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Sources and Sinks of Microplastics in Canadian Lake Ontario Nearshore, Tributary and Beach Sediments

Anika Ballent
The University of Toledo


Patricia L. Corcoran
The University of Western Ontario

Odile Madden
Smithsonian Institution

Paul A. Helm
Ontario Ministry of Environment and Climate Change

Fred J. Longstaffe
The University of Western Ontario, flongsta@uwo.ca

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Corresponding Author: Ms. Anika Ballent,

Corresponding Author's Institution: University of Western Ontario

First Author: Anika Ballent

Order of Authors: Anika Ballent; Patricia L Corcoran, Ph.D.; Odile Madden, Ph.D.; Paul A Helm, Ph.D.; Fred J Longstaffe, Ph.D.

Abstract: Microplastics contamination of Lake Ontario sediments is investigated with the aim of identifying distribution patterns and hotspots in nearshore, tributary and beach depositional environments. Microplastics are concentrated in nearshore sediments in the vicinity of urban and industrial regions. In Humber Bay and Toronto Harbour microplastic concentrations were consistently >500 particles per kg dry sediment. Maximum concentrations of ~28,000 particles per kg dry sediment were determined in Etobicoke Creek. The microplastic particles were primarily fibres and fragments < 2 mm in size. Both low- and high-density plastics were identified using Raman spectroscopy. We provide a baseline for future monitoring and discuss potential sources of microplastics in terms of how and where to implement preventative measures to reduce the contaminant influx. Although the impacts of microplastics contamination on ecosystem health and functioning is uncertain, understanding, monitoring and preventing further microplastics contamination in Lake Ontario and the other Great Lakes is crucial.

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6

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21 further microplastics contamination in Lake Ontario and the other Great Lakes is crucial.
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26 **1. Introduction**
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28 Studies concerning plastics contamination of marine environments began in the 1970s (e.g.
29 Carpenter and Smith, 1972; Colton et al., 1974), and since that time, investigations have shown
30 that plastic waste is consistently found in aquatic environments including beaches, ocean surface
31 waters, deep-sea sediments, freshwater lakes, and tributaries (e.g. Eriksen et al., 2013; Van
32 Cauwenberghe et al., 2013; Vianello et al., 2013; C  zar et al., 2014; Turra et al., 2014; Eerkes-
33 Medrano et al., 2015). Contamination of aquatic environments with plastics has become a global
34 issue as a result of low recycling rates, designs that do not include the post-consumer stage of the
35 product, and lack of policies that support a circular plastics economy (Neufeld et al., 2016
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39 Microplastics, defined as plastic particles < 5 mm long (NOAA, 2015), are either manufactured
40 for use in applications such as cosmetics, personal care products, industrial abrasion processes
41 and synthetic fabrics (Fendall and Sewell, 2009; Eriksen et al., 2013; Sundt et al., 2014), or are
42 generated by the degradation of larger plastic products (Carpenter and Smith, 1972; Sundt et al.,
43 2014). In a recent study, microbeads, many of which were comparable to particles isolated from
44 cosmetic products, comprised an estimated 58% of microplastics < 1 mm in size collected from
45 the surface waters of the Great Lakes (Eriksen et al., 2013). Several countries, including Canada
46 and the United States, have since begun the process of banning the sale of cosmetic products
47 containing microplastics. Polyethylene (PE) and polypropylene (PP), often used in the
48 production of cosmetic microplastics, have densities less than that of fresh water, but have
49 nonetheless been regularly identified in submerged sediments (Claessens et al., 2011; Vianello et
50 al., 2013; Corcoran et al., 2015). The study of microplastics in sediment is therefore needed for a
51 comprehensive understanding of microplastics contamination in the Laurentian Great Lakes.
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57 Microplastics pollution has been reported from Great Lakes surface waters (Eriksen et al., 2013),
58 along shorelines (Zbyszewski and Corcoran, 2011; Zbyszewski et al., 2014; Corcoran et al.,
59 2015) and in offshore lake bottom sediments (Corcoran et al., 2015), but its presence in
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subsurface sediments of nearshore regions and tributaries is still largely unknown (Driedger et al., 2015). Allan et al. (2013) modeled current environmental stressors affecting the Great Lakes and found that cumulative stress levels were greatest in nearshore regions and generally decreased with distance from the shore. Only one investigation concerning microplastics pollution in Lake Ontario sediments has been published to date (Corcoran et al., 2015), despite it being the terminal lake within the Great Lakes system. In addition, the lake is surrounded by highly urbanized and industrialized regions, particularly along the northern shore, which have been associated with the accumulation of plastic debris (e.g. Barnes et al., 2009; Turra et al., 2014). We herein provide a regional investigation of the abundance and depositional patterns of microplastics in nearshore, tributary and beach sediments along the Canadian shoreline of Lake Ontario in an effort to provide: (i) a baseline for future monitoring, (ii) a means to assess potential sources of microplastics to the lake, and (iii) a distribution map of current microplastics hotspots.

Microplastics in benthic ecosystems pose an environmental threat primarily because organisms that feed on the benthic community may potentially ingest microplastics. Littoral and profundal fish species in large temperate lakes have diets that consist primarily of benthic organisms, and in Lake Ontario ~92% of fish and ~96% of invertebrate species are found in littoral habitats (Vadeboncoeur et al., 2011). The majority of aquatic species in Lake Ontario are closely connected to nearshore benthic habitats, and microplastics contamination of these sediments may be directly affecting the health of the lake ecosystem at many trophic levels. Although plastics are not classified as hazardous waste in Canada, certain plastic products, such as polycarbonate CD-ROM discs and polyurethane foams commonly used in furniture, are manufactured from hazardous derivatives (Lithner et al., 2009). Additives such as polybrominated diphenyl ethers (PBDEs; e.g. flame retardants) (Lithner et al., 2011; Rochman et al., 2014) and plasticizers such as Bisphenol-A are also often included to change the physical properties of the plastics (Oehlmann et al., 2009). Plasticizers have been shown to have negative impacts on the hormonal systems of invertebrates, fish and amphibians (Oehlmann et al., 2009). Conclusive evidence for the transfer of associated hazardous compounds from plastics to organisms is lacking, but several studies have suggested correlations between plastics ingestion and compromised physiological function (e.g. Teuten et al., 2009; Wright et al., 2013; Syberg et al., 2015). An additional threat of microplastics is their role as colonization surface habitats for bacteria or other organisms, which may lead to the invasive transport of microbial communities (e.g. Ye and Andrady, 1991; Zettler et al., 2013; Harrison et al., 2014; McCormick et al., 2014; Nauendorf et al., 2016).

A comprehensive overview provided by Eerkes-Medrano et al. (2015) highlights our limited understanding of microplastics contamination in freshwater systems. Rivers and urban tributaries, however, have been shown to be major transport pathways for microplastics and macroplastics (Moore et al., 2011; Gasperi et al., 2014; Lechner et al., 2014; Rech et al., 2014; Zhao et al., 2014; Corcoran et al., 2015; Naidoo et al., 2015). Whereas buoyant materials are transported on surface waters (Gasperi et al., 2014), non-buoyant materials are transported along the tributary bed (Moore et al., 2011; Morritt et al., 2014). Microplastic debris loads can be introduced into tributaries via non-point spill and litter sources as well as point sources such as effluent pipes (Lechner & Ramler 2015), storm water drainage outlets (Armitage & Rooseboom

2000) and possibly wastewater treatment plants, particularly during combined sewage overflow and bypass events during heavy precipitation conditions (MacDonald and Podolsky, 2009). Rech et al. (2014) and Corcoran et al. (2015) showed that macroplastic and microplastic debris loads carried by rivers are also deposited along river banks, suggesting that rivers are both depositional and erosional zones for anthropogenic debris.

2. Setting and Methods

2.1 Setting

Lake Ontario drains an area of slightly more than 64,000 km² and is the terminal lake in the Laurentian Great Lakes chain (Fig. 1). Prevailing wind patterns in the Toronto region from November to February are mainly NW to WSW, and gradually shift to ENE-dominated from March to October. The western end and the northwestern shore of the lake are characterized by several urban and industrial regions, including the cities of Hamilton, Mississauga and Toronto, with populations of 0.52, 0.71 and 2.62 million, respectively. Plastics manufacturing in Canada is concentrated in Southern Ontario (Statistics Canada, 2012). According to a 2014 statistical report of the Canadian Chemical Industry (CIAC), production of synthetic resins, fibres and rubbers included PE, ethylene vinyl acetate, polystyrene (PS), polyvinyl chloride (PVC), polyacrylamides, polyethylene terephthalate (PET), nylons, latex emulsions, polyesters, silicones and butyl and halobutyl rubbers (CIAC, 2014, p. 34). High-density resins comprised 16% and PE comprised 80% of Canadian synthetic resin/rubber exports by weight in 2013 (CIAC, 2014, p. 35). In total, ~3,500 kt of PE were produced in 2013 (CIAC, 2014, p. 34).

2.2 Sample collection

Nearshore, tributary and beach sediments were sampled using a combination of sediment trap, core and grab sampling techniques in order to obtain as many data points as possible. The original objective was to collect all nearshore samples using a Glew gravity corer, but this method proved to be especially challenging as a result of the high wave action in Lake Ontario. The Ontario Ministry of the Environment and Climate Change (MOECC) donated Shipek grab samples, in addition to passive sediment trap samples that they had collected from Lake Ontario (Nearshore Index and Reference sites, Great Lakes Nearshore Monitoring Program). Tributary sediment was best sampled using a petite ponar grab that could be lowered from a standing position. Beach sediment was sampled using a split spoon corer, as it was originally thought that depth below the surface could be related to microplastics abundance. However, given the highly variable hydrodynamic conditions that take place on beaches, we have chosen to represent all sediment depths at each beach as a whole. The use of various sampling methods enabled a greater spatial resolution, but temporal (annual and seasonal) constraints were not possible. Sample names, dates, sampling instruments, depositional environments, locations, and water depths are detailed in Table 1.

A total of 33 nearshore samples were collected (Table 1; Fig. 1). Five nearshore sediment samples were collected using a Glew gravity corer in August, 2014. The PVC cores measured 6.5 cm in diameter, and ranged between 6 and 15 cm thick. Immediately after recovery, the gravity cores were extruded into 1 cm intervals ($N \leq 15$) and stored in sealed polyethylene bags.

Twenty-five samples were collected by the MOECC from the Canadian shoreline of Lake Ontario and the proximal St. Lawrence River in August, 2012 and July, 2014 using a Shipek grab sampler with a square opening of 20 cm and a half-cylindrical cup with a radius of 10 cm (Wildco, Yulee, FL). Three replicate grab samples were collected from each station, homogenized in a metal tray and transferred into 0.5 L PET collection bottles. Three additional nearshore samples were collected by the MOECC from sediment traps in November, 2014. The traps comprised an array of four acrylic cylinders, secured at approximately 2 m above the lake bed, and were allowed to collect sediment for 6 months

Tributary sediment samples from Red Hill Creek, Etobicoke Creek, Humber River and Don River (Fig. 1) were obtained with a 16 x 14.5 cm hand-held, stainless steel Petite Ponar sediment grab (Wildco, Yulee, FL). Sample locations were constrained foremost by accessibility and second by the presence of sediment depositional zones. At each of seven sample locations, 2 grab sample replicates were combined in a metal tray and were spooned into 0.5 L PET collection bottles.

Sediment was also sampled from five east and south facing beaches along the north-western shore of Lake Ontario in June, 2015: Beachway Park, Bronte Beach, Marie Curtis Park, Sunnyside Beach and Woodbine Beach (Fig. 1). Much of the northern shore of Lake Ontario has been built up with large boulders or is inaccessible due to privatization of lands, which limited sample sites to public sandy beaches. Using a stainless steel split spoon corer with a height of 30 cm, two cores were collected from each beach at approximately 2 m and 6 m from the waterline, representing the proximal and distal foreshore, respectively. The foreshore sediments are regularly submerged with seasonal changes in water level as well as during storm surges. The corer was equipped with an inner, segmented PVC core, each segment measuring 10 cm long. Upon opening the core, each segment was covered with a polyethylene cap. All proximal foreshore sites, except at Marie Curtis Beach, were limited to a sampling depth of 20 cm due to the high water content below that depth. All samples were kept cool until returned to the laboratory where they were stored at -25°C until analysis.

2.3 Microplastic quantification

The sediment samples were dried at 70°C and weighed. All samples, except those collected by gravity core, were sieved using a Taylor sieve shaking apparatus with sieve mesh sizes of 5.6 mm, 2.0 mm, and 0.063 mm for 5 minutes at 60 Hz. Consolidated sediment samples with high clay fractions were first wet sieved through a 0.063 stainless steel sieve, then dried and weighed again to calculate the fine fraction. Following sieving, each fraction was weighed, the >2 mm and >5 mm fractions were visually examined for microplastic, and the >0.063 mm fraction was transferred through a sample splitter. Half of the >0.063 fraction was used for density separation. Gravity core samples were not separated by sieving because their very fine grain size resulted in flocculation during drying. The resultant clumps did not disaggregate during sieving, but the estimated lower grain size limit was 0.025 mm. All samples were density separated using a sodium polytungstate (SPT) density separation technique. Each sample was combined with SPT, (1.5 g cm⁻³), magnetically stirred for 2 minutes and then transferred to a glass separation funnel to settle. After draining the non-buoyant material, the buoyant material was drained into a

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4 polycarbonate/polyester 0.053 μm sieve, rinsed thoroughly with filtered deionized water,
5 transferred to a second separation funnel containing 500 mL of filtered deionized water, and then
6 allowed to settle. The resulting non-buoyant and buoyant fractions were consecutively drained
7 through a polycarbonate/polyester 0.053 μm sieve and transferred to glass vials. Samples were
8 dried at 70°C and were covered until visual processing. Samples collected using the gravity corer
9 were not split or sieved; the microplastic-containing fraction was separated by decanting and
10 filtering through VWR[®] Grade 114 qualitative fast flow 25 μm filter paper.
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14 Microplastics were visually identified using a Nikon SMZ1500 stereo microscope at
15 magnifications ranging from 15 \times to 225 \times . With decreasing particle size, the ability to identify a
16 particle as plastic becomes reduced due to fewer visual and textural clues as discussed by Lenz et
17 al. (2015) and Song et al. (2015). Even though particles at the lower end of the size limit defined
18 by the filter or sieve can be magnified sufficiently, lower observation limits are constrained by
19 the ability to distinguish synthetic particles from other particles using color (bright, non-natural
20 colors are easier to detect), presence of shiny lustre (often associated with plastics) and other
21 visual and textural cues. In addition, the higher the magnification, the more time is needed to
22 process a sample. In light of these limitations, the lower size limit of visual identification of
23 plastics is estimated to be ~0.25 mm. Microplastic particles were counted and sorted by type into
24 3 categories: fibres, fragments and spherical beads. All particles were photographed using a
25 Nikon digital camera DXM1200F connected to the microscope.
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31 Throughout the sampling and laboratory analysis, precautions were taken to minimize
32 contamination of the samples from airborne microplastic. Containers holding samples were kept
33 covered with aluminum foil throughout the process except during periods when the samples were
34 drying in the closed oven. The laboratory surfaces were routinely wiped down and all beakers,
35 trays, containers, funnels, tools and sieves were thoroughly washed and rinsed with filtered
36 deionized water before and after each use and were stored with openings covered in aluminum
37 foil. Metal and glass containers and tools were used in all analyses, except for a
38 polycarbonate/polyester mesh sieve. Sampling containers used in the field were plastic; however,
39 precautions were taken that all materials were either cleaned prior to use or were new and
40 unopened containers. Clothing worn by researchers were of natural fibres and in the laboratory,
41 white cotton laboratory coats were worn. During analysis, doors to the corridors were kept closed
42 whenever possible. To test for airborne microplastic contamination levels during sample
43 processing in the laboratory, petri dishes (cleaned and microscope inspected) were set in the
44 working space of each lab room and the drying oven for 2 hours, immediately followed by visual
45 inspection with the same stereo microscope used for sediment sample analysis. Two replicate
46 tests were conducted for each space.
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52 *2.4 Polymer identification*

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54 In order to determine the relative abundance of various polymer compositions, microplastic
55 fragments and beads were randomly selected for analysis by Raman spectroscopy and X-ray
56 fluorescence spectroscopy (XRF) at the Museum Conservation Institute at the Smithsonian
57 Institution. Raman spectroscopy was conducted with a NXR Fourier-transform Raman module
58 coupled to a 6700 Fourier transform infrared spectrometer (Thermo Electron Corporation,
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Madison, WI, USA). The FT-Raman module was equipped with a continuous wave near infrared ND:YVO₄ excitation laser (1064 nm), a germanium detector cooled with liquid nitrogen and a CaF₂ beam splitter. Laser power was chosen empirically to maximize signal-to-noise ratio (SNR) without damaging the sample, and ranged from 0.01-0.07 Watts across a 50 micron round laser spot. Spectra comprised a co-addition of 64-2048 scans collected at 8 cm⁻¹ resolution across 98-3994 cm⁻¹ Raman shift. The performance of the spectrometer was checked against a reference scan of polystyrene and recalibrated as necessary each day of analysis. Raman spectra were plotted with OMNIC™ software (Thermo Scientific, Madison, WI, USA) and compared to commercial spectral libraries¹ and custom libraries prepared by the Smithsonian's Museum Conservation Institute. All spectra were analyzed using the automated search function of the software and also visually before identification was finalized. Of the 6,331 plastic particles identified in the sediment samples, a total of 90 particles were analyzed by Raman spectroscopy. Using a random number generator, up to twelve particles were selected from three tributary samples and two 1 cm intervals from each of the five nearshore gravity core samples. Particles selected by the random number generator that could not be successfully analyzed (i.e. the material was too dark or thin) were replaced by those associated with the next randomly generated number. Fibres were excluded due to their insufficient diameter and volume. Some samples were also analyzed by X-ray fluorescence spectroscopy (XRF) to confirm the presence of chlorides in suspected polyvinyl chloride plastics. The instrument used was a Bruker Artax 400 μXRF spectrometer equipped with a Rh-tube, a poly-capillary lens with a ~100 μm focal spot and a Peltier-cooled Si-drift detector. The excitation voltage ranged between 25-50 kV and the current ranged between 490 and 492 μA.

2.5 Geographic spatial analysis

Population density and plastics-related industries were mapped on a watershed basis to gain insight into the relative levels of urban and industrial waste that may be expected to enter the lake through tributaries along the shoreline of Lake Ontario. Esri® geographic information software ArcGIS was used to calculate and map these two variables and to map the spatial variation of microplastics. Geographic shapefile datasets for the primary-level watershed boundaries (areas draining to Lake Ontario and the St. Lawrence River) and quaternary-level watershed boundaries (river and coastal stream catchment basins) were retrieved from the Government of Ontario Open Source Data Catalogue and Land Information Ontario. The datasets were used to generate a shapefile for quaternary-level watershed boundaries in which only those watersheds that drain into Lake Ontario and the St. Lawrence River were included, and in which all watersheds include a portion of the Lake Ontario or St. Lawrence River shoreline (Fig. 1). Multiple drainage points per watershed were permitted where small coastal creeks were located and for which separate quaternary-level watersheds were not delineated.

¹ HR FT-Raman Polymer Library (copyright 1997-2001, 2004 Thermo Electron Corporation for Nicolet Raman). HR Pharmaceutical Excipients FT-Raman Library (copyright 1999, 2004 Thermo Electron Corporation for Marcel Dekker, Inc.). FDM Retail Adhesives & Sealants (Fiveash Data Management, Inc., Madison, Wisconsin, USA)

Canadian 2011 Census data obtained from Statistics Canada were used to determine the approximate human population of each watershed. Wastewater treatment plants proximal to the Ontario shoreline of Lake Ontario and St. Lawrence River were plotted using facility addresses. Storm drain outlets and combined sewer overflow outfalls along the tributaries and lake shore are densely located, particularly in urban regions. The abundance of outfalls would be expected to correlate well with population levels on the scale of the study, as indicated by a map showing storm drain outfalls along Taylor Creek, tributary of the Don River (City of Toronto, 2006). They are therefore not individually mapped in this study. Plastics-related industry contact addresses were collected from ThomasNet, an online supplier discovery and product-sourcing directory. Through the search engine, businesses with descriptions or names containing the word 'plastic' were selected, and verified to be in the plastics industry by qualitative analysis of the description. Results were constrained to suppliers located in Ontario, Canada and categorized according to type: manufacturer, distributor and service. All businesses were considered equally in counting the number of businesses located in each watershed. For businesses with multiple locations, all locations in Ontario were included. The search was not exhaustive, and the list of locations used in this study may be incomplete; however, registration in the ThomasNet directory is free and includes a large number of plastics-related businesses. Geographic locations of microplastics samples ($N=51$), recorded using a handheld Geographic Positioning Device (GPS), were formatted into a point feature shapefile. Datasets were converted within ArcGIS to a common projected coordinate system used for this project: North American Datum 1983, Universal Transverse Mercator, Zone 18 North (NAD83 / UTM-18N). Spatial analysis was completed primarily in vector space, but drainage area and population were calculated in raster space at a resolution of 100 m. Plastics-related industry counts were calculated using a spatial join function.

3. Results

3.1 Microplastics abundance

A total of 6,331 particles were visually identified as microplastics. Microplastics abundance for each sample site was normalized to particles per kg of dry sediment ($N \text{ kg}^{-1}$, dw) using the initial mass of the dried sediment sample (Table 1). Microplastics were identified in every sediment sample, and abundances varied between 20 and 27830 kg^{-1} (Fig. 2). Maximum microplastics abundance was found at site P-EC2, at the mouth of Etobicoke Creek. The sample was primarily composed of algae, and therefore the extrapolated total number of microplastics is exceptionally high due to the low mass of the dried algae. Additional results and analyses presented here exclude this data point unless specifically noted. On average (not including P-EC2), microplastics abundance was 760 kg^{-1} . In the sample processing lab, the drying oven and microscopy lab airborne contamination levels were 2, 3 and 1.5 fibres hr^{-1} of exposure, respectively on the scale of a standard glass Petri dish (area: $A=64 \text{ cm}^2$). During processing, extreme caution was taken to minimize sample exposure. Contamination of the sediment samples with fibres may have occurred during the drying stages of sample preparation during which sample containers ($A=2-315 \text{ cm}^2$) were exposed for up to 24 hours.

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4 Nearshore sediments contained on average the greatest abundance of microplastics (980 kg^{-1}),
5 followed by tributary sediments with 610 kg^{-1} and beach sediments with 140 kg^{-1} (Fig. 3a).
6 Microplastics in nearshore sediments were most concentrated in Humber Bay and Toronto
7 Harbour. Microplastic abundances of $> 1000 \text{ kg}^{-1}$ were found only at sites in the Greater Toronto
8 Area (GTA) and offshore of Oakville (sample S-7541), west of Toronto. Microplastics
9 abundance, as recorded in the sediments sampled from the gravity core, generally decreased
10 within the top 2 g cm^{-2} of sediment accumulation, but microplastics were found at all sampled
11 depths up to 15 cm (equivalent to 11.2 g cm^{-2} accumulated sediment mass) below the sediment
12 surface (Fig. 4a). Nearshore sediments collected with the gravity cores had relatively high
13 microplastic concentrations compared to the trap and grab samples; average microplastic
14 abundance for gravity core, trap and grab samples were 2130 kg^{-1} , 1070 kg^{-1} and 730 kg^{-1} ,
15 respectively.
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21 Microplastics abundance in tributary sediments appear to display variability on small spatial
22 scales (Fig. 2, Table 1). For example, in Humber River and Etobicoke Creek, downstream sites
23 contained microplastic counts one to two orders of magnitude greater than sites within 1 km
24 upstream. Abundances of microplastics in beach samples appear to decrease with greater
25 distance from Toronto (Fig. 4c). Microplastic contamination loads in Lake Ontario are
26 comparable to those reported in similar studies from around the world, as summarized in Table
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30 31 *3.2 Microplastic morphology*

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33 Microplastics in nearshore depositional zones were almost exclusively $< 2 \text{ mm}$ in size.
34 Microplastics $> 2 \text{ mm}$ were found only in one nearshore sediment sample in Toronto Harbour
35 (site S-3030). Plastic particles $> 2 \text{ mm}$ were slightly more common in tributary sediments with
36 84 particles at site P-EC2 and 1 particle at site P-RC2; overall, 4% of the microplastics found in
37 tributary samples were $> 2 \text{ mm}$ in size. Approximately 8% of plastics in beach samples were > 2
38 mm, the majority of which were found at Marie Curtis and Sunnyside (Fig. 5a) beach sites and at
39 the mouth of Etobicoke Creek (sample P-EC2).
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43 Fragments and fibres were the dominant morphologies for microplastics $< 2 \text{ mm}$ across all
44 depositional zones, with beads being a minor component (Fig. 3b). Fragments (including films)
45 and beads (industrial pellets) were the most common microplastics $> 2 \text{ mm}$. Fibres were most
46 abundant in nearshore samples and least abundant in tributary samples. Beads were found in all
47 depositional environments, but were not found in the sediment traps.
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51 In general, microplastics were of variable colour, texture, grade of degradation, size and shape.
52 Fibres were highly regular in diameter along their entire length and varied in colour and length
53 from tens of microns to several millimeters (Fig. 5b). Irregularly shaped, bulbous to wispy
54 fragments with smooth surfaces and translucent diaphaneity were common (e.g. Fig. 5c, f).
55 Beads were mainly spherical, translucent, amber or black, and sometimes cracked or fragmented
56 (Fig. 5c, far right column). Oblong, helical forms composed of rigid, opaque material were
57 common in the tributary and nearshore sediments, particularly in Etobicoke Creek (Fig. 5d).
58 These particles had smooth clean surfaces and did not appear fragmented. In a conservative
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classification, we estimate that at least 4% of all microplastics were of this form. At Etobicoke Creek, however, these particles made up ~30% of all fragments < 2 mm. Non-rigid, black, opaque particles appearing as amorphous chunks or long thin helical twists accounted for 7% of microplastics collected in the study (Fig. 5e).

3.3 Polymer composition

Of 90 suspected microplastics analyzed using Raman spectroscopy, 60 (67%) were positively identified as synthetic polymers. An additional five particles could not be identified but were found to contain phthalates, which are plasticizers, and toluidine red, a pigment commonly used for plastic products. Three samples were identified as non-plastic. These particles, mistaken for microbeads, were identified as quartz and calcium carbonate and were likely well-rounded sand grains. Thirty-one percent of the analyzed particles were identified as PE, 10% as PS and 4% as polyurethane (PU) (Fig. 6). Polypropylene (PP), PVC, and polystyrene sulfonate (PSS) each made up 3% of the analyzed particles. Other polymers including PET, polymethyl methacrylate (PMMA), polyvinyl/vinyl acetate copolymer, PMMA-PS copolymer or mixture, acrylonitrile butadiene styrene (ABS), nylon, phenoxy/epoxy resin, and polymethylsiloxane (silicone) were identified (Fig. 6). Twenty-two samples remain unidentified. Microplastic counts were not adjusted to reflect the plastic to non-plastic ratio of the Raman analysis results due to the low percentage (1.4%) of particles analyzed.

Several patterns relating plastic morphology and composition were recognised. Fragments identified as PE were consistently characterised by translucent diaphaneity and irregular wispy and bulbous forms, also noted by Corcoran et al. (2015). The black opaque rubbery particles could not be identified using Raman spectroscopy due to their tendency to combust even at very lower laser power. This problem was also noted by Lenz et al. (2015); however, their analyses provided identification of similarly described particles as black tire rubber. Yellow, translucent, rigid fragments were commonly identified as PU. Several of the smooth, spherical, transparent, amber-red beads (Fig. 5c, bottom right) were identified as polystyrene sulfonate (PSS).

3.4 Watershed analysis

A total of 66 watersheds were analyzed for their population and abundance of plastics-related industries. The Don River watershed had the greatest population, whereas the Toronto Urban Catchment watershed had the highest population density with ~7380 people km⁻² (Fig. 7). Both of these watersheds empty into the Toronto Harbour. There are approximately 20 major wastewater treatment plants along the Canadian shore of Lake Ontario and the upper St. Lawrence River (Fig. 7), which could contribute plastic debris to the lake. With regards to industry intensity, Etobicoke Creek contains the largest number of plastic product manufacturers, distributors and service businesses combined, at 62 business facilities. There is a clear clustering of the plastic industry in the Greater Toronto Area extending towards the Hamilton Region (Fig. 8). Several watersheds did not include any plastic industry facilities, as identified through the ThomasNet directory.

4. Discussion

4.1 Lateral trends in deposition

The intense microplastics contamination in samples from the Greater Toronto Region may be attributed to the high population density and industrial activity in the watersheds draining into this region. The Etobicoke Creek, Mimico Creek, Humber River, Toronto Urban Catchment, and Don River watersheds have a combined population of 3.4 million, which accounts for 40% of the total population of all watersheds draining into Lake Ontario, in Canada. Likewise, half of the plastics production facilities in the study region are located in the same five watersheds. Higher resolution sediment sampling along the northwest shore of Lake Ontario may reveal a clearer trend between microplastics abundance and watershed population- and industry-density. For the same reason, accounting for sewage and storm outlets as point sources of microplastics by sampling consistently near these outlets is suggested for future studies.

The high concentrations of microplastics in Toronto Harbour and Humber Bay may also be influenced by the morphology of the shoreline at those locations. It has been shown that deposition of microplastics in bottom sediments occurs in low energy environments, such as harbours and lagoons, where fine particles supplied by fluvial and anthropogenic outputs can settle (Claessens et al., 2011; Vianello et al., 2013). Average circulation patterns in Lake Ontario as modeled by Beletsky et al. (1999) show that in both summer and winter, currents in the vicinity of Toronto Harbour move along the shore from southwest to northeast. We expect that the peninsula located just west of Humber Bay protects the southeast shore of Humber Bay and the Inner Toronto Harbour from severe waves during the dominant anticyclonic surface water circulation during the summer and winter in the northwestern basin of Lake Ontario (Beletsky et al., 1999). Similarly, the Toronto Islands may reduce water flow velocity in the Inner Toronto Harbour and Humber Bay during easterly long-fetch storm events. The relative abundance of microplastics in this region may result from the lower frequency of resuspension and transport events associated with weaker hydrodynamic forcing. Although the “harbouring effect” may allow greater accumulation of microplastics contamination, it is only one of many variables affecting the spatial variability of plastic debris. For example, Hamilton Harbour has relatively low microplastic contamination levels in comparison to what would be expected given the coastal morphology of the harbour. This indicates that microplastics in sediments are foremost dependent on source loads.

The dense concentration of microplastics at the mouth of Etobicoke Creek compared to upstream sediments is consistent with reduced boundary shear stress, the driving force of particle motion in a transport fluid, as tributaries widen and flatten into the lake. In a marine model study, strong unidirectional flows associated with internal waves and storm events were needed to transport non-buoyant plastic particles down-slope in a submerged environment (Ballent et al., 2013). As theoretically applied to Lake Ontario, microplastics should be transported by rapid flow in tributaries and during storm events and deposited as turbulence and bottom currents subside, for example, at the mouths and banks of tributaries. Our results show, however, that microplastics are also found in tributary sediments where higher flow regimes dominate, and in nearshore, open environments where sediments are exposed to erosion and large-scale transport. Further research is needed to understand the similarities and differences between the transport mechanics

of microplastics and other sedimentary particles such as clastic grains, clays and organic aggregates in aquatic environments.

Microplastic contamination levels may also be influenced by the presence of obstructing structures, variations in topography, and areal extent of the watershed. Reduced flow rates due to obstructing structures or flatter topography may be associated with increased microplastic deposition upstream, thereby reducing microplastics abundance at the river mouth. Several of the watersheds in the study are amalgamated river systems, many of which have control structures such as dams, weirs, reservoirs and lakes. The input locations of microplastics should be considered in geographic relation to the topographic and anthropogenic features of the tributary. Several questions to be addressed by future investigations of microplastics transport through tributaries are: What distances are microplastics being transported before being deposited? What are the minimum flow velocities and shear stresses found in tributaries and how much do these values vary over the length of a tributary? Where in the tributaries are shear stresses sufficient to transport microplastic? What percentage of the plastics found in the sediments were originally positively buoyant?

Spatial variability on scales of tens to several hundreds of meters was exhibited in the Toronto Harbour and Humber Bay region where sample sites were close and where multiple samples were collected from the same site. Tributary site P-EC2, for example, had two orders of magnitude more microplastic particles than the beach sediments within 100 m away at Marie Curtis Park, and one order of magnitude more microplastics than sediments only 350 m upstream. Similarly, the four separate grab and trap samples taken from the Humber Bay Index station (S-7546 and S-HB14, S-3025, T-2047) along the Toronto waterfront had microplastic counts between 40 and 2210 kg⁻¹ highlighting the temporal variability and meter-scale spatial variability of samples collected from the same station. The observed variability is possibly reflective of the turbulent and random nature of fluid flows and surface characteristics such as substrate type, topography, roughness and presence of vegetation (e.g. Vianello et al., 2013; Corcoran et al., 2015).

4.2 Vertical trends in deposition

The nearshore gravity core samples provide insight into the vertical variability of microplastics in lake bottom sediments where plastic concentrations are possibly the highest in Lake Ontario (Fig. 4a). Microplastic abundance was extrapolated according to sediment weight, and therefore the relative abundance of microplastics in the top 2 cm can be attributed to the greater content of water and natural organic material in the upper unconsolidated layer.

Sediment accumulation rates of ~1.7 mm yr⁻¹ are estimated for the nearshore environment of the western basin of Lake Ontario based on pollen dating of *Ambrosia* (first occurrence 120 years B.P.) as calculated by Rukavina (1976). The deepest gravity core, which penetrated to a depth of 15 cm, may therefore represent the last ~90 years. However, the increase in water content in the unconsolidated sediment of the upper layer points to a much shorter time period. Furthermore, frequent resuspension of surface sediments in nearshore (depth < 40 m) lake environments by storm events (Klump et al., 2000) can resuspend material on scales equal to those deposited

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4 annually in lake basins within a period of days, as shown for Lake Michigan (Eadie et al., 1996;
5 Schwab et al., 2000). Material resuspended by these events may be confined to the nearshore in
6 the presence of a coastal plume (Mortimer, 1988; Hall, 2008). Further research is needed to
7 constrain the timescales of microplastics transport and deposition in transient nearshore and more
8 permanent offshore basins. Microplastics in offshore basin sediments of Lake Ontario were
9 reported to extend to a maximum of 8 cm below the sediment surface (Corcoran et al., 2015),
10 which is consistent with a lower sediment accumulation rate compared with that of the nearshore
11 locations studied here.
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15 *4.3 Physical characteristics and transport behaviour*

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17 The density and shape of microplastic particles may also impact distribution patterns identified
18 in Lake Ontario. Plastics range in density from ~ 0.8 to 1.4 g cm^{-3} , which is less than the average
19 density of mineral sediments ($1.6\text{-}2.7 \text{ g cm}^{-3}$, (Fettweis et al., 2007; Hidalgo-Ruz et al., 2012).
20 Theoretically, PE and PP particles should float because their densities are lower than that of
21 freshwater, however, PE was the most common type of plastic among the Raman analyzed
22 samples. Plausible mechanisms for the deposition of low-density polymers in submerged
23 sedimentary environments include net density increase of microplastic particles by biofouling
24 (e.g. Ye and Andrady, 1991; Andrady, 2011; Zettler et al., 2013; McCormick et al., 2014),
25 adsorption of natural substances to the surface (Frias et al., 2016), inclusion of inorganic fillers
26 during manufacturing (Corcoran et al., 2015) and faecal express (Cole et al., 2013; Setälä et al.,
27 2014; Zalasiewicz et al., 2015). Inorganic fillers were not identified in any of the Raman spectra
28 of the particles identified as PE and PP in this study, however, many microplastics in our study,
29 particularly those with irregularly shaped, textured or degraded surfaces, appeared to have clay-
30 like particles adhered to their surfaces. Microplastic particles made of PE and PP and with
31 greater surface area to volume ratios, such as fibres and irregularly shaped fragments, are
32 expected to have lower settling velocities and lower shear stress values needed to initiate particle
33 motion than microplastic particles of high-density polymer types and mineral sediments. They
34 may therefore be transported at lower flow and turbulence levels and be more readily transported
35 offshore, consistent with the findings of Corcoran et al. (2015). Denser microplastics may behave
36 more similarly to mineral sediments and be more constrained to nearshore areas.
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43 Comparisons of microplastic types recovered from samples of different sedimentary
44 environments is challenging because of the variations in sampling methods. Simply comparing
45 abundances, as normalized to sediment mass, the high proportion of fibres to fragments in the
46 nearshore sediments compared to the tributary sediments may suggest that fibres are transported
47 through suspension for greater distances than fragments, which may have higher settling
48 velocities or are transported as bedload. Sediment traps, which collect particles falling out of
49 suspension from the overlying water column, contained almost exclusively fibres. The overall
50 low proportion of microbeads in all environments contrasts with the surface water results of
51 Eriksen et al. (2013), suggesting that microbeads remain suspended in the water column as a
52 result of their low density. Notwithstanding, the low sampling resolution combined with the use
53 of multiple sampling techniques make these observations speculative. More thorough analysis of
54 microplastic morphology and composition may reveal trends regarding which types of plastics
55 are most easily transported to depth in an aquatic environment.
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59 *4.4 Potential sources*

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4 Our results indicate that microplastics in coastal sediments of Lake Ontario likely originate in
5 proximal watersheds and are likely transported to the site of deposition through tributaries.
6 Future studies of microplastics in sediments directly adjacent to storm water, wastewater
7 treatment plant, and combined sewer outfalls may reveal a clearer trend regarding how much
8 plastic is contributed by these sources, and how microplastics abundance in the sediment varies
9 with distance from outfalls. Assigning particular origins to the microplastics is challenging due
10 to their small size, fragmented nature, and the unknown range of possible sources. A speculative
11 discussion of the potential sources of microplastics contamination in Lake Ontario follows.
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15 Polyurethanes are commonly used in the production of foams for furniture, as well as in
16 adhesives such as construction glue products, surface coatings and sealing applications. The
17 black, opaque fragments with rubber-like consistency (Fig. 5e) may originate from vehicle tires
18 as suggested by Lenz et al. (2015). The natural wear down process of tires during driving may
19 contribute small particles such as those seen in our study. These particles could easily be washed
20 from roads to storm drains during rain events. Similarly, the shredding of used tires for recycling
21 purposes referred to as crumb rubber, as defined by Regulation 347: General Waste Management
22 under the Environmental Protection Act, Revised Statute of Ontario, 1990, may also contribute <
23 2 mm particles. Fibrous microplastics are thought to originate from the production, washing and
24 the natural aging of textiles, such as synthetic clothing and carpets (e.g. Browne et al., 2011).
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29 The amber-colored beads (Fig. 5c, right most column) identified to contain PSS may be
30 polystyrene resin beads that are commonly used as an ion exchange medium for water
31 purification and softening, as well as in various medical and industrial applications (Dardel,
32 2016). The beads represent a source of microplastics not yet discussed in the literature.
33 Microbeads have recently become an environmental focus in the scientific and political realms
34 with the result of new legislation. The Microbead-Free Waters Act, adopted in the United States
35 in 2015, bans the manufacture of microbead-containing cosmetic products by July 2017. In
36 Canada, microbeads manufactured for use in cosmetic products may be added to the List of
37 Toxic Substances in Schedule 1 of the Environmental Protection Act of 1999. This ban would
38 not, however, address the majority of the beads identified in the sediments, assuming they are
39 indeed used for non-cosmetic purposes. It is possible, however, that many of the irregularly
40 shaped particles in our samples which were categorized as fragments, originate from cosmetic
41 products. Leslie (2014) reports that microplastics used in cosmetics, such as facewashes and
42 toothpastes, range in shape from spherical to amorphous, suggesting that 'microbeads' are not
43 limited to bead morphologies.
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50 The oblong, helical fragments (Fig. 5d) could originate from finishing processes during the
51 manufacture of injection-moulded plastics. A common finishing process, termed deflashing,
52 involves the removal of extraneous material, flash, from the seams and edges of solidified
53 products where resin may have leaked into voids between the mould halves (SME, 2016). The
54 shaving-like particles identified in our samples could potentially be the waste flash particles
55 resulting from this process. A study by Lechner and Ramler (2015) identified an industrial point
56 source of microplastics along the Danube River in Austria. According to their investigation, the
57 allowable plastic loads in wastewater of the manufacturing plant was 30 mg l⁻¹, which translates
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to the equivalent of almost 95 tons of plastic waste per year, as calculated for flow rates of 100 l s⁻¹. Considering that plastics are not a regulated constituent of waste water in Ontario under the Ontario Environmental Protection Act, R.S.O. 1990, it is possible that substantial loads of microplastics are being released by the manufacturing and moulding facilities and draining directly into Lake Ontario.

5. Conclusions and Outlook

The spread and quantity of microplastics found in nearshore, tributary and beach sediments of Lake Ontario suggest that this contaminant may be entering the food web through ingestion by benthic fauna and higher-trophic organisms such as birds and fish. The wide range of polymers and particle morphologies represented in Lake Ontario sediments reflects various sources, potentially traced to industrial, transportation, construction and consumer activities. Reducing the influx of microplastics contamination will similarly require a range of measures addressing the various sources, for example, continued improvement and monitoring of waste management programs in cities and in industrial settings, such as Operation Clean Sweep, as well as an accelerated transition to a circular plastics economy (Neufeld et al., 2016). We suggest future monitoring of microplastics in the sediments of Lake Ontario and feeding tributaries, particularly in the GTA region, as well as a more detailed investigation of the input locations of microplastic along the tributaries and lake shoreline to further improve strategies targeting contamination reduction.

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57 Tables

58
59 Table 1. Summary of sediment samples by name, site location description, depositional
60 environment, sampling instrument type, date, geographic coordinates and underwater depth. A
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depth of zero is assigned to beach samples taken above the lake water level. Microplastic abundance is reported as particles kg of dry sediment; $N \text{ kg}^{-1}$. The fine fraction (clay and silt sized particles) is reported as the percent of sediment $< 63 \mu\text{m}$.

Sample	Site*	Environ.	Type	Year	Month	Lat. (°N)	Long. (°)	Depth (m)	$N \text{ kg}^{-1}$	% sed. $< 63 \mu\text{m}$
S-7481	Six Mile Cr	Nearshore	Grab	2012	Aug	43.320	-78.979	18	320	6.79
S-7486	Port Dalhousie	Nearshore	Grab	2012	Aug	43.228	-79.283	19	290	92.0
S-7491	Stoney Cr	Nearshore	Grab	2012	Aug	43.268	-79.671	22	70	24.4
S-7541	Oakville	Nearshore	Grab	2012	Aug	43.426	-79.661	21	1360	78.9
S-7546	Humber Bay	Nearshore	Grab	2012	Aug	43.623	-79.447	15	280	59.0
S-7553	Toronto Hb	Nearshore	Grab	2012	Aug	43.632	-79.370	9	3210	96.7
S-7501	Pickering	Nearshore	Grab	2012	Aug	43.794	-79.085	20	230	4.40
S-7506	Chub Point	Nearshore	Grab	2012	Aug	43.953	-78.012	21	140	4.23
S-7514	Trenton	Nearshore	Grab	2012	Aug	44.088	-77.544	3	800	31.4
S-7509	Prince Edward	Nearshore	Grab	2012	Aug	43.958	-76.812	21	430	81.7
S-7521	North Channel	Nearshore	Grab	2012	Aug	44.181	-76.735	24	780	97.4
S-7526	McDonnell Bay	Nearshore	Grab	2012	Aug	44.234	-76.375	4	120	15.8
S-7531	Prescott	Nearshore	Grab	2012	Aug	44.698	-75.532	3	40	1.41
S-7536	Lake St. Francis	Nearshore	Grab	2012	Aug	45.137	-74.416	12	80	5.84
S-3025	Humber Bay, index	Nearshore	Grab	2014	Jul	43.623	-79.447	15	230	46.6
S-3026	Humber Rv, mouth	Nearshore	Grab	2014	Jul	43.633	-79.464	8	730	3.47
S-3027	Humber Bay, STP	Nearshore	Grab	2014	Jul	43.626	-79.466	8	2550	71.2
S-3028	Toronto Hb, index	Nearshore	Grab	2014	Jul	43.632	-79.370	9	1590	91.6
S-3029	Don Rv, mouth	Nearshore	Grab	2014	Jul	43.642	-79.361	9	1250	96.2
S-3030	Toronto Hb, west	Nearshore	Grab	2014	Jul	43.633	-79.390	7	2790	82.0
S-HB14	Humber Bay	Nearshore	Grab	2014	Jul	43.623	-79.447	15	50	66.4
S-TH14	Toronto Hb	Nearshore	Grab	2014	Jul	43.632	-79.370	9	530	92.6
S-3031	Hamilton Hb, index	Nearshore	Grab	2014	Jul	43.289	-79.836	24	130	N/A
S-3032	Hamilton Hb, west	Nearshore	Grab	2014	Jul	43.281	-79.872	13	210	N/A
S-3033	Hamilton Hb, SE	Nearshore	Grab	2014	Jul	43.285	-79.794	22	160	N/A
T-258	Hamilton Hb, index	Nearshore	Trap	2014	Nov	43.289	-79.836	24	260	90.0
T-2047	Humber Bay, index	Nearshore	Trap	2014	Nov	43.623	-79.447	15	2210	97.0
T-1364	Toronto Hb, index	Nearshore	Trap	2014	Nov	43.632	-79.370	9	750	96.0
G-HB1	Humber Bay	Nearshore	Core	2014	Aug	43.630	-79.466	6	1240	77.9
G-HB2	Humber Bay	Nearshore	Core	2014	Aug	43.629	-79.469	4	990	88.4
G-HB3	Humber Bay	Nearshore	Core	2014	Aug	43.626	-79.473	4.5	3470	91.7
G-TH1	Toronto Harbour	Nearshore	Core	2014	Aug	43.631	-79.409	2.5	4270	36.7
G-TH2	Toronto Harbour	Nearshore	Core	2014	Aug	43.627	-79.383	5	670	66.2
P-DR1	Don Rv	Tributary	Grab	2015	Jun	43.691	-79.360	0.18	480	23.3
P-HR1	Humber Rv	Tributary	Grab	2015	Jun	43.652	-79.493	0.23	100	0.28
P-HR2	Humber Rv	Tributary	Grab	2015	Jun	43.642	-79.491	0.26	1740	51.8
P-EC1	Etobicoke Cr	Tributary	Grab	2015	Jun	43.587	-79.545	0.09	1210	27.7
P-EC2	Etobicoke Cr	Tributary	Grab	2015	Jun	43.585	-79.542	2.5	27830	0.00
P-RC1	Red Hill Cr	Tributary	Grab	2015	Jun	43.240	-79.774	0.35	100	22.7
P-RC2	Red Hill Cr	Tributary	Grab	2015	Jun	43.240	-79.774	0.11	40	1.01
C-BW1	Beachway Park	Beach	Core	2015	Jun	43.312	-79.800	0	60	0.09
C-BW2	Beachway Park	Beach	Core	2015	Jun	43.312	-79.800	0	60	0.09
C-BB1	Bronte Beach	Beach	Core	2015	Jun	43.392	-79.710	0	20	0.04
C-BB2	Bronte Beach	Beach	Core	2015	Jun	43.392	-79.710	0	70	0.03
C-MC1	Marie Curtis Park	Beach	Core	2015	Jun	43.584	-79.542	0	50	0.06
C-MC2	Marie Curtis Park	Beach	Core	2015	Jun	43.584	-79.542	0	190	0.09
C-SS1	Sunnyside Beach	Beach	Core	2015	Jun	43.637	-79.450	0	470	0.05
C-SS2	Sunnyside Beach	Beach	Core	2015	Jun	43.637	-79.450	0	250	0.09
C-WB1	Woodbine Beach	Beach	Core	2015	Jun	43.666	-79.299	0	170	0.03

C-WB2 Woodbine Beach Beach Core 2015 Jun 43.666 -79.299 0 50 0.05

*Abbreviations: Hb: harbour; Rv: river; Cr: creek; STP: sewage treatment plant outfall

Table 2. A summary of average microplastics contamination in various marine and freshwater sediments, worldwide, is given. Microplastics contamination is reported as average particle abundance per kilogram dry sediment, $N \text{ kg}^{-1}$ (dw).

Study	Study Area	Depositional Environment	Avg. $N \text{ kg}^{-1}$ (dw)
Turra et al., 2014	Sao Paulo, Brazil	Marine Beach	0.1 ^b
Dekiff et al., 2014	Germany	Marine Beach	2 ^a
VanCauwenberghe et al., 2015	Belgium	Marine Beach	6
Browne et al., 2010	UK	Marine Beach	~60 ^{a,b}
Claessens et al., 2011	Belgium	Marine Beach	95 ^a
This study	Ontario, Canada	Lacustrine Beach	140
Costa et al., 2010	Brazil	Marine Beach	310 ^{a,b}
Mathalon and Hill, 2014	Halifax, Canada	Marine Beach	5000
Claessens et al., 2011	Belgium	Marine Harbour	165 ^a
Naidoo et al., 2015	Durban, S. Africa	Marine Harbour	1165 ^{b,c}
Vianello et al., 2013	Venice, Italy	Marine Lagoon	1500 ^a
Frias et al., 2016	Portugal	Marine Nearshore	55
Claessens et al., 2011	Belgium	Marine Nearshore	90 ^a
This study	Ontario, Canada	Lacustrine Nearshore	980
Corcoran et al., 2015	Ontario, Canada	Lacustrine Offshore	352
This study	Ontario, Canada	Tributary	760 ^d

^a Modified from Van Cauwenberghe et al. (2015) Table 1

^b Using an average sediment density of 1600 kg m^{-3} (Fettweis et al., 2007)

^c Using a 1.25 average wet/dry ratio (Van Cauwenberghe et al., 2015)

^d Not including site P-EC2.

Figures

Figure 1. Sampling sites by depositional environment and instrument type for which microplastics in sediments in Lake Ontario and the St. Lawrence River were analyzed. Watershed boundaries indicate the regions that drain directly into Lake Ontario and the St. Lawrence River.

Figure 2. Microplastics abundance normalized to particles $N \text{ kg}^{-1}$ sediment (dry weight, dw), for 50 study sites in Lake Ontario. The inset shows the Greater Toronto Area in detail.

Figure 3. (a) Microplastics ($< 2 \text{ mm}$) abundance, $N \text{ kg}^{-1}$ sediment (dw), for beach, tributary and nearshore sediments of Lake Ontario averaged (error bars indicate standard deviation) across depositional environment. (b) The relative abundance of fibers, fragments and beads averaged across beach, tributary and nearshore samples. Samples collected from the St. Lawrence River are considered as nearshore samples.

Figure 4. (a) Microplastics (< 2 mm) abundance, $N g^{-1}$ sediment (dw), plotted against cumulative sediment mass ($g cm^{-2}$), dw, for nearshore sediments of Humber Bay and Toronto Harbour as sampled by gravity core. Cumulative sediment mass (CSM) is a function of the core radius (r),

sediment mass of the core interval (m), and depth of interval in cm (d): $CMS = \sum_{i=1}^d \left(\frac{m_d}{\pi r^2} \right)$. (b)

Microplastics (< 2 mm) abundance, $N kg^{-1}$ sediment (dw), averaged (error bars indicate standard deviation) for tributaries draining into Lake Ontario including Don River (DR), Humber River (HR), Etobicoke Creek (EC) and Red Hill Creek (RC). In each tributary (except DR), two proximal sites were sampled. (c) Microplastics (< 2 mm) abundance, $N kg^{-1}$ sediment (dw), at Beachway (BW), Bronte (BB), Marie Curtis Park (MC), Sunnyside (SS) and Woodbine (WB) beaches. One proximal and one distal foreshore sample from each beach was collected along a transect perpendicular to the waterline. Proximal and distal sites are denoted with a '1' and '2', respectively, in the sample name as listed in Table 1.

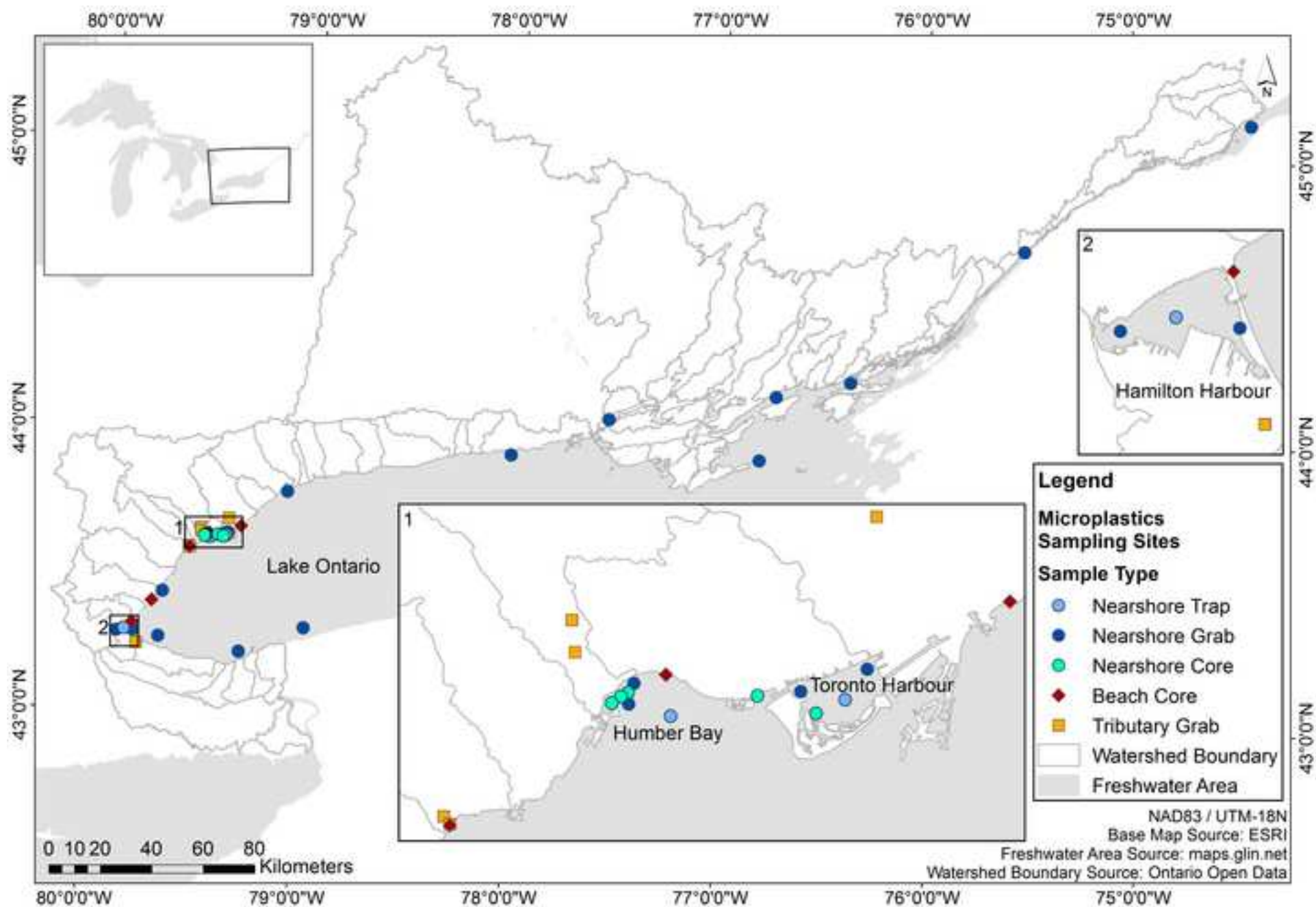
Figure 5. Examples of microplastics identified in sediment samples from tributaries, beaches and the nearshore lake bottom of Lake Ontario. All scale bars are 1 mm. (a) Macro- and microplastic fragments, fibers, foams and pellet isolated from the upper 10 cm of proximal foreshore sediments at Sunnyside Beach. (b) Microplastic fragments and fibers isolated from nearshore sediment in Toronto Harbour as collected in a sediment trap. (c) Microplastics found in grab sediments (sample S-3027) in Humber Bay. Fragments include hexagonal glitter (N=4) and whole and fragmented PSS beads (N=4), on right, among other fragments of unidentified source. (d) Microplastics from Etobicoke Creek. Long helical fragments may be derived from deflashing processes used for finishing injection moulded plastic products. (e) Examples of black, opaque fragments with rubber-like consistency found in Humber Bay. (f) Microplastic fragment exhibiting bulbous to wispy form and isolated from Toronto Harbour sample G-TH1 at a depth of 1-2 cm below the sediment surface.

Figure 6. Synthetic polymers and compounds which were identified in the FT-Raman spectroscopic analysis of particles isolated from Lake Ontario nearshore and tributary sediments. Of ninety particles analyzed, 60 were plastic in composition, 3 were non-plastics (e.g. quartz, calcium carbonate), 5 were plastic-associated compounds (e.g. phthalates, toluidine colourant), and 22 could not be identified.

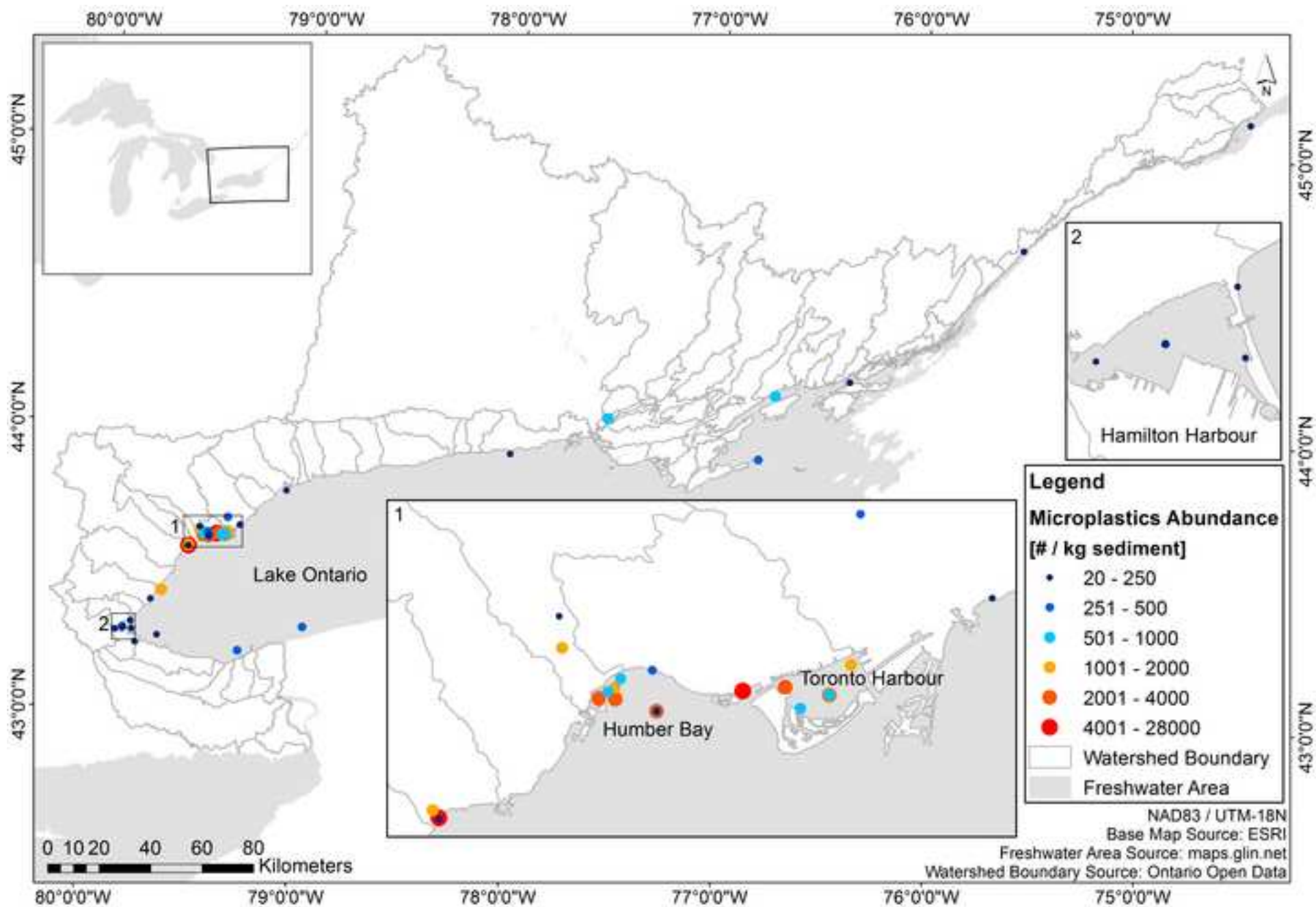
Figure 7. Human population map for watersheds draining into Lake Ontario and the St. Lawrence River. Locations of wastewater treatment plants on the shoreline of the study are included.

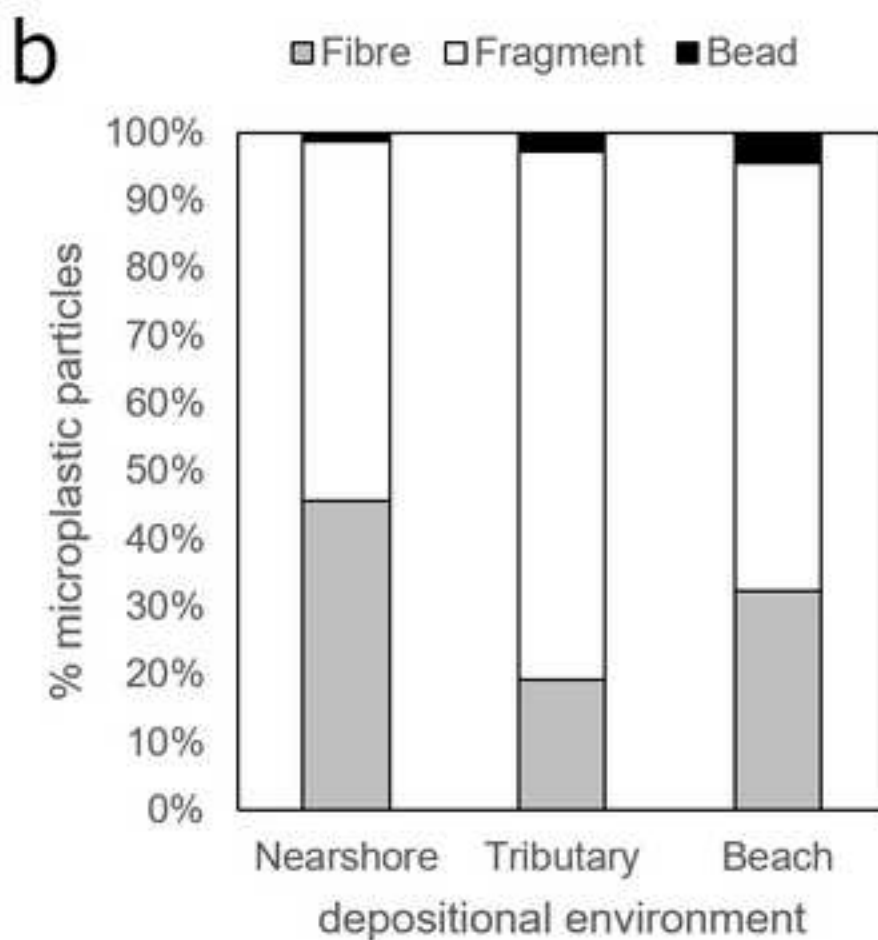
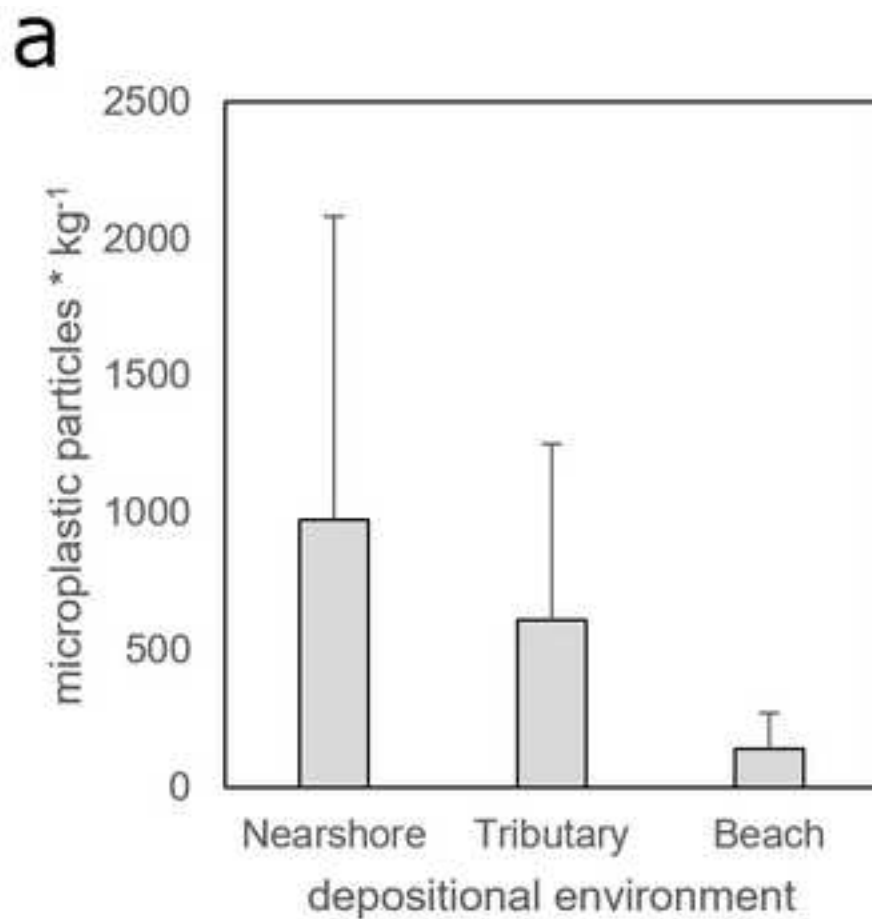
Figure 8. Locations of plastics-related manufacturing, distributing and service facilities within the watersheds draining into Lake Ontario and the St. Lawrence River.

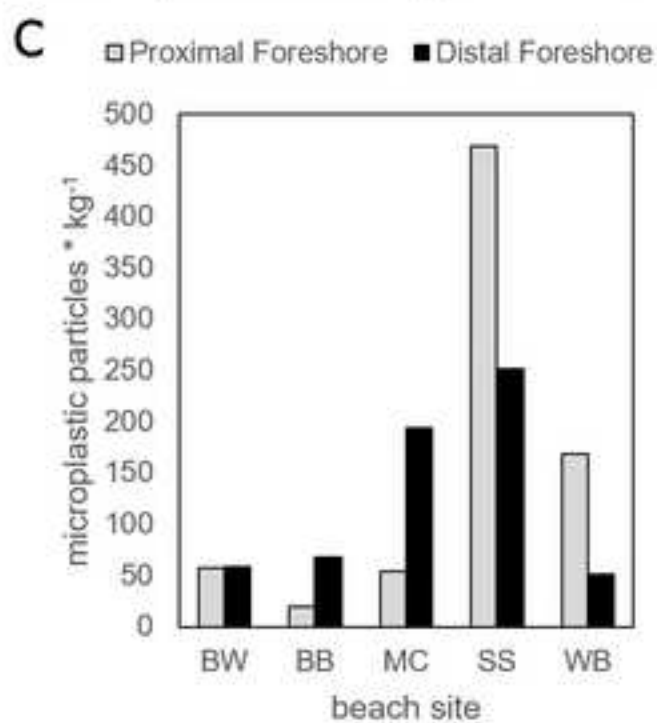
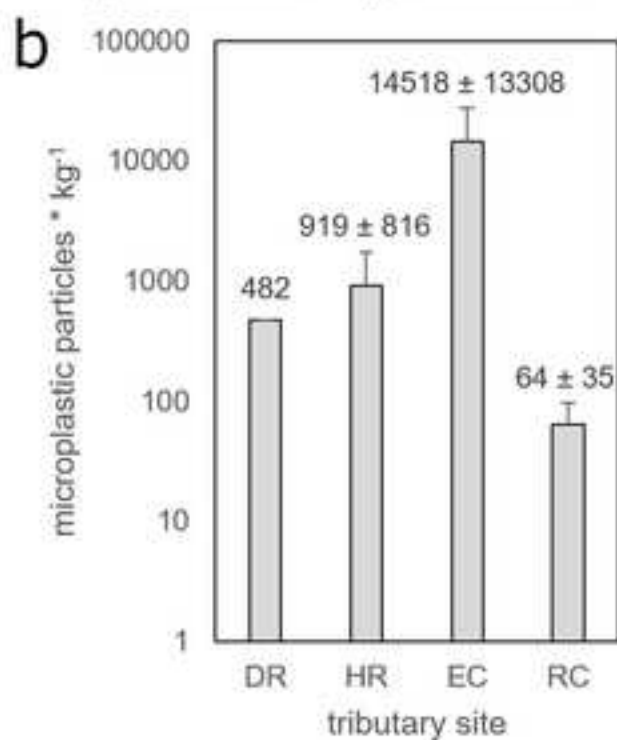
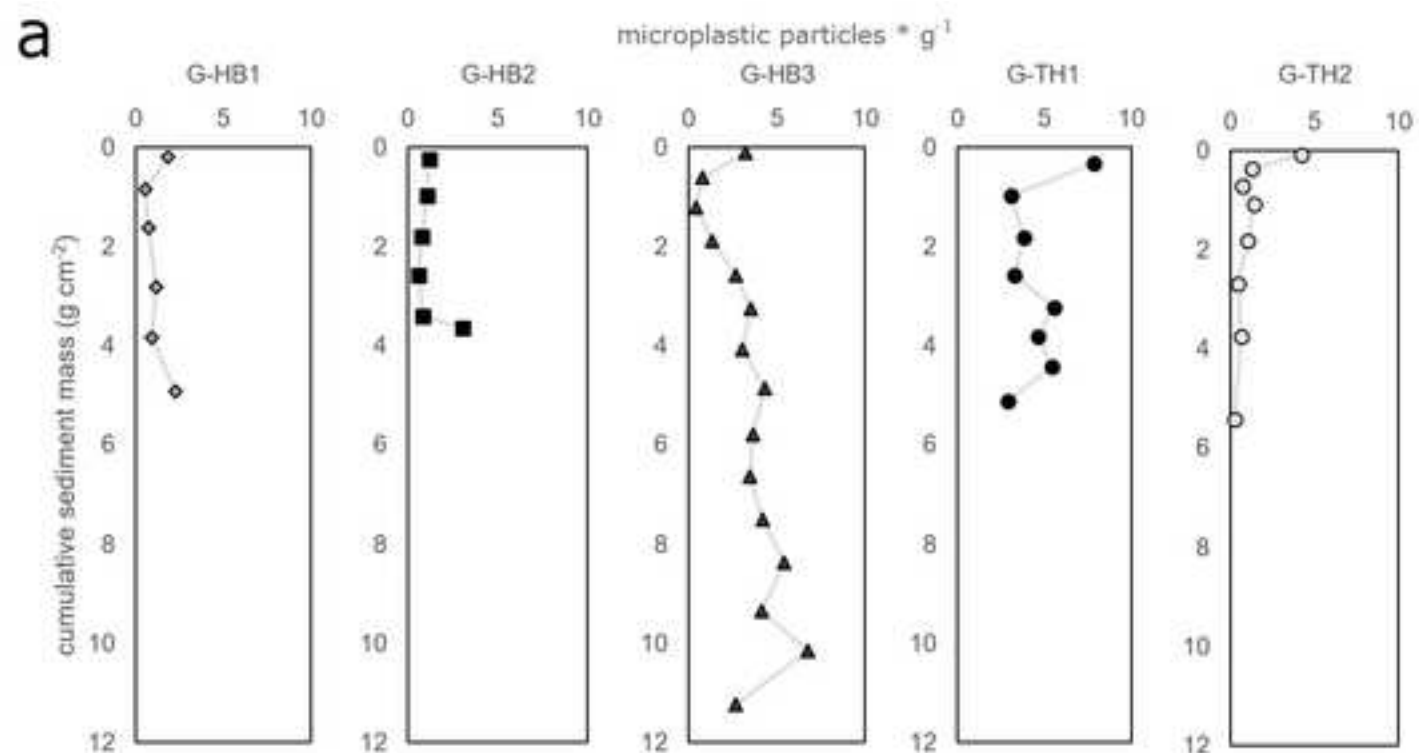
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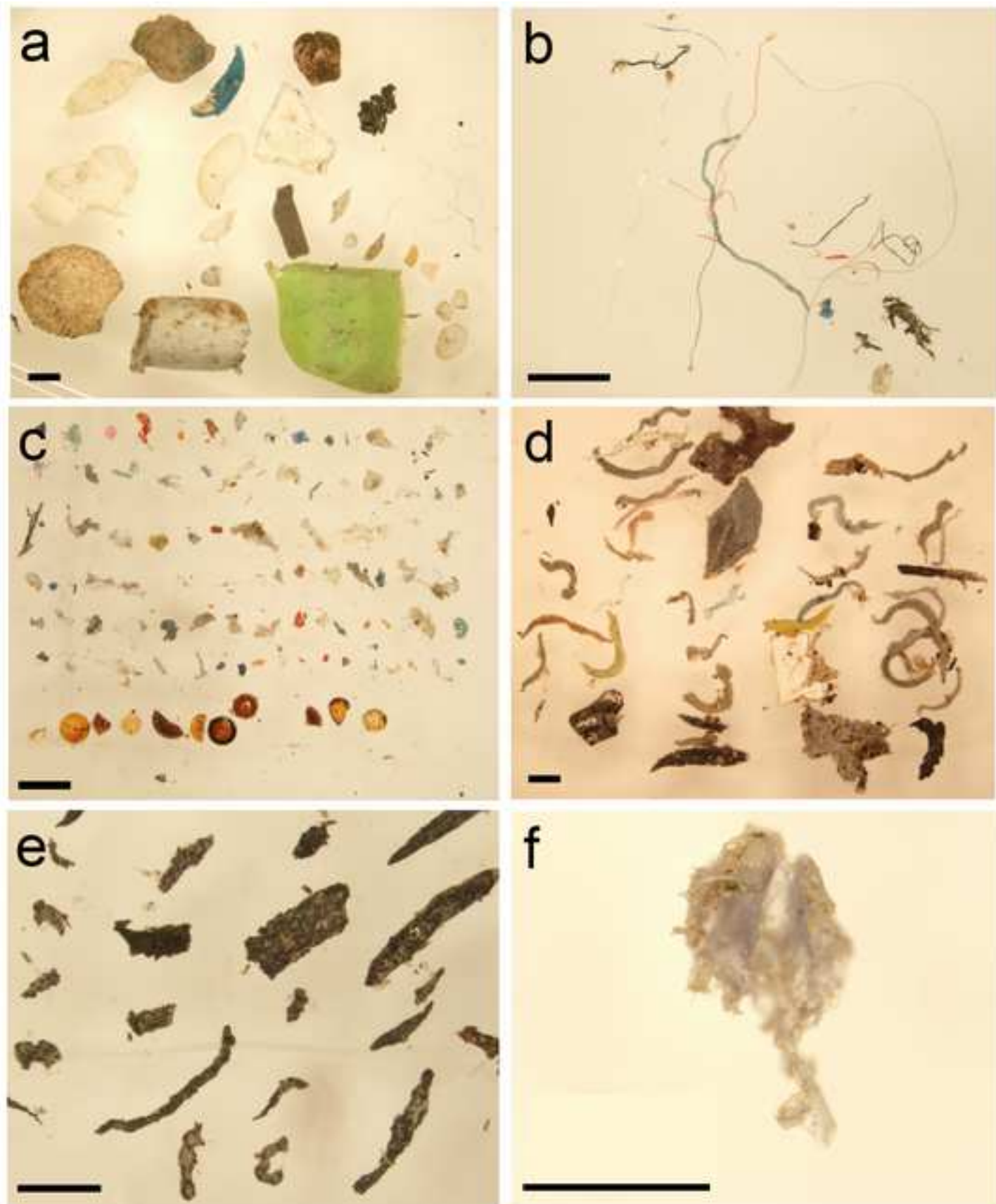


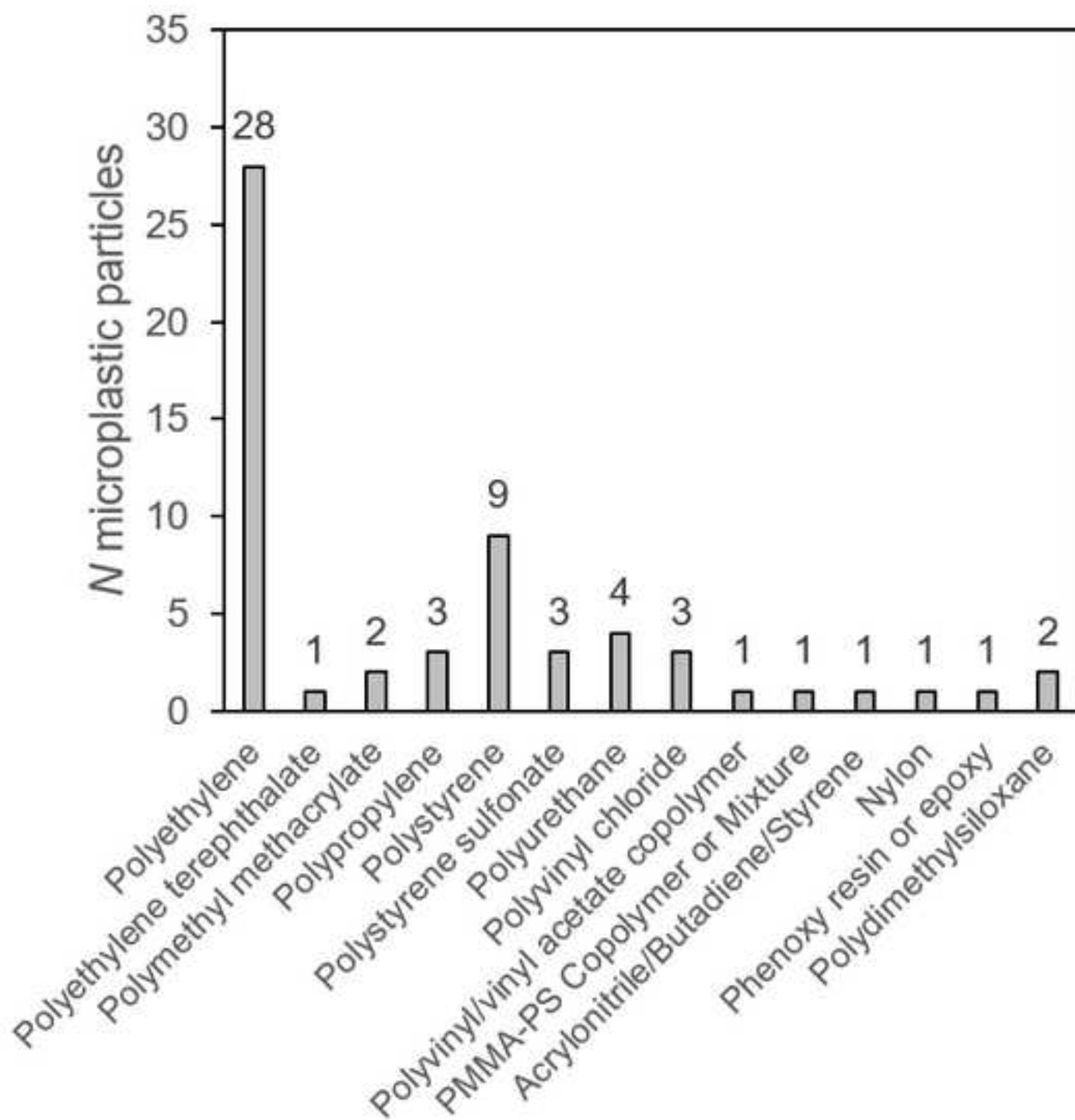
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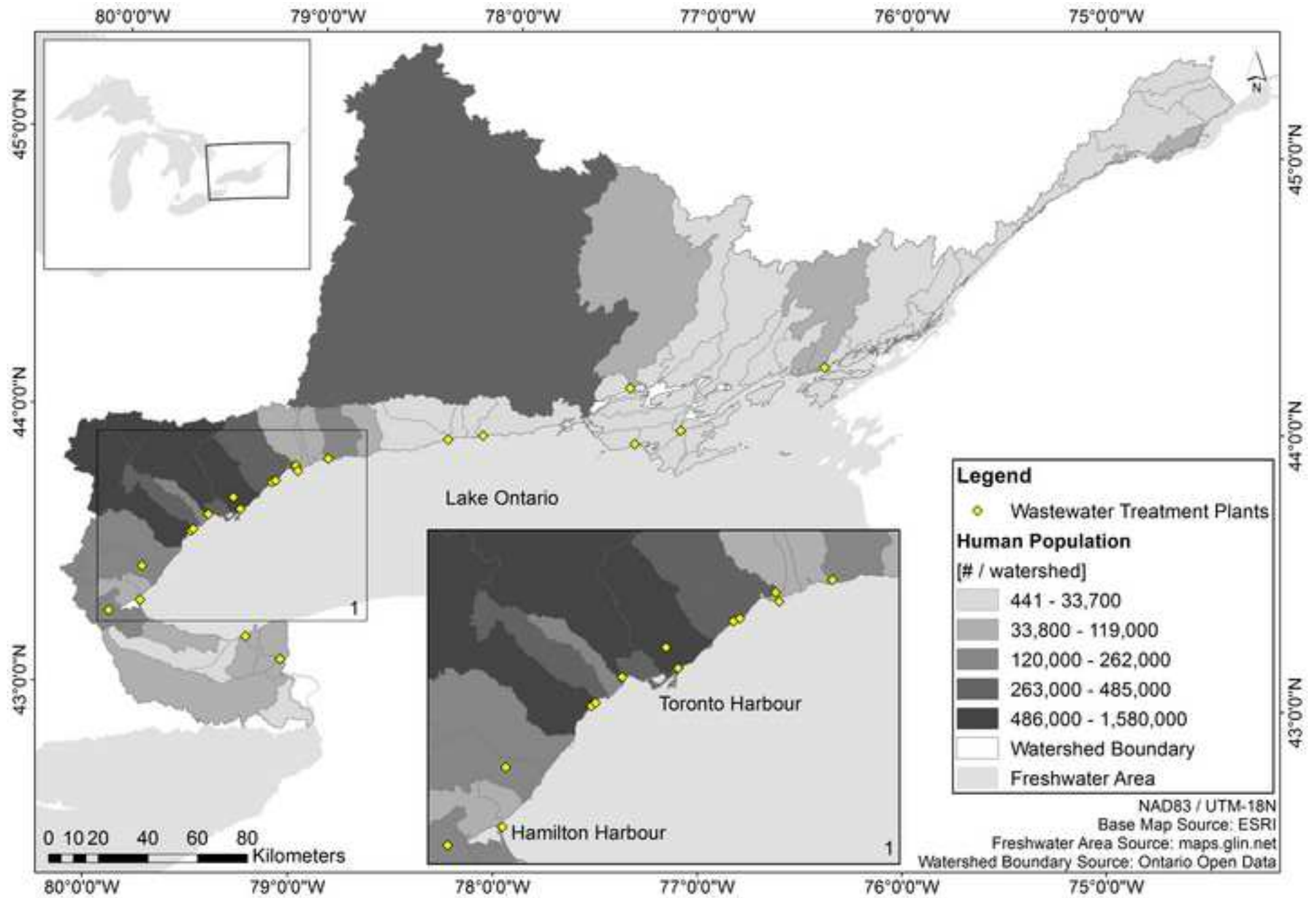






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