3.1. Pellet compositions

The FTIR results show that 85.8% of the pellets are composed of polyethylene (PE), 8.5% are polypropylene (PP), 1.6% are thermoplastic olefin (TPO), and 1.0% are ethylene vinyl acetate (EVA) (Table S1). Polystyrene (PS), butadiene-styrene, acrylonitrile butadiene styrene (ABS), nylon, PP/PE blend, and PE/Polyamide blend make up <2.2% of the pellets. Two pellets were unknown polymers and 2 were minerals. Lakes

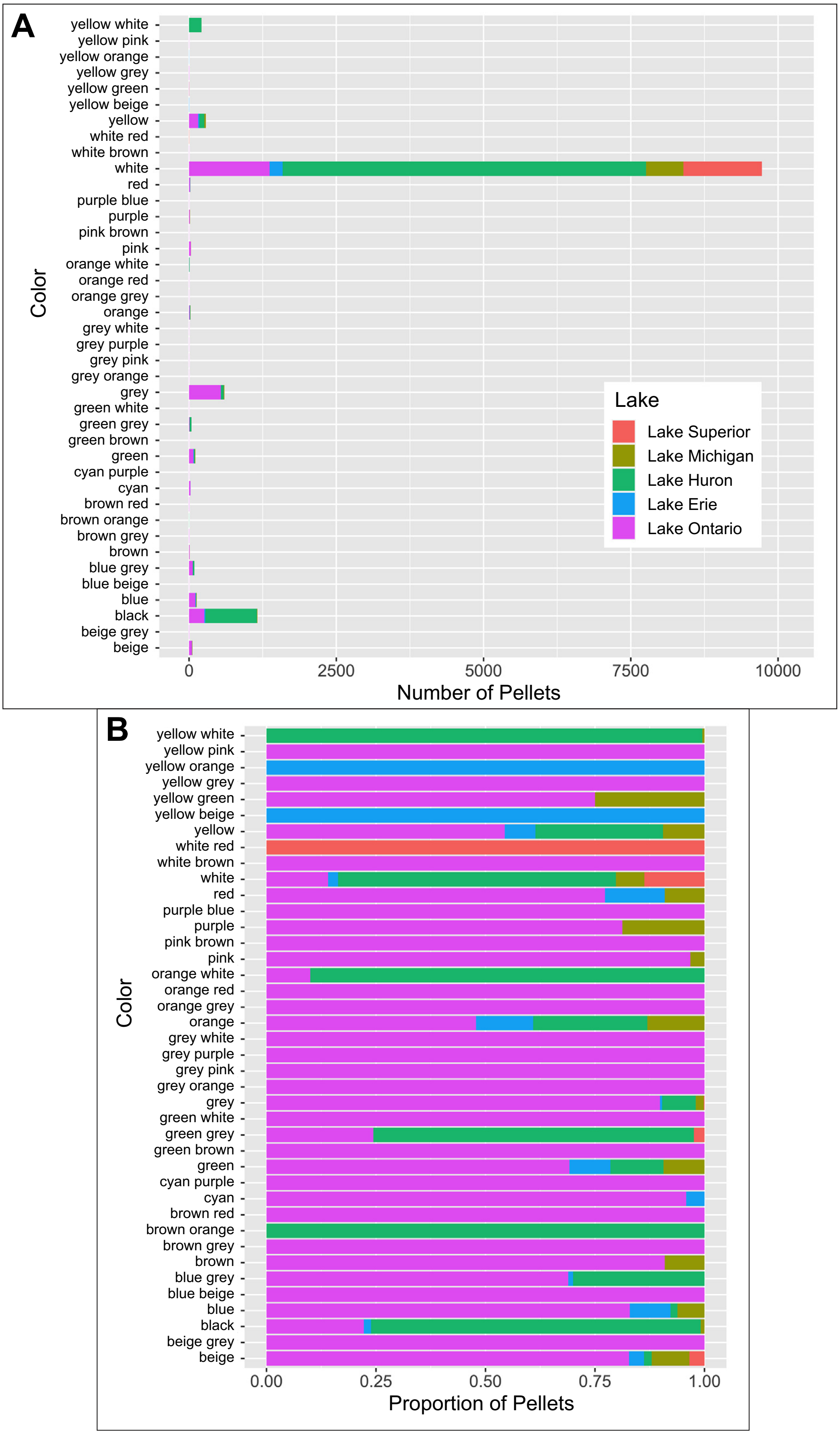

Fig. 5. Graphs illustrating the distribution of pellet colors for each lake. A) The main pellet color found on beaches of all lakes was white. The second most common color was black, although black pellets were not found on Lake Superior beaches. B) Lake Ontario beaches contained the greatest variety of pellet colors with 35/40 colors, whereas the lake with the poorest variety of pellet colors was Superior with only 5/40 colors. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

Erie and Ontario contained the greatest variety of polymer compositions, whereas the pellets analyzed from Lake Superior were composed of only PE and PP.

3.2. Pellet characteristics

Pellet types on each beach were assigned to pellets of the same size, and with the same color, shape, diagnostic feature, and visible surface weathering (Table S2; Fig. 2b). Pellet types on different beaches within the same lake were compared only for Lake Superior. The sheer volume of different pellets on beaches of the other four lakes prohibited type comparisons for this part of the study. Pellet sizes varied between 2 and 42 mm^2, with a median of 12 mm^2 and a mean of 13.7 mm^2 overall. Mean sizes for each lake were 17.9 (Superior), 14.5 (Michigan), 13.4 (Huron), 14.5 (Erie) and 11.9 (Ontario). A one-way analysis of variance (ANOVA) was conducted to compare the lake effect on individual pellet size, that is, to test that there is a significant difference in mean pellet size between lakes. This was significant at the $p < 0.01$ level. The predominant pellet shape overall was oblate (Fig. 4a). Cylindrical, oblate, and circular pellets were the most common shapes and were identified in samples from beaches on all five Great Lakes (Fig. 2c). A total of 9329 pellets had no diagnostic traits. Of the other 3266 pellets, dimples were by far the most common (Lakes Ontario, Huron, Michigan and Superior), followed by lines (Lakes Ontario and Huron) and nodules (Lake Ontario) (Figs. 2d, e, 4b). The sampled quadrats on the three beaches with the greatest number of pellets show that Baxter Beach (Huron) and Bronte Beach (Ontario) contained the largest variety of pellets with respect to distinguishing traits, whereas Rossport beach (Superior) mainly contained pellets with no distinguishing traits.

Pellet color, although a subjective category, shows that the majority of the pellets on beaches in the Great Lakes Basin were white, followed by black and grey (Fig. 5a). Lake Ontario, and specifically Bronte Beach, contained the greatest variety of pellet colors, with thirty-five of the forty color types (Fig. 5b). Pellets from Lake Huron and Lake Michigan contained thirteen of forty colors, twelve of forty colors were represented in Lake Erie pellets, and Lake Superior pellets were of only four color types (Fig. 5b). A total of 96.4% of the pellets on Lake Superior beaches were white, oblate, 18 mm^2 and with no diagnostic traits (Type 3405 in Table S2). These pellets are the result of a train derailment that occurred in 2008 near Terrace Bay, Ontario, which spilled pellets into Lake Superior (https://www.sootoday.com/local-news/pellets-mystery-solved-182329).We also documented the relative degree of weathering of each of 436 PE pellets analyzed by FTIR. Peaks in the spectra between 1710 and 1775 cm^{-1} are increased absorption peaks, which are indicative of oxidized material. Oxidation was classified as low if there was little to no evidence of absorption peaks at about 1715 cm^{-1} relative to the characteristic PE peak height at around 1471 cm^{-1} (Fig. S2). The results in Table S1 indicate that 13.7% of the pellets analyzed were weathered, with the greatest percentage of weathered pellets in Lakes Ontario, Michigan and Erie. We also attempted to visually note the number of pellets that appeared extensively weathered, as determined through color change (fading or yellowing), increase in surface roughness (e.g. pits and microfractures), embrittlement, and abundance of external particles adhered to surfaces (e.g. Zbyszewski and Corcoran, 2011; Brandon et al., 2016; Cai et al., 2018) (Fig. 2f). Of the 15,595 pellets examined, 2.4% showed extensive evidence of weathering (Table S3). By individual lake, the percentages of pellets that were visually identified as weathered were <0.01% (Superior), <0.01% (Michigan), 2% (Huron), 10.6% (Erie), and 0.04% (Ontario). A comparison of the results indicates that visual characterization of weathering is not as precise as chemical identification. This may be due to challenges in recognizing discoloration in pellets with dark colors (e.g. black, blue).

The number of pellets in each beach quadrat was related to grain size of the beach sediment. All samplers used the grain size classification chart of Wentworth (1922), except for the very fine silt to coarse silt categories, which were grouped into "silt" because only two beaches fell into the category (Fig. 6). The "mixed" category was added to indicate beaches with polymodal grain sizes (containing more than two), and the "organics" category was added for the Pigeon River beach on Lake Superior, which was completely covered in logs, sticks and leaves. The results in Fig. 6 indicate that the majority of pellets were identified on beaches composed of very fine sand, fine sand, medium sand and mixed grain populations. These grain size categories also showed the

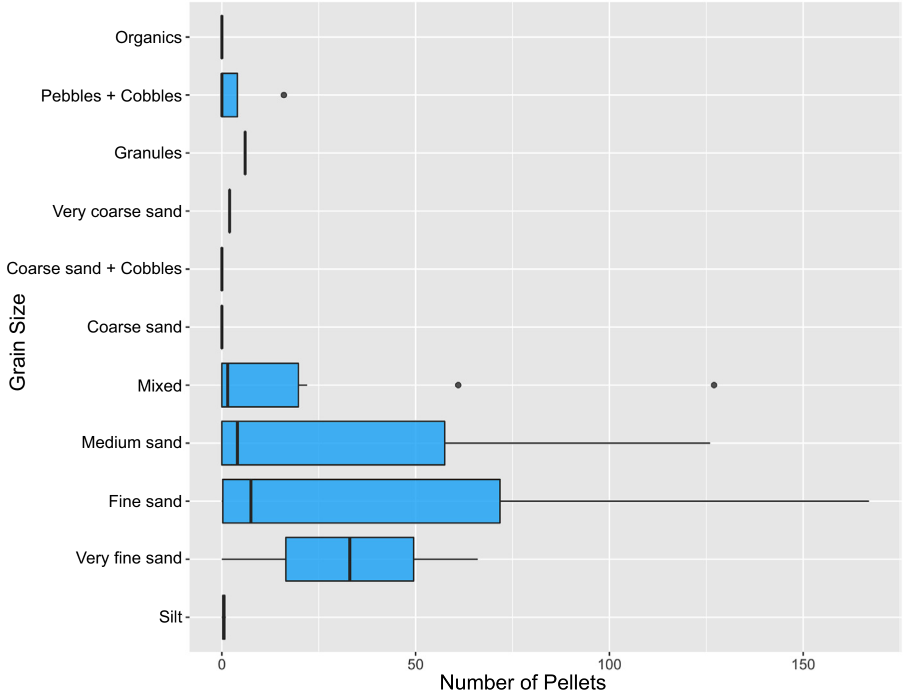

Fig. 6. Boxplots displaying the relationship between pellet abundance and grain size on beaches. The means for very fine sand, fine sand, and medium sand were greater than other grain size grades, which suggests that pellets preferentially accumulate on sandy beaches compared to silty, granular, pebbly and cobble beaches.

greatest ranges in number of pellets, especially fine sand and medium sand. Therefore, although pellets appear to accumulate preferentially on sandy beaches, some sandy beaches contained no pellets.

3.3. Population and plastic industry

High pellet abundances across the Great Lakes were correlated with large population numbers (Fig. 7). We recognize that the general population does not have access to pre-production plastic pellets, however, distinguishing between industry proximity and watershed population is challenging because most pellet manufacturers, suppliers and distributors are located in high population areas (Fig. 7). An exception is Lake Superior, which has the lowest overall watershed populations of all the Great Lakes, with only four of the twenty-five watersheds containing >50,000 people. Removal of the high pellet count at Rossport beach, which is a direct result of the train derailment, leaves a total of 368 pellets (2.0 pellets/m^2). Furthermore, removal of all pellets from Lake Superior beaches that originated from the spill (Type 3405 in Table S2) leaves only 48 pellets (0.3 pellets/m^2).

Lake Michigan pellet counts appear to be greater with an increased number of plastic industries, particularly in the southeastern and southwestern portions of the lake (Fig. 7). The three northernmost sampling locations and two southernmost sampling locations contained a mean of 2 pellets (0.2 pellets/m^2) and these were the regions with the lowest number of industries.

Beaches along the northern half of Lake Huron contained no pellets and twenty-one of those twenty-four watersheds had total populations <100,000 (Fig. 7). In contrast, the southern half of Lake Huron (including the southern coastline of Georgian Bay) is composed of twenty watersheds, nine of which have a population >100,000; pellet abundances are substantially greater in the southern (67.1 pellets/m^2) compared to the northern (0 pellets/m^2) parts of the lake (Fig. 7). In particular, the location with the highest pellet counts across all five Great Lakes is in the Sarnia region at the southern end of the lake. Although most of the plastic industries in this region are found along the St. Clair River, which flows south into Lake St. Clair, several creeks draining the industrial sector flow into southern Lake Huron. We have visually identified numerous pellets within and along at least one of these creeks. Although pellets are widely distributed across Lake Erie, there appears to be no relationship between pellet abundance and the location and density of plastic industries (Fig. 7). Notwithstanding, the great variety in pellet compositions and colors for Lake Erie samples compared to other lakes supports the hypothesis that multiple industrial sources are involved.

Of the twenty-two watersheds surrounding Lake Ontario, sixteen have watershed populations >100,000 and two watersheds contain

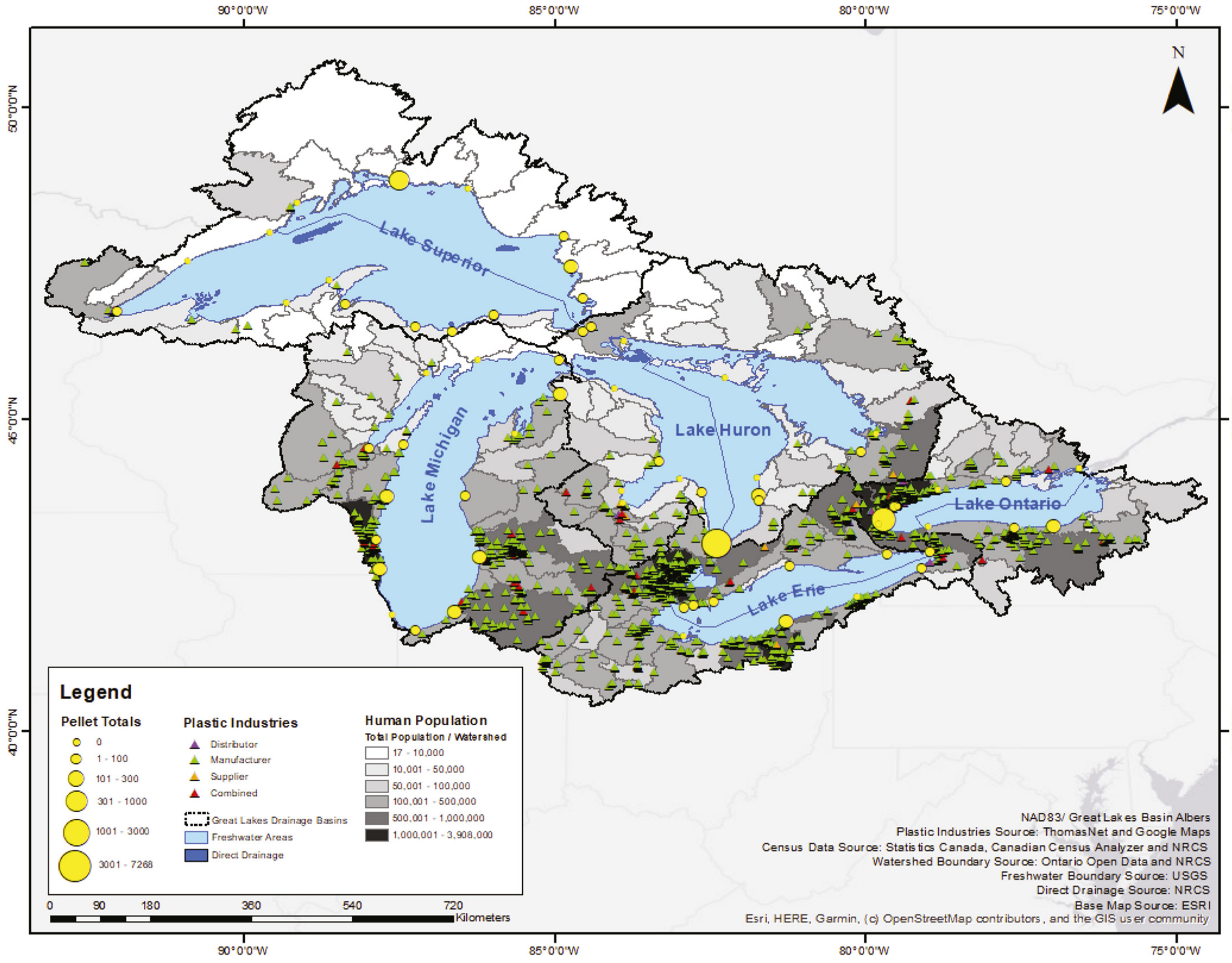

Fig. 7. GIS map displaying pellet totals at each beach (yellow dots), and locations of plastic distributors, manufacturers, and suppliers throughout the entire Great Lakes basin. Watersheds are divided by thick black lines and total population per watershed is indicated in various shades of grey. Note how the pellet totals are greatest in Lake Huron and Lake Ontario. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

>1,000,000 people. The high pellet count of 26.4 pellets/m^2 at Bronte Beach falls within one of these watersheds (Fig. 7). Bronte Beach is located within an approximately 160 km long corridor spanning two watersheds (Credit-16 mile, Humber-Don) that contain 415 identified plastic industries (Fig. 7).

3.4. Location relative to bays, river mouths and major highways

The number of pellets at each sampling quadrat was compared with proximity to rivers, location within bays, and proximity to major highways. Thirty-one of the sixty-six beaches sampled are located within 5 km of a river, and these contained a mean number of 373 pellets (Table S2). Notably, the three beaches with the highest pellet counts (Baxter, Bronte, and Rossport) are all located within 5 km of a river. For sampling locations with <200 pellets, the mean was 36. The thirty-five beaches located >5 km from a river had a mean of 20 pellets. A two-sample, one-tailed *t*-test on the data containing all locations produces a p-value of 0.0747, indicating that there is a significantly greater amount of pellets on beaches within 5 km of a river mouth.

Of all of the beaches sampled, seventeen were located within a bay and the mean number of pellets for these beaches was 21 (Table S2). The 49 beaches located outside bays produced a mean pellet abundance of 240. For locations with <200 pellets, the means for pellet abundance within bays (21) and outside bays (29) were not statistically significant. A simple two-sample *t*-test on the data containing all locations produces a p-value of 0.4214, indicating that there is no significant difference in mean number of pellets on beaches within and outside bays. The number of pellets at each sampling location was also plotted against distance to a major highway. The data points display no correlation between the two variables (Fig. S3).

4. Discussion

The main objective of this study was to determine the factors controlling the abundance and distribution of plastic pellets along beaches throughout the Great Lakes watershed. The results of this study highlight some of the major controls on pellet distribution during late Autumn, 2018.

4.1. Influence of plastic industries and watershed population

Not surprisingly, the data show that in the basin as a whole, high pellet abundance can be related to high watershed population and greater numbers of plastic industries, as the two factors are positively correlated. The two beaches with the greatest number of pellets (Bronte-Lake Ontario; Baxter-Lake Huron) are spatially associated with a significant number of plastic industries, indicating their influence on pellet abundance. Lake Superior contained the beach with the third highest number of pellets (Rossport), but this abundance is neither due to watershed population nor industry, as both factors are relatively negligible for this lake. Instead, the pellets on Lake Superior beaches are a result of a spill from a train derailment, indicating that pellets are not only lost at the source or destination, but also in transit.

Interestingly, Lake Erie beaches contained the lowest number of pellets, but displayed a wide pellet distribution across its beaches. The plastic debris distribution models of Hoffman and Hittinger (2017) and Cable et al. (2017) suggest that most plastic pollution in Lake Erie would accumulate in the water along the southern shoreline. However, we did not see a correlation between our beached pellet data and the surface water accumulation models. The lack of a statistical relationship between pellet abundance, watershed population, and number of plastic industries for Lake Erie may be a function of its short hydraulic residence (retention) time. According to the EPA (2019), Lake Erie has a residence time of approximately 2.6 years, whereas in contrast, the residence time for Lake Superior is 191 years.

There does appear to be a correlation between our pellet data and Hoffman and Hittinger's (2017) modelled movement of plastic debris for Lake Michigan. Both data sets show a greater accumulation of plastic debris in the southern portion of the lake. Unlike Lake Erie, Lake Michigan has a high hydraulic residence time of 99 years (EPA, 2019), and therefore, residence time does not appear to control the low number of pellets on Lake Michigan beaches overall. The low pellet numbers, especially for the high population watershed in which 12th Street beach was sampled (with 0 pellets found), and wide distribution of pellets across beaches of Lake Michigan may be a result of: i) the drainage of Chicago's land into the Mississippi River system instead of Lake Michigan through a complex natural and artificial hydrological network known as the Chicago Area Waterway System (CAWS) (Duncker and Johnson, 2016), ii) a low overall release/spill of pellets, or iii) the pellets being retained in surface water and not deposited on the beaches. Comparing the movement of surface water currents within each lake is beyond the scope of this project, but retention of pellets in surface waters may be a function of the complexity of the current patterns (e.g. number of gyres, current strength and speed) in different seasons of the year.

Interestingly, there were statistically significant differences in pellet sizes between lakes. The largest pellets were identified on Lake Superior and the smallest were found on Lake Ontario. We considered that this could be related to a decrease in size with extended weathering as a pellet travels through the Great Lakes system, but the results show that Lake Erie beaches contained the most highly weathered pellets and their average size of 14.5 mm^2 is equivalent and greater than the size averages for Lake Michigan and Lake Huron, respectively. Instead, the average size differences, much like color and shape, probably reflect the producers' preferences, and have little to do with environmental effects.

We investigated the characteristics of each pellet in order to identify different pellet types on each beach, which would help find clues concerning source. The number of different pellet types on the most pellet-rich beach of each lake included: 6/973 (Rossport-Superior), 426/7268 (Baxter-Huron), 79/167 (Sheboygan-Michigan), 58/127 (Fairport Harbor-Erie), and 1291/2635 (Bronte-Ontario). These results suggest that there is an order of magnitude fewer sources of pellets to Lake Superior than to Lake Huron, and that Lake Huron contains an order of magnitude fewer pellet sources than Lakes Ontario, Erie and Michigan. Lake Ontario, and specifically Bronte Beach, contained the greatest number of pellet types per total pellets. If plastic industries are purposefully or inadvertently allowing pellets to spill into or near tributaries in the Credit-16 mile and Humber-Don watersheds (which flow into Lake Ontario), this could explain the wide range of pellet types washing up on Bronte Beach. While large, instantaneous spills have required cleanups (e.g. Terrace Bay, ON and Pocono Creek, PA), industries responsible for long-term, regular releases of pellets have not had such requirements. However, a recent federal decision sets a new precedent. For decades, the Formosa Plastics Facility in Point Comfort, TX discharged plastic pellets and powders into Lavaca Bay despite their permit allowing only trace amounts of floating debris (Waterkeeper v. Formosa, June, 2019a). In the Final Consent Decree, the US District Court required, among other remedial measures, that past discharges of plastics had to be cleaned up, future discharges abated, and that Formosa put $50 million USD towards Environmental Mitigation Projects (Waterkeeper v. Formosa, Nov., 2019b).

4.2. Influence of spatial location (river mouths, bays, highways)

Plastic pellets are normally transported from manufacturer to processer by train, truck or ship. The pellets spilled from a rail car into Lake Superior in 2008 continue to be deposited along the lake's shoreline, but the long hydraulic residence time of the lake suggests that the pellets may remain suspended in the water column for a century or more. Plastic pellets spilled during transport via trucks (e.g. https://

www.wfmz.com/news/tractor-trailer-spills-plastic-pellets-in-pocono-creek/article_970503dd-90a8-578c-8a51-bfe2f0214693.html), could also make their way into drainage systems, creeks and other watercourses, and eventually become deposited into large bodies of water. We attempted to discern if a positive relationship exists between pellet abundance on Great Lakes beaches and proximity to major highways. The data indicate that no correlation could be found, which may be a function of distance between source (truck) and sink (lake).

We also hypothesized that once pellets enter a lake, they would preferentially accumulate in bays or other protected inlets. No statistical support for this hypothesis was evident in our data, which suggests that the low density of pellets causes them to recirculate throughout each lake, become beached during high onshore wind and wave events, and then become transported back into the lake during high rain, lake water or offshore wind events.

One factor that positively affects the abundance of pellets on beaches in our study is proximity to river mouths. A statistically greater number of pellets were found on beaches located within 5 km of a river mouth. This supports models showing that rivers are one of the main pathways transporting plastic debris from land to larger bodies of water (Lebreton et al., 2017; Schmidt et al., 2017). Studies have also shown that microplastics, including pellets, are abundant in tributaries flowing into the Great Lakes watershed (Corcoran et al., 2015; Baldwin et al., 2016; McCormick et al., 2016; Lenaker et al., 2019), and therefore, tributaries could represent major pathways for input of pellets to the lakes.

4.3. Influence of depositional processes as related to beach grain size

The preferential deposition and/or retention of pellets on beaches with sand-size sediment is likely a function of wave energy in the depositional environment. Beaches composed of grains larger than medium sand require relatively high energy, and cobble beaches, such as Marathon (Superior) and Porter on the Lake (Ontario) contained no pellets. Although high wave energy would transport pellets onto the beach, that same energy redistributes the pellets back into the water. Beaches composed of silt or organics (e.g. Port Clinton-Huron; Pigeon River-Superior) also contained no pellets, but this is instead due to the wave energy being too low for sufficient landward transport. Although it is expected that grooming affects the distribution of pellets across a beach, only 6 of the 66 beaches had been groomed prior to sampling (Table S3), and therefore, we could not statistically test this hypothesis.

5. Conclusions and future work

The present basin-wide investigation of plastic pellets represents the largest simultaneous sampling campaign for pellets in the world. The study emphasizes the major controls on pellet pollution in the Laurentian Great Lakes. Factors that correlate positively with elevated pellet totals are high watershed population numbers, high density of plastic industries, <5 km distance from a river mouth, past evidence of pellet spills, and beach grain sizes ranging between very fine and medium sand. The low number of pellet types and compositions on Lake Superior beaches and the high number of types and compositions on Lake Ontario beaches support the influence of plastic manufacturers and processers on pellet pollution. Ideally, pellet types across all five lakes should be compared in order to determine whether pellets are being transported throughout the Great Lakes system. This is the next logical, albeit lengthy extension of the present study. We are planning to survey each sampling site and collect pellets in October 2023, to compare pellet abundances from Year 1 and Year 5. The results will hopefully highlight whether awareness and mitigation of pellet spills has improved. An additional next step in this comprehensive investigation is to determine whether the pellets on Great Lakes beaches have adsorbed persistent organic pollutants (POPs) on their surfaces. The combination of plastic debris and harmful pollutants could prove detrimental to aquatic animals and grazing birds in the various ecosystems of the Great Lakes.

Finally, the identification of shape, size, diagnostic trait, and color provided in Table S2 could prove very useful for pellet manufacturers and processers who wish to determine whether their products are contributing to pellet pollution in the Great Lakes watershed.

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CRediT authorship contribution statement

Patricia L. Corcoran: Conceptualization, Data curation, Formal analysis, Methodology, Project administration, Supervision, Visualization, Writing - original draft. **Johanna de Haan Ward:** Formal analysis, Software, Visualization, Writing - review & editing. **Ian A. Arturo:** Data curation, Formal analysis, Investigation, Methodology, Software, Visualization, Writing - review & editing. **Sara L. Belontz:** Conceptualization, Data curation, Formal analysis, Investigation, Methodology, Software, Visualization, Writing - review & editing. **Tegan Moore:** Conceptualization, Data curation, Formal analysis, Investigation, Writing - review & editing. **Carolyn M. Hill-Svehla:** Data curation, Formal analysis, Writing - review & editing. **Kirsty Robertson:** Conceptualization, Data curation, Formal analysis, Writing - review & editing. **Kelly Wood:** Conceptualization, Data curation, Formal analysis, Writing - review & editing. **Kelly Jazvac:** Conceptualization, Data curation, Funding acquisition, Writing - review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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