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Distribution, abundance and spatial variability of microplastic pollution on the surface of Lake Superior

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ABSTRACT

In 2014, 94 paired neuston net samples (0.5 mm mesh) were collected from the surface waters of Lake Superior. These samples comprise the most comprehensive surface water survey for microplastics of any of the Great Lakes to date, and the first to employ double net trawls. Microplastic abundance estimates showed wide variability, ranging between 4000 to more than 100,000 particles/km^2 with most locations having abundances between 20,000 to 50,000 particles/km^2. The average abundance in Lake Superior was ~30,000 particles/km^2 which was similar to previous estimates within this Laurentian Great Lake and suggests a total count of more than 2.4 billion (1.7 to 3.3 billion, 95% confidence interval) particles across the lake's surface. Distributions of plastic particles, characterized by size fraction and type, differed between nearshore and offshore samples, and between samples collected in the eastern versus western portion of the lake. Most of the particles found were fibers (67%), and most (62%) were contained in the smallest classified size fraction (0.50–1 mm). The most common type of polymer found was polyethylene (51%), followed by polypropylene (19%). This is consistent with global plastics production and results obtained from other studies. No statistically significant difference was detected between the paired net samples, indicating that single net sampling should produce a representative estimate of microplastic particle abundance and distribution within a body of water.

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Introduction

Plastic pollution first appeared in noticeable quantities in aquatic environments during the 1970's (Carpenter and Smith, 1972). It was initially thought that this pollution was not harmful and was a natural consequence of increasing industrial production since the 1950's (vom Saal et al., 2008). Plastic is currently the largest contributor to marine pollution (vom Saal et al., 2008; Lebreton et al., 2017; Sarijan et al., 2021). Millions of tons of plastics enter the world's waters with the United States and Asia being the largest contributors (NOAA, 2014; Jambeck et al., 2015; Lebreton et al., 2017; Law et al., 2020). With plastic now being found in the air, oceans, soils, sediments, and surface waters, the prevalence of plastics in our everyday life is at an all-time high (Alimi et al., 2018).

Large plastic debris, such as derelict fishing gear, was initially the main concern of scientists assessing plastic pollution. However, fishing gear lost at sea accounts for only a small percentage of aquatic plastic pollution (Andrady, 2011). Microplastic particles have been a growing concern since the early 1990s (Frias et al., 2014). Microplastics fall under two categories depending on their origin. Primary microplastics are plastic particles that were created within the defined size range of 1 μm–5 mm (Talvitie et al., 2017). This includes scrubbers in cleaning and cosmetic products, pellets used in plastic feedstock or productions (Eerkes-Medrano et al., 2015) and textile fibers (Talvitie et al., 2017). Secondary microplastics are particles that have fragmented from larger parent pieces (Talvitie et al., 2017). This includes fibers and fragments from items such as fishing nets, line fibers, films, raw materials, consumer products, household products, and degradable plastics that are designed to fragment in the environment (Eerkes-Medrano et al., 2015). Regardless of their origin or use, dominant polymers used in plastic production tend to be less dense than water (e.g., polyethylene and polypropylene with densities of ~0.91–0.95 and 0.90–0.92, respectively) and thus plastic particles tend to float within aqueous systems (GESAMP, 2019). Biofilms can develop on these surface floating particles, nonetheless, causing the density of the particle to increase and thus sink (Corcoran et al., 2015).

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Most research on microplastic pollution has occurred in marine ecosystems, but microplastic pollution is also widespread throughout freshwater lakes, rivers, estuaries, and other bodies of water around the globe (Free et al., 2014; Lebreton et al., 2017). Less is known about the abundance, distribution, transportation, and behavior of microplastic particles in freshwater as compared to marine systems (Wagner et al., 2014; Sarijan et al., 2021). Similarities, however, do exist between marine and lacustrine systems in terms of transportation, distribution, and occurrence of microplastics in aquatic animals (Zbyszewski and Corcoran, 2011; Dris et al., 2015; Eerkes-Medrano et al., 2015; Sarijian et al., 2021).

Average microplastic concentrations reported in the Laurentian Great Lakes (0.04 items/m^2, Driedger et al., 2015) are as high as or greater than that reported from some oceanic gyres (South Pacific Gyre, 0.03 items/m^2, Eriksen et al., 2013; North Atlantic Gyre 0.03 items/m^2, Law et al. 2010; North Pacific Gyre 0.33 items/m^2, Moore et al., 2001). These findings lead to increased scrutiny to determine the source, abundance, and impact of microplastic pollution in the Great Lakes. Most of this research has focused on the more populated and industrialized lakes, Lakes Erie, Michigan and Ontario (Eriksen et al., 2013; Corcoran et al., 2015; Baldwin et al. 2016; Mason et al., 2016; Cable et al., 2017; Grbic et al., 2020; Mason et al., 2020). Lake Superior, the largest of the five lakes, is often considered to be relatively pristine due to its low population density, largely forested versus urban or industrial land-use (Baldwin et al., 2016), and its location at the head of the Great Lakes drainage basin. Though some microplastic sampling was reported in the eastern part of the basin by Eriksen et al. (2013) and in the western basin by Hendrickson et al. (2018) and Minor et al. (2020), a comprehensive study of the presence of microplastics in Lake Superior has not been completed. Here we report on the first full lake study of microplastics in Lake Superior. Additionally, in this study, side-by-side, paired nets were used to determine if single net sampling produces a representative estimate of microplastic particle abundance and distribution within a large body of water.

Materials and methods

Sampling sites

Lake Superior sample sites (Fig. 1) were selected by the U.S. Geological Survey (USGS). Nearshore sites (Fig. 1; inside the grey dotted line) were originally identified in 1978 (USA) and 1989 (Canada) to assess nearshore fish communities in water <100 m deep, while offshore sample sites were selected in 2011 to assess offshore fish populations in water depths >100 m (Rosinski et al., 2020). Surface water sampling for floating plastic particles was conducted in conjunction with an annual fisheries assessment conducted using the USGS Research Vessel KIYI from May to July 2014.

Sampling methods

Surface water samples were collected using a paired 1 × 1 m^2, 0.500 mm-mesh, three-meter long, neuston nets (Sea-Gear Model 9450-2.0, http://www.sea-gear.net/). Neuston nets were originally designed to collect neuston, such as zooplankton and larval fish that live within the upper 10 cm of the sea surface, but have recently found additional use in sampling microplastics within this same few centimeters of water surfaces. The paired nets were positioned side-by-side and towed at a depth of a half meter (half in and half out of the water) to reduce the likelihood of waves washing over the top of the net. The nets were deployed near mid-ship using the ship's crane and were towed ~1–2 m away from the port-side. As winds can lead to wave action that entrain neuston and surface floating particles to greater depths, the sea state for all sample sites was fairly calm with all wave heights <0.9 m, and most<0.3 m. Trawls were made at ~4 km/h (range = 3.2–4.8 km/h) for 10 min, with an average trawl distance (tow length) of 0.7 km (range = 0.6–0.8 km), at 94 locations across Lake Superior (Fig. 1).

Material collected from each net was rinsed into a labelled glass jar and preserved with 90% ethanol. To prevent confusion between the paired samples, the net closer to the boat was labelled Net A and the net further from the boat was labelled Net B. Samples obtained from Net A were given even identification numbers, and samples from Net B were given odd identification numbers.

Sample processing

The samples were processed using a modified National Oceanic and Atmospheric Administration marine debris protocol (Masura et al., 2015), briefly described here (for details see Cox, 2018). Samples were first emptied into a set of two stacked stainless-steel sieves (0.355 mm and 4.75 mm). Solids within the largest size fraction (≥4.75 mm) were manually sorted to remove visible plastic debris from organic material. To collect any microplastics initially stuck to the organic material, all material was rinsed with deionized water, with the rinse water running through the sieves, before being discarded. The solids in the smaller sieve (0.355 mm) were then subjected to a wet peroxide oxidation (WPO), which digests labile organic material using 30% hydrogen peroxide in the presence of an iron (II) catalyst. No external heat was added to accelerate the reaction; however, it is an exothermic process. No density separation process was employed in the processing of samples.

After the wet peroxide oxidation, each sample was sieved through a set of two stacked stainless-steel sieves (0.355 mm and 1.00 mm) separating particles by size fraction. All particulates within each sieve were transferred to individual glass Petri dishes using deionized water for visual analysis. Using a dissection microscope (Leica EZ4 HD, 40×), all microplastic particles within each size classification (0.500–0.999 mm, 1.00–4.749 mm, and ≥4.75 mm) were removed, enumerated, categorized by morphology as fragment, pellet, line/fiber, film, or foam (Mason et al., 2016; GESAMP, 2019) and archived for later spectroscopic analysis. Note that despite the smallest sieve-size being 0.355 mm, the lower end of the smallest size faction is 0.500 mm due to the mesh size of the trawl net being slightly larger than the sieve used during sample processing.

Quality assurance and quality control

To reduce potential contamination throughout the sample processing from external sources, such as airborne fibers, work occurred in a laminar airflow cabinet and the workspace was wiped down every week. All glassware was covered with a watch glass when not in use and washed thoroughly between trials. Cotton lab coats and sterile nitrile powder free exam gloves were worn throughout the methodological procedure.

To account for possible laboratory contamination that could be coming from atmospheric deposition, the chemicals used, the glassware or other aspects of the testing environment, laboratory blanks containing deionized water (used to wash all glassware) were processed in a manner identical to the samples themselves. Particle counts within samples were reduced based upon the average counts across all lab blanks (n = 7; average = 5.6 particles; all fibers).

It is important to note that the WPO method is stated (Masura et al., 2015; Tagg et al., 2017) to have negligible impact upon the most common plastics. Nevertheless, as demonstrated by Munno et al. (2018), the elevated temperatures that occur as a result of the WPO method, even in the absence of added heat, can lead to

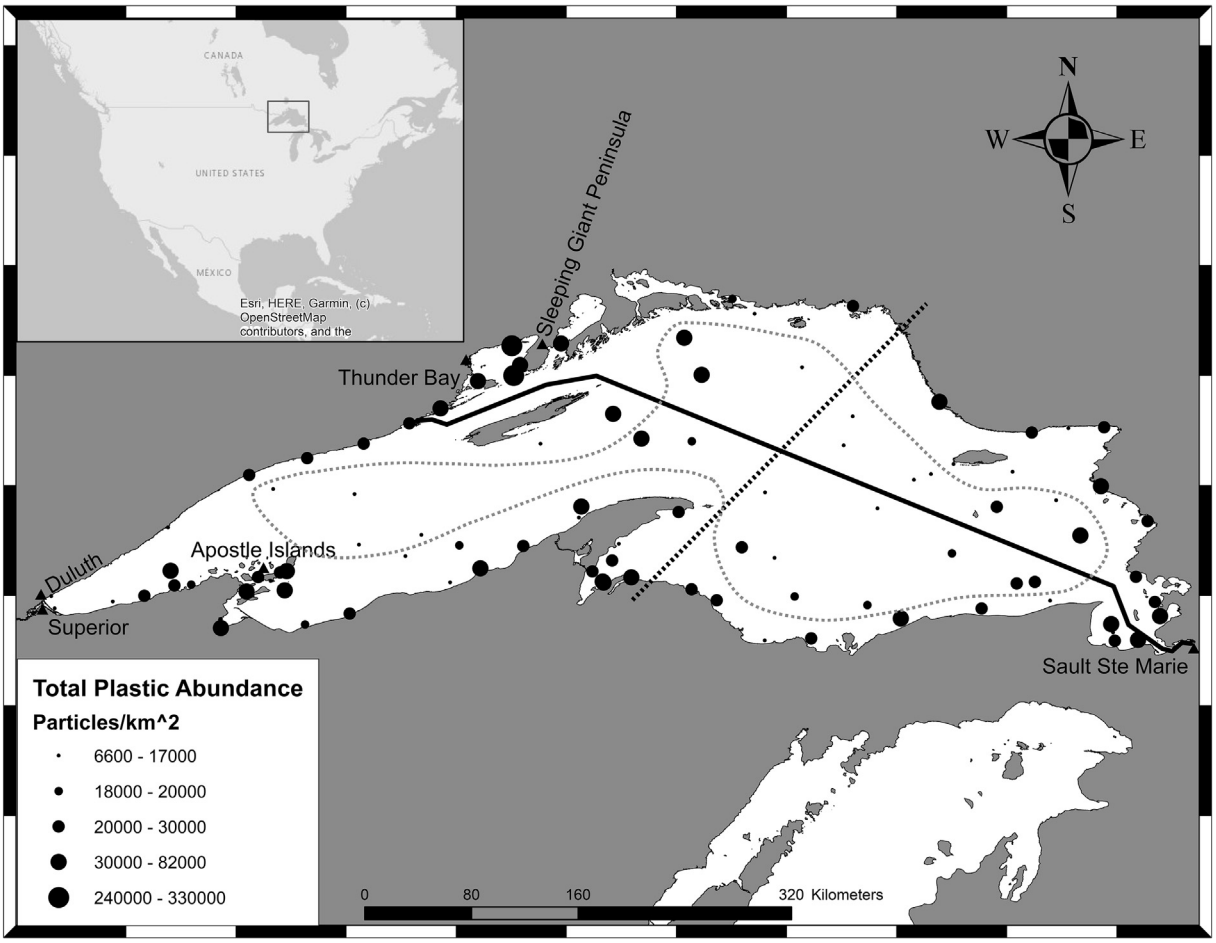

Fig. 1. Average surface water plastic abundance (particles/km^2) between two paired samples collected at 94 locations across Lake Superior during summer 2014. For analytical purposes, sampling regions were divided: east or west of the Keweenaw Peninsula (black dotted line), north–south of the USA/Canada border (black solid line), and nearshore-offshore, based on the approximate location of the 100 m bathymetric depth zone around the lake (grey dotted line).

the melting and loss of some spherical plastic, particularly those 'microbeads' which have wax character. As the temperature of the reaction was not monitored throughout the process but showed characteristic boiling behavior of temperatures in excess of 75 °C, it is likely that some microbeads/pellets were 'lost' via this chemical processing and thus the numbers reported here should be considered conservative.

Spectroscopic analysis

Due to the time intensive nature of Fourier Transform Infrared (FTIR) spectroscopic analysis and the quantities of retrieved particles, not all particles from all samples could be analyzed. To analyze a representative number of particles, approximately 10% of the total particles counted were targeted for FTIR analysis. To reach a total of 10% analysis, samples with high counts of microplastics (Electronic Supplementary Material (ESM) Table S1, starred samples) were selected and, from these samples, no fewer than 10, and no more than 30 particles (across different sizes and morphologies) were selected and analyzed. The rest were returned to their respective labelled glass vial.

In preparation for FTIR analysis, sample contents were rinsed from their glass vials and emptied into labelled, clean, dry petri dishes (separated by size fraction) and placed in a Thermo Scientific Heratherm oven at 50 °C until the petri dish and its contents were dry. Individual particles were then removed from the petri dish and placed on the Attenuated Total Reflectance (ATR) FTIR (PerkinElmer Spectrum Two ATR; 450 cm^{-1} to 4000 cm^{-1}, 64 scans, 4 cm^{-1} resolution) for analysis. Polymer identification was made by comparing sample spectra to an internal manufacturer provided spectrum library, requiring a minimum 70% match for acceptance.

Data analysis

To determine if the spatial heterogeneity between the side-by-side nets was statistically significant a paired T-test was performed using the total number of particles found in each individual net (i.e., not separated by size or morphology). Because the raw data were not normally distributed, but were skewed right, a log (X + 1) transformation was performed. The log-transformed data showed a normal distribution, allowing a paired T-test to be performed (SYSTAT 13 for Windows, version no. 13.1).

Plastic abundance per square kilometer of lake surface was calculated based on the surface area sampled (tow length × trawl net width) and the number of plastic particles in each sample. Relative comparisons in plastic particle abundances across the lake were made in relationship to predominant currents, cities, north–south using the USA/Canada border (Fig. 1, solid line), east–west (Fig. 1, black dotted line to the east of the Keweenaw Peninsula); and nearshore-offshore, based on the approximate location of the 100 m bathymetric depth zone around the lake (Fig. 1, grey dotted line).

Results and discussion

During the summer of 2014, 94 paired surface water samples were collected across Lake Superior yielding a total of 187 individual samples as one (sample ID 679) was broken in transit (ESM Table S1). Previous to this study, five samples each had been in collected in the eastern (Eriksen et al., 2013) and western portions (Minor et al., 2020) of the lake in 2012 and 2018, respectively, one sample (in triplicate) in eastern Lake Superior in 2014 (Cable et al., 2017), and twelve samples in western Lake Superior in 2016–2017 (Hendrickson et al., 2018). Therefore, this study represents the most intensive survey of surface floating microplastics in Lake Superior to-date. Plastic particles were found in all 187 samples, yielding a total of 3887 particles.

Differences between nets

There was no difference in mean plastic abundances between the two individual samples simultaneously collected at each site (paired *t*-test, T = −0.648, p = 0.519, N = 93 paired samples; data log transformed), however the variation in plastic abundance among sites was higher for the net positioned closer to the boat (Net A; Table 1). This was likely caused by its proximity to the boat that, depending on the wind direction, may have increased or decreased the volume of water passing through the net and illustrates the need to take such physics into consideration. These results indicate single net sampling methods can produce representative samples.

Plastic particle abundance

The abundance of microplastic particles was relatively homogeneous across the surface of Lake Superior (Fig. 1). Of the 94 locations sampled, 51 (54%) had an abundance between 20,001–50,000 count/km^2 and another 34 (36%) locations had an abundance between 10,001–20,000 count/km^2 (Table S1). This accounted for 90% of all sample locations throughout the lake, indicating that microplastic abundance was similar regardless of where the samples were obtained.

Of the 94 locations sampled, two sites had an average abundance across the paired nets higher than 100,000 particles/km^2 (ESM Table S1). These two samples were located less than 20 km from Thunder Bay, ON, which is the largest city (by population) on the shores of Lake Superior. It should be noted that while Thunder Bay, ON, Canada, has a total population (~108,000) greater than that of either Duluth, MN, USA (~85,000) or Superior, WI, USA (~26,000) individually, the combined Twin Ports (as they are called) of Duluth-Superior do exceed that of Thunder Bay, especially when considering the surrounding area, according to the 2020 USA population census. We do not, however, have samples obtained as close to the Twin Ports as we do Thunder Bay. Given the larger population center of the Twin Ports, it might be expected that similar results would be obtained from the surface waters of the Duluth-Superior Harbor as from Thunder Bay.

Table 1
Descriptive statistics on log-transformed numbers of plastic particles captured by paired nets (n = 93) across the surface of Lake Superior. Net A was closer to the boat than Net B.

	Net A	Net B
Minimum	7.99	8.53
Maximum	12.9	12.5
Median	10.0	9.92
Mean	9.93	9.99
Standard Deviation	0.85	0.68

Other areas of high lake surface particle abundance were near Sault Ste Marie, Ontario, Canada, at the east end of the lake where Lake Superior enters the St. Marys River, and surrounding the Apostle Islands, USA near the west end of the lake (Fig. 1). Six locations contained fewer than 10,000 count/km^2 and were mostly located offshore (ESM Fig. 1; Table S1).

Across the lake surface, microplastic abundance averaged ~30,000 particles/km^2 (95% confidence interval, ~21,000 to ~39,000 particles/km^2). Based on this average and the surface area of Lake Superior (82,100 km^2), this suggests a total count of more than 2.5 (1.7–3.2) billion plastic particles across the total surface area of the lake. While higher than expected given the low human population density within the Lake Superior watershed, a combination of Lake Superior's long residence time (199 years, Quinn, 1992), cold temperatures, and relatively low microorganism concentrations may explain these relatively high abundances of surface-floating plastic within Lake Superior as compared to the other Laurentian Great Lakes (described below). Many plastics are known to have limited biodegradability, favoring mechanical degradation, which simply forms smaller plastic particles, and much slower (with regard to mineralization) photooxidative pathways (Singh and Sharma, 2008). The on-average colder water temperatures of Lake Superior, as compared to the other Laurentian Great Lakes (given its enormous size and northern location), lead to reduced microorganism concentrations within the lake, which are necessary for biodegradation, as well as reduced biofilm growth that would lead to an increase in particle density and possible sedimentation.

Plastic particle distribution

The majority of microplastic particles collected were contained in the smallest (0.500–0.999) mm size fraction (62%) with fibers/-lines being the most common morphology (67%). Fragments (23%) were the next most common morphology, followed by films (9%), with foams and pellets each accounting for only 1% of the total (Table 2). Of the pellets, only a few matched the size (<1 mm), shape and color of microbeads commonly associated with personal care products (Chang, 2015), with most, especially within the larger size fraction (>1mm), seeming to be pre-production plastic pellets, commonly referred to as nurdles.

Areas of low and high calculated abundances may be attributed to currents and water circulation patterns (Beletsky et al., 1999; Bennington et al., 2010), as well as proximity to large population centers, which are sources of microplastic pollution (Eriksen et al., 2013; Hoffman and Hittinger, 2017; Mason et al., 2020). Surface currents in Lake Superior (down to a depth of 15 m) during the summer months would account for the accumulations around the Apostle Islands, as well as near the St. Marys River (Beletsky et al., 1999; Bennington et al., 2010). The high accumulations near Thunder Bay, Ontario were found in the bay itself and would be less influenced by lake currents. Currents outside of Thunder Bay travel southwest towards Duluth but do not enter the bay because of the Sleeping Giant Peninsula which juts out and traps water and microplastics, possibly preventing large quantities from being transported elsewhere by currents (Beletsky et al., 1999; Bennington et al., 2010). General circulation patterns are counter-clockwise (cyclonic) and run along the coastline of the lake driving particles toward the outflow at the southeastern end of the lake (Beletsky et al., 1999; Bennington et al., 2010). However, at the mouth of the St. Marys River the flow is clockwise (anti-cyclonic) (Bennington et al., 2010), which could act to trap particles within the southeastern bay leading to the higher lake surface abundances within this region (Fig. 1).

Average particle abundance was higher in the: north (v. south), west (v. east), and nearshore (v. offshore) regions of the lake

Table 2
Plastic abundances (count/km^2) for 2014 Lake Superior averaged over all 187 samples, distinguished by size and particle type.

	0.500–0.999 mm	1.000–4.749 mm	>4.75 mm	% of total
Fragment	4189	2755	55	23%
Pellet	111	131	0	1%
Fiber/Line	13,556	6471	234	67%
Film	1006	1448	188	9%
Foam	125	149	16	1%
Total	18,987	10,961	493	
% of total	62%	36%	2%	

Table 3
Average plastic abundances (count/km^2) by size classifications for various location designations on Lake Superior. Designations are visualized in Fig. 1 and specified in Table 1.

	0.500–0.999 mm	1.000–4.749 mm	>4.75 mm	% of total
North (n = 59)	25,293 (61%)	18,483 (71%)	798 (69%)	65%
South (n = 128)	16,080 (39%)	7482 (29%)	353 (31%)	35%
East (n = 73)	16,655 (45%)	7589 (37%)	281 (31%)	42%
West (n = 114)	20,480 (55%)	13,107 (63%)	629 (69%)	58%
Near (n = 136)	21,417 (63%)	11,618 (56%)	582 (69%)	61%
Off (n = 51)	12,505 (37%)	9179 (44%)	259 (31%)	39%

(Table 3). As noted earlier, the two samples with the highest abundance were located less than 20 km from Thunder Bay. These samples, along with those from near Sault Ste Marie, Ontario, Canada, lead to the higher average within the northern region of the lake, despite other areas of population density (such as Duluth-Superior, Minnesota, USA) being located along the southern shores (Table 3). Similarly, the higher counts in the western samples may be explained by the high population centers on the western side of the lake, as both Thunder Bay and Duluth-Superior are located there. Given their proximity to sources (i.e., people), as well as the tendency for particles to accumulate through surface currents along coasts, nearshore samples contained higher abundances on average as compared to offshore samples (Table 3).

It should be noted that sampling occurred from May through August, a 4-month time period. As surface currents within the Great Lakes tend to change seasonally, this could influence the results obtained and discussed here. Within Lake Superior surface currents are driven by winds in the winter and lake temperature variations in the summer leading to a tendency to slow throughout these months and quicken during the winter (Beletsky et al., 1999; Bennington et al., 2010). These seasonal changes in the drivers of surface currents lead to variations in the seasonal gyres (Beletsky et al., 1999; Bennington et al., 2010). As particles are carried by surface currents, these seasonal changes can influence the spatial variations of microplastic abundance on temporal scales.

Polymer identification

A combined total of 3887 particles were identified and counted from the 187 samples in this study. Of this total, 408 individual particles (~10%), taken from the 28 samples with the highest abundances (Table S1), were acceptably analyzed using FTIR to confirm polymer identity (Table 4). The most common polymer identified was polyethylene, with a total of 204 particles, which accounted for 50% of all particles identified (Table 4). Polypropylene was the second most common polymer but was much less common than polyethylene, with a total of only 82 particles, or 20% (Table 4). The remaining particles were identified as polyester (7%), polyamide (7%), polystyrene (2%), and others (14%).

As polyethylene and polypropylene are the most produced polymers, representing ~30% and ~20% of the market, respectively ((PlasticsEurope, 2020)), it is not surprising that they would be the most prominent within these field samples. The low density of these two polymers (<1 g/mL) are likely also a factor in their prominence in surface water samples. These findings are consistent with other studies within the Laurentian Great Lakes (Mason et al., 2016; Mason et al., 2020).

On a more granular level, polymeric identity varied by particle morphology (Table 4). In total, 168 lines/fibers were analyzed using FTIR with polypropylene (25%) being slightly more prevalent as compared to polyethylene (23%). Polyester (17%) and nylon (15%) were also common. In contrast, fragments and films were more commonly composed of polyethylene (70% and 76% respectively). Both results are reflective of the most common applications for these polymers (GESAMP, 2019).

Comparison to previous great lakes' studies

To-date there have been six studies (prior to this one) within the open-waters of the Laurentian Great Lakes, of which four have included some sampling within Lake Superior (Table 5). Two of these studies (Eriksen et al., 2013; Cable et al., 2017) only sampled

Table 4
Summary of FTIR results on retrieved pelagic plastic particles. PE = polyethylene; PP = polypropylene; PEST = polyester; PA = polyamide; PS = polystyrene.

Particle Morphology	Number of Particles			Type of Polymer					
	Retrieved	Analyzed	%	PE	PP	PEST	PA	PS	Other
Fragments	891	149	17%	104 (70%)	26 (17%)	–	2 (1%)	–	17 (11%)
Pellets	30	6	20%	5 (83%)	1 (17%)	–	–	–	–
Fibers/Lines	2593	168	6%	39 (23%)	42 (25%)	28 (17%)	25 (15%)	–	34 (20%)
Films	336	74	22%	56 (76%)	13 (18%)	–	1 (1%)	–	4 (6%)
Foams	37	11	30%	–	–	–	–	11 (100%)	–
Total	3887	408	10%	204 (50%)	82 (20%	28 (7%)	28 (7%)	11 (3%)	55 (14%)

Table 5
Comparison of results of this study to previous Laurentian Great Lakes studies in percentages by size and particle type.

		Laurentian Great Lakes Sampled	Size Classification			Morphology				
	n		0.35–0.99 mm	1.00–4.74 mm	>4.75 mm	Fragment	Pellet	Fiber/Line	Film	Foam
Eriksen et al., 2013	21	Superior (5), Huron (8), Erie (8)	81%	17%	2%	42%	48%	<1%	1%	8%
Mason et al., 2016	59	Michigan	59%	32%	9%	79%	4%	14%	2%	1%
Cable et al., 2017	38	Superior (1), Huron (9), Erie (28)	93%[a]	6%	1%	97%	1%[b]	<1%[c]	<1%	1%
Hendrickson et al., 2018[d]	12	Superior	n/a	n/a	n/a	38%	2%	39%	21%	1%
Mason et al., 2020	39	Erie (17), Ontario (22)	73%	24%	3%	63%	26%	4%	2%	6%
Minor et al., 2020[d]	5	Superior	n/a	n/a	n/a	12%	0%	70%	14%	4%
This study	187	Superior	62%[e]	36%	2%	23%	1%	67%	9%	1%

[a] The lower limit within the smallest size classification in this study was 106 μm.
[b] To be consist with previous studies, the pellet category here includes the combination of nurdles and spheres detailed in this study.
[c] Does not include fibers since, as the authors note, they could not be quantified with equal high confidence across all sizes.
[d] These studies did not reported size distributions. To be consistent with other studies the 'other' category reported in this study was included with fragments.
[e] The lower limit within the smallest size classification in this study was 500 μm.

within eastern Lake Superior, while the other two (Hendrickson et al., 2018; Minor et al., 2020) only sampled within western Lake Superior. Thus, this is the only study to-date to survey the entire Lake Superior surface and, with its 187 total samples, it is the most extensive survey of any of the Laurentian Great Lakes (Table 5).

Consistent with the results of other Great Lakes studies (that reported such data), the majority of (lake surface) retrieved microplastic particles were contained in the smallest size classification (Table 5). Given that one microplastic item can form thousands (if not millions) of microplastic particles through its (largely mechanical) degradation (Singh and Sharma, 2008), the consistency of these results is not surprising. Studies do show differences in particle morphology, both across the lakes and within Lake Superior itself (Table 5). Both studies which sampled only eastern Lake Superior (Eriksen et al., 2013; Cable et al., 2017) reported much lower counts of fibers than the present study, favoring fragments as the dominant morphology, though it should be noted that in both these studies the data presented were averaged across Lakes Erie, Huron, and Superior.

This study is more consistent with the more recent studies which focused solely on western Lake Superior (Hendrickson et al., 2018; Minor et al., 2020) and found fibers to be the dominant morphology (Table 5). Hendrickson et al. (2018) reasoned that the high count of fibers could indicate atmospheric deposition as a primary pathway for microplastics into Lake Superior. This hypothesis is consistent with the analysis of the ubiquity of fiber occurrence (regardless of land-use and flow conditions) within a study of 29 tributaries that feed into the Great Lakes (Baldwin et al., 2016), as well as their common occurrence within atmospheric samples (Brahney et al., 2021). Fibers were not as common in Lakes Superior, Erie, and Ontario as in Lake Michigan samples (Table 5). This difference could be owing to a combination of smaller lake surface areas and higher human population densities for Lakes Erie and Ontario, which would reduce the atmospheric deposition area and lead to higher emission rates of other particle morphologies increasing their ratios relative to those of fibers (Baldwin et al., 2016), as well as sampling and analytical differences among studies.

Our lake-wide average lake surface abundance of ~30,000 plastic particles/km^2 is consistent with that reported by Hendrickson et al. (2018). In comparison to other Great Lakes, a surface water survey of Lake Michigan showed an average of ~17,000 particles/km^2 (Mason et al., 2016), which is approximately 13,000 fewer particles/km^2 than Lake Superior, while Lakes Huron, Erie and Ontario were higher, averaging ~46,000, ~160,000 and ~230,000 particles/km^2, respectively (Earn et al., 2020). While the lower average of Lake Michigan might seem surprising given that it is technically downstream from Lake Superior, the flow of water (and thus the associated transport of microplastics) largely by-passes Lake Michigan, favoring a flow through Lake Huron into Lake Erie and, then, Lake Ontario. In this light, the consistent increase in average lake surface plastic abundance from Superior to Huron to Erie to Ontario may indicate that plastic loadings are both carried by the flow of water and enhanced by population centers along that flow. On the whole, these inter-lake variabilities seem to arise from a combination of lake surface areas, population densities within the watershed, and lake residence times.

Conclusions

This project represents the first comprehensive study of surface water microplastic pollution in Lake Superior and it is the only study completed in the Great Lakes using side by side nets for sample collection. The results show that there was no difference between the results obtained from Net A or Net B, other than slightly more variability in Net A samples nearer the boat. This indicates that single net surveys can produce a representative result and helps to provide validity to previous studies involving single neuston net sampling.

Lake surface plastic abundances estimated for Lake Superior in this study are consistent with estimates for other Laurentian Great Lakes. Lake Superior had higher average abundances than Lake Michigan despite its larger size and overall lower industrial and population densities, which may be attributed to Lake Superior's larger surface area, longer residence time, and perhaps sampling and analytical methodology differences among studies. Conversely, average abundances for Lake Superior were less than those previously estimated for Lakes Huron, Erie and Ontario, which are likely due to an additive effect of these lakes being downstream of Lake Superior and their higher population densities and industry. While Lake Michigan is technically downstream of Lake Superior, the dominate water flow from Lake Superior is through Lake Huron into Lake Erie.

Similar to other studies the highest concentration of particles was found in the smallest size fraction. The most common polymer type was polyethylene, which is similar to other studies, as well as global production patterns, which show that polyethylene is the most commonly produced polymer. Polypropylene was the second most abundant, which also agrees with other studies and global production statistics.

Given Lake Superior's large size, volume of water and long residence time, continued monitoring of microplastic pollution will be required to understand the overall health of the lake and the status of microplastic debris. This study provides a lake-wide baseline assessment for future studies. Many climatic conditions may affect microplastic abundance, distribution, and transport, including warming caused by climate change, seasonal changes in currents, as well as changes in population and industry along the shoreline.

Monitoring how these conditions impact not only the lake but pollution within the lake will increase the overall understanding of how microplastic pollution interacts with the surrounding ecosystem and may provide insight into management practices which can help protect the overall health of Lake Superior.

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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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jglr.2021.08.005.

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