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Microplastic contamination of sediments across and within three beaches in western Lake Superior

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ABSTRACT

Microplastic pollution of the environment is ubiquitous, but the processes by which microplastics accumulate within beach sediments are not yet well understood. We isolate microplastic pollution from the sediments at three western Lake Superior beaches. Samples of both surface and subsurface sediments are considered. We find that the average microplastic contamination is 65 microplastic particles kg^{-1} sediment across our sites with significant variability across beaches, but the microplastic composition is always dominated by polyester fibers. The variation across beaches does not seem to relate to the distance from suspected sources of microplastics to the lake. Within each beach, we find no significant variation of the mean microplastic concentration in the cross-shore direction or in the surface vs. subsurface sediments at the wrack line. We interpret this mean microplastic concentration in the sediment as a measure of the bulk microplastic concentration in the nearshore water at each beach. In subsurface sediments, we observe a significant difference in the variance of microplastic concentrations, and we attribute this variation to the intermittency of the extreme hydrodynamic conditions that deposit microplastics deeper into the sediment.

Introduction

Plastic production, use, and waste have been increasing significantly since the first synthetic polymers were created. Plastic particles have been found all over the world, with oceans and lakes acting as significant sinks (Jambeck et al., 2015; Law, 2017). Due to the ubiquity of plastics around the world, some have even proposed that plastics could be used as a marker for geologic time noting the significance of human pollution impact on the geologic record, calling this the Anthropocene Epoch (Corcoran et al., 2014).

Plastic pollution in the environment comes in many forms; plastics encompass many different polymers of various colors, densities, shapes, and sizes. When classifying plastic particles by size, we define microplastics to be particles that are less than 5 mm (Masura et al., 2015). Microplastics include 'primary microplastics,' which are particles that were processed in their original microscale size like the microbeads in cosmetic products, and 'secondary microplastics,' which are fragments or fibers that have broken down from macroplastics or larger microplastics (Barrows et al., 2018).

Microplastics are abundant in lakes and oceans around the world (Ballent et al., 2016; Isobe et al., 2014; Zhang et al., 2020). These plastic particles are transported from bodies of water to benthic sediments and beach sediments (Forsberg et al., 2020), although these processes and mechanisms are still not well understood. These microplastics in both the water and the sediments of many important water resources continue to be of great concern. We continue to see the expanding reach of microplastic pollution throughout the environment and adverse impacts on ecosystems and human health.

Plastic pollution in freshwater lakes, including the Laurentian Great Lakes, is a critical issue to understand. Such waters are an important freshwater resource and fishery ecosystem, and thus the impacts of plastic pollution are far reaching but not yet well understood. Microplastics have been shown to transport toxic chemicals into the environment (Mato et al., 2001) and these particles have been shown to collect persistent organic pollutants (Rios et al., 2007; Hirai et al., 2011). Due to their small size, they are easily ingested by small organisms which are at the lowest trophic levels of the aquatic food web. Ingestion of plastics can lead to chemical leaching from the plastics into an organism and is very likely transported to higher trophic levels (Andrady, 2011).

From a fate and transport perspective, the spread of microplastics has been declared one of the grand challenges in environmen-

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tal fluid mechanics (Dauxois et al., 2021). From numerous bodies of water, it has been hypothesized and demonstrated that microplastics can accumulate in benthic and beach sediments, which make such sediments a significant sink for microplastic particles (Critchell and Lambrechts, 2016; van Sebille et al., 2020; Kunz et al., 2016; Vermeiren et al., 2021; Bridson et al., 2020).

Current methods for understanding microplastic accumulation on beaches focus on sediment samples collected from the surface of the wrack line or high water mark - an area of significant debris accumulation (Sartain et al., 2018; Browne et al., 2010). This method works for collecting buoyant microplastic particles that are deposited at the maximum wave run-up, but may not account for plastics that are deposited on the beach due to other methods, specifically the turbulent mixing of sediments in the swash zone and burial of plastics in this region. Additionally, microplasitcs within surface sediments show the amount of plastics on the beach, but do not readily provide insight to the removal of plastics from the lacustrine environment as plastics within surface sediments could be easily re-suspended.

This lack of understanding of microplastic-beach interactions has led to an increase in studies of microplastic transport and influence at coasts (Browne et al., 2011; Critchell and Lambrechts, 2016; Isobe et al., 2014; Zhang et al., 2020). Specifically within the Laurentian Great Lakes, there have been a number of studies concerning microplastics within surface waters (Eriksen et al., 2013; Hendrickson et al., 2018; Mason et al., 2016; Mason et al., 2020) and benthic sediments (Lenaker et al., 2021) but only a few studies on the interaction of plastics with the beaches Dean et al., 2018; Ballent et al., 2016; Minor et al., 2020). Whitmire et al. (2017) examined microplastic pollution on beaches from various National Park units, including four beaches on Lake Superior, and found that the Apostle Island beaches on Lake Superior contained the most plastic per kilogram of sediment of all 35 locations studied around the United States. The authors attributed the high microplastic concentration partly due to close proximity to the Saint Louis River Estuary as a potential significant source for microplastic pollution into the Lake. Contrasting these results, Minor et al. (2020) found lower quantities of microplasitcs in the beach sands from Lake Superior in similar locations.

Motivated by the lack of mechanistic understanding of microplastic fate and transport in coastal areas, specifically the contamination of beach sediments in Lake Superior, we investigate here the microplastic contamination of three beaches in western Lake Superior. We choose three representative beaches in this area that are near potential microplastic sources (*e.g.*, a freshwater estuary and wastewater treatment plant) and have different properties such as sediment size distributions and wave angle of attack, all while being subject to similar lake-scale dynamics. We also investigate trends in microplastic contamination within each beach, focusing on differences in microplastic concentrations in the surface vs. subsurface sediments and wrack vs. inner swash.

Methods

Study site

We selected three beaches on the shores of western Lake Superior for sample locations, as shown in Fig. 1. Wisconsin Point beach (WIP) and Minnesota Point beach (MNP) are both located near the mouth of the St. Louis River Estuary, as it flows into Lake Superior. Burlington Bay beach (THMN) in Two Harbors, MN is adjacent to the effluent of the Two Harbors waste water treatment plant, which is located approximately 800 m south of the beach.

In Fig. 1, we show an aerial photograph of each beach with a red line marking the 100 meter span alongshore where we obtained samples. Using the US Army Core of Engineers Wave Information Study (WIS) buoys on Lake Superior, we found the direction of the prevailing wind driven waves for the buoy located the closest to each study area. This dominant wave direction is marked by a white arrow pointing towards each beach (Engineers et al., 2022).

The estuary is a suspected source of microplastic contamination as rivers have been shown to be a source of microplastic pollution in other regions (Lebreton et al., 2019; Lenaker et al., 2019). Waste water treatment effluent has also been shown to be a source of microplastic pollution, especially fibers from fabrics and clothing due to washing machine effluent (Mason et al., 2016; Browne et al., 2011). Although MNP and WIP are close to each other geographically, they differ in that the area surrounding MNP is far more developed than the area surrounding WIP. One additional difference is that MNP has been nourished in the past with sediments dredged from the adjacent harbor. We did not find any information about previous beach nourishment projects at WIP nor THMN. Sediment size distributions at each beach show that WIP and MNP have quite fine and well-sorted sediment whereas the sediment at THMN is coarse and less well-sorted (Fig. 2; see Sediment processing and microplastic isolation section for the methods used to generate this data).

Beach sediment sampling

Fig. 3A shows the sampling structure at each beach. We sampled beach sediments along three cross-shore transects separated in the alongshore direction. We placed down a 100 meter tape measure along the wrack line of each beach. We selected the sample locations using three previously chosen random distances to prevent bias in sample selection from beach conditions. At each sample location along the beach, we collected a surface sample and core sample at the wrack line and a core sample at the swash line. Thus, we collected surface sediment samples at the wrack line [following *Microplastics Sampling and Processing Guidebook,*] (Sartain et al., 2018) and many other studies (Wessel et al., 2016), but we augmented this procedure by also collecting sediment core samples at the wrack line and swash line. In Fig. 3B, we show a beach cross section specifically noting the wrack line, or high water mark that is often characterized by significant debris accumulation from the water. The swash zone is the region starting from the waterline at the wave drawdown location to the maximum active wave run-up location. For the sediment sampling in this study, we define the swash line as the location of maximum active wave run-up on the beach. Sampling multiple transects across the beach allow us to look at the variability of microplastics in the alongshore direction, while the wrack line and swash line samples allow us to consider the cross-shore concentration change of microplastics, and the surface and core samples at the wrack line lets us look at variability of microplastic concentrations with depth. Thus, our modified sampling method facilitates microplastic concentration analysis across all three spatial dimensions (cross-shore, alongshore, and depth).

For the surface sediment samples along the wrack line, we collected the top 1–2 cm of sand from a 25 cm x 25 cm wood frame placed at the sample location. We generally followed the methods from Sartain et al., 2018, specifically noting that we used a stainless steel scoop, did not first sieve the sediments in the field, and stored sediments in covered aluminum foil pans. Between samples, we rinsed the shovel with DI water to mitigate contamination between samples.

For the core samples, along the wrack line and swash line, we sampled with a 7.6 cm (diameter) x 15.2 cm (depth) cylindrical stainless steel core sampler (AMS Inc.). With a small stainless steel shovel, we emptied the core into an aluminum foil pan and covered it. Between samples we cleaned all equipment first with lake water

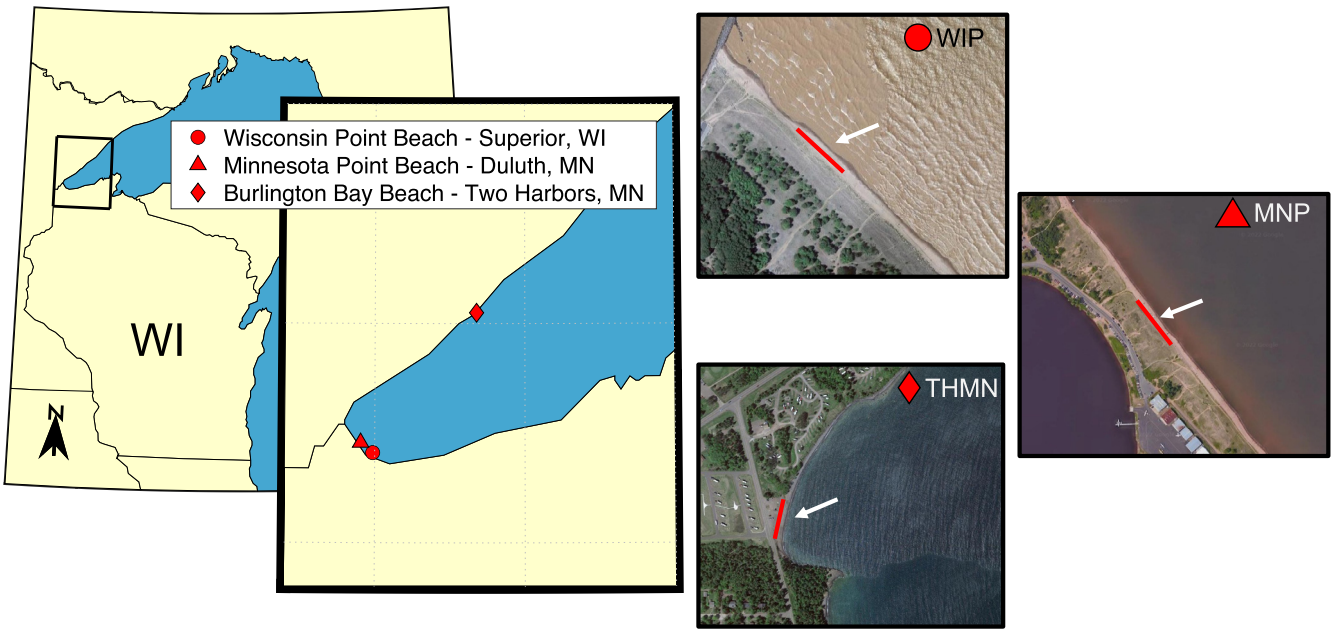

Fig. 1. Maps and aerial photographs showing study sites: Wisconsin Point beach (WIP; Superior, WI), Minnesota Point beach (MNP; Duluth, MN), and Burlington Bay beach (THMN; Two Harbors, MN). The red line indicates the alongshore distance of sample spread, and the white arrow indicates the dominant direction of wave forcing.

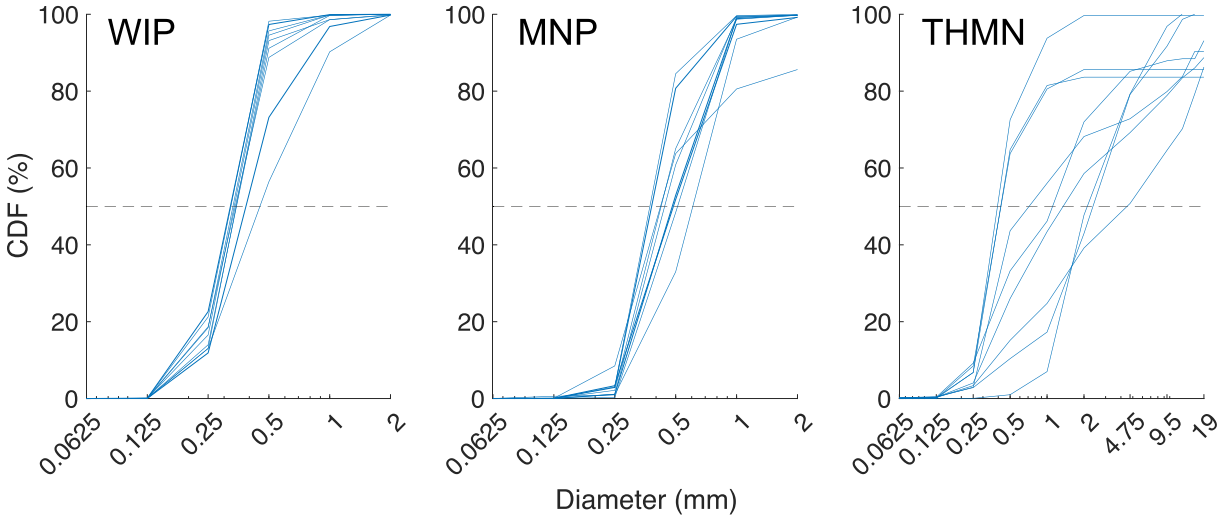

Fig. 2. Cumulative distribution function of sediment grain size at each study beach. Median grain diameter for each sample is where distribution crosses dashed line at 50%, indicating the d_{50}.

to remove any visible sediments followed by a rinse with DI water to mitigate any cross-sample contamination.

Sediment processing and microplastic isolation

We used a widely accepted protocol for microplastic separation from sediments (Zobkov and Esiukova, 2017), which is based on a method published by the National Oceanic and Atmospheric Administration (Masura et al., 2015). Our extraction differs slightly from Zobkov and Esiukova (2017) by repeating the density separation for a total of three times and increasing the solution settling time on the third separation from one hour to overnight. Both of these improvements were based on the results from Besley et al. (2017).

In order to reuse $ZnCl_2$ solution between different samples, we filtered the $ZnCl_2$ solution through a GF/F filter and periodically measured the density to ensure it remained within acceptable tolerances ($1.4 < \rho < 1.6$ g/ml). We filtered the settled sediment solutions through a 125μm stainless steel mesh sheet folded into a basket using a stainless steel ladle to scoop the solution off the top of the beaker, before pouring the rest of the solution into the filter.

Following the third density separation, we performed a wet peroxide oxidation (WPO) to dissolve organic matter and calcite digestion. We performed the peroxide digestion at room temperature based upon results from Lenaker et al., 2021 to prevent possible degradation of any plastics. We let this solution sit for 5 min in a fume hood, covered with a watchglass. If significant organic matter remained, we removed large organic chunks with tweezers, rinsing

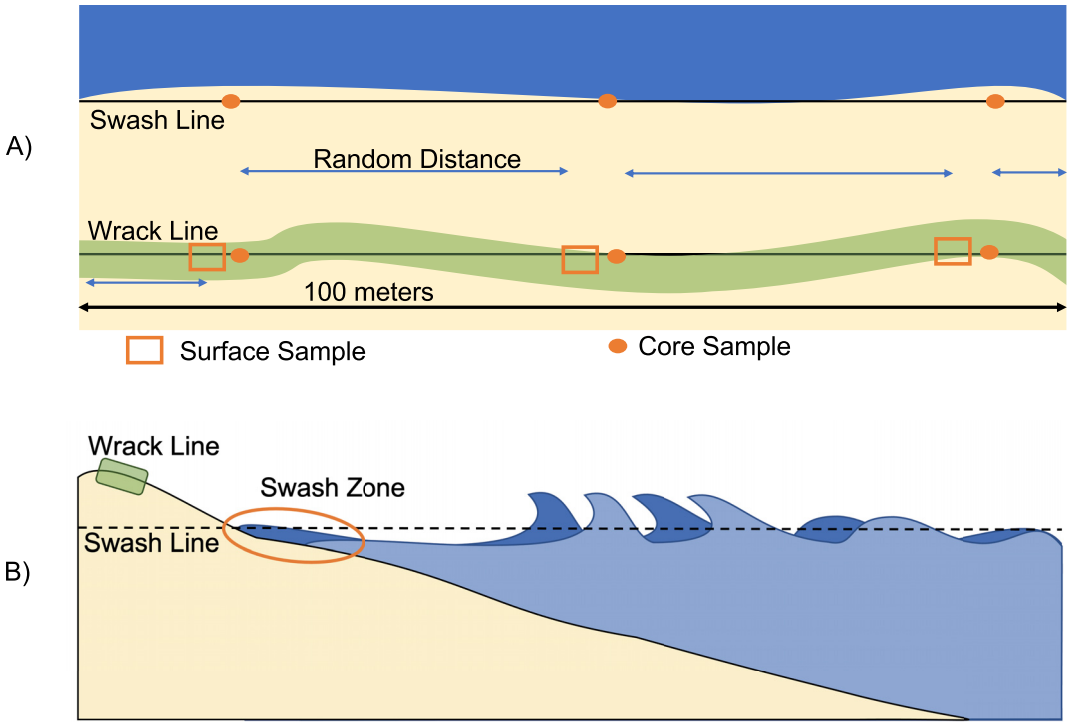

Fig. 3. A) Diagram of beach sample collection. We collected surface samples at three points along the wrack line and core samples at three points along both the wrack and swash lines. Surface samples consisted of the top 1–2 cm of sand in a 25 cm x 25 cm area and the core samples consisted of 7.6 cm diameter core plug from the surface to a depth of 15.2 cm. B) Beach cross section noting the swash zone - region from wave draw down to maximum wave run-up, swash line - maximum wave run-up location, and the wrack line - high water mark area with significant debris accumulation.

with Milli-Q into the solution before setting the organics aside. We then added 25 ml of 4.5% HCl solution to the WPO solution beaker and stirred. We left this beaker covered in the fume hood to sit overnight (> 15 h) increasing this time on account of not heating the solution for the WPO.

We then performed a final density separation in the original zinc chloride solution using the density separator shown in Fig. 9 of Masura et al., 2015. We collected the supernatant and buoyant solids in a beaker which was then poured through a glass fiber filter with 0.7μm pores using vacuum suction. We rinsed the filter with DI water through the vacuum filter and subsequently placed the filter in a plastic petri dish (Polystyrene) and covered with aluminum foil until ready for microplastic identification.

While we used 400 g of wet sediment from each sample for microplastic isolation ($M_{\text{wet,plastic}}$), we also measured out a second 400 g sample of the wet sediment to do a grain size analysis ($M_{\text{wet,sed}}$). We dried the sample for sediment analysis to a constant mass ($M_{\text{dry,sed}}$). We calculated for a ratio of the dry to wet sediment analysis masses, then solved for the dry mass of the sediment used for plastic separation ($M_{\text{dry,plastic}}$) via

$$M_{\text{dry,plastic}} = \frac{M_{\text{dry,sed}}}{M_{\text{wet,sed}}} M_{\text{wet,plastic}}. \quad (1)$$

For each sediment sample, we found the sediment grain size distribution, following the methods outlined by the *American Society for Testing and Materials* (ASTM International, 2020).

Microplastic identification

We began the process of microplastic identification and quantification by visually counting the number of particles that were in the filter using an optical microscope (up to 50x magnification). We counted the particles following the micropalstic sampling guidebook (Sartain et al., 2018), categorizing the plastics into: fibers, fragments, films, and microbeads. We did not count particles that had a visible cellular structure (Norén, 2007). We counted particles under white light and under a violet *Nightsea* light (400–415 nm) and filter (450 nm longpass) following a customized method (developed by Dr. Rios at the University of Wisconsin - Superior). Clear plastics are easier to see and count under the violet light as they fluoresce at the wavelength of this light.

We selected 20–30 particles from each sample for Fourier-transform infrared spectroscopy (ATR-FTIR; Thermo Scientific Nicolet iN10 spectrometer with a Germanium tip ATR and detector cooled with liquid nitrogen) analysis to confirm if the particle is plastic and to identify the polymer type. This amounted to at least 4% of each sample, but usually 16%, and up to as much as 73% of particles that were visually identified were analyzed with ATR-FTIR. We analyzed particles with 16 scans for 5 s with a resolution of 4 cm^{-1} and a detector spectral range of 4000 to 650 cm^{-1}. We collected a background spectra before each sample. We compared the generated spectra to an internally generated spectral library, searching the whole spectrum or using a region search as necessary, requiring a minimum match of 70% to confirm the particle identity as plastic, although we generally were able to match above 80% throughout the analysis.

Microplastic quantification

We visually counted 3981 particles across all non-blanks samples (3763 fibers) and analyzed 630 particles with ATR-FTIR analysis for identification (577 fibers). Among the 9 sample blanks, we counted 697 particles (690 fibers) and analyzed 312 with ATR-FTIR (309 fibers). We show two example ATR-FTIR spectra for synthetic and semi-synthetic fibers, plotted with a library standard spectra, in Fig. 4.

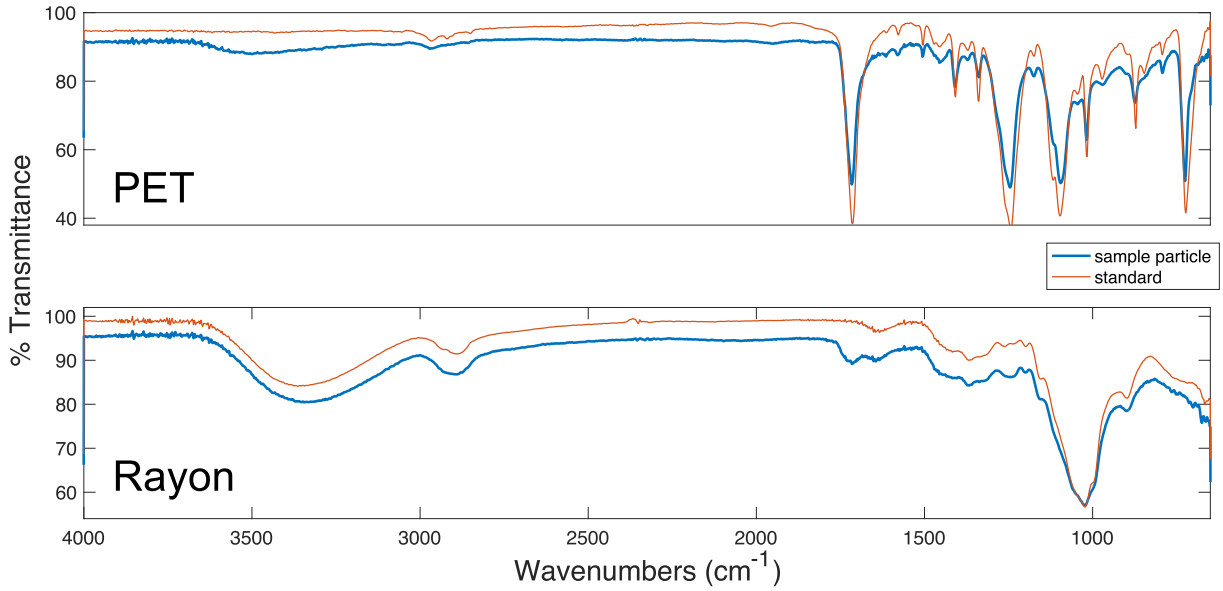

Fig. 4. Example ATR-FTIR spectra from the two most common synthetic (PET) and semi-synthetic (Rayon) fibers. The blue line shows the spectra collected from the sample and the orange line shows the standard spectra from the internal library.

Following Lenaker et al., 2021, the count of particles of a specific morphology and polymer type in each sample (C_{polymer}) is calculated from the number of particles verified as a specific polymer (N_{polymer}), the total number of particles analyzed with ATR-FTIR ($N_{\text{ATR-FTIR}}$), and the total number of visually identified particles (N_{count}) using

$$C_{\text{polymer}} = \frac{N_{\text{polymer}}}{N_{\text{ATR-FTIR}}} N_{\text{count}}. \quad (2)$$

The plastic contamination as quantified by particle counts in the blank samples was small: Fibers: PET - 4.04 p sample^{-1}, Modacrylic 0.11 p sample^{-1}, Rayon 32.36 p sample^{-1}, Fragments: Alkyd Resin - 0.2 p sample^{-1}, Cellophane - 0.1 p sample^{-1}. To account for sample contamination in the particle/polymer counts, we subtract the average blank particle/polymer counts from each non-blank sample count. We do this by particle shape and polymer rather than over all plastics to avoid over compensating for contamination within specific categories. We then normalize the number of particles by the mass of the dry sediment for the respective sample.

Quality assurance and quality control

We followed careful methods to prevent possible plastic contamination of the samples while performing the microplastic separation. We covered all samples with aluminum foil or glass lids when we were not actively manipulating or observing. When working with the samples, we wore 100% cotton lab coats and blue nitrile gloves. While working through this experiment, we often wore various face masks while in the lab. Although we did not restrict the material of the face covering, we suspect there may be benefits of the face mask limiting the contamination or disruption from breathing near a sample. We were careful to use glass and stainless steel laboratory equipment whenever possible, only using plastic where necessary. Even with such contamination controls, we included a measure of potential sample contamination due to the difficulty of preventing all contamination from microplastics and other anthropogenic particles during laboratory analysis of samples. To account for quality assurance and quality control throughout the laboratory analysis, we included 9 sample blanks that went through the entire isolation process, although no sediment was added at the start. We processed each blank simultaneously with three other non-blank samples. Whenever the samples were uncovered and exposed to the air, the blank was left open, and the blank was also covered whenever the samples were covered. Although blanks were processed in conjunction with certain samples, we treat all blanks as aggregate measures of contamination throughout the isolation process. After the isolation, during microplastic counting and identification, we counted and processed the blanks the same as the non-blank samples. In this way, the blanks were exposed to the same open air conditions for similar duration for counting and processing.

In addition to laboratory blanks, during the ATR-FTIR analysis, we estimated contamination by using a clean filter that we left in a glass petri dish on the lab bench, and uncovered whenever any sample was exposed for counting or analysis. After concluding all ATR-FTIR analysis, the particles on the control filter numbered 200 fibers. We found that 15.8% of the fibers were plastic (2 PET and 1 Modacrylic) from ATR-FTIR analysis on 19 fibers, resulting in 31.6 total plastic fibers on the control filter. We estimate that each of the samples (27 field samples and 9 blank samples) would each have been exposed to the laboratory air for 0.5 h, whereas the control filter was exposed for 18 h. From this, we can estimate a rate of contamination from laboratory air to be 1.8 plastic fibers/hour, or less than 1 fiber in each sample filter. This is considered to be a negligible level of contamination, which is already accounted for in our blanks, and we therefore do not account for it again numerically in our results.

Finally, the blank samples do not account for contamination specifically during the sample collection in the field. Sample collection contamination at the field sites could result from exposure to air, contamination from sampling equipment, or clothing fibers from individuals collecting samples. This contamination is expected to be small since the sediment was quickly covered once collected and since we cleaned sampling equipment with DI water between samples.

Results and discussion

Microplastic concentration

We sum the microplastic count kg^{-1} of dry sediment for each particle shape and polymer to get a total count of microplastics

in one kg of dry sediment from each sample. Over all samples, we find the mean microplastic (fully synthetic) concentration of 65 particles kg^{-1} of dry sediment (fibers and fragments) with a median of 35 particles kg^{-1} of dry sediment, showing that there is some skewness in the distribution towards extreme values. Fibers dominated the recovered particles, we counted no microbeads, and the amount of fragments and films were low compared to fibers such that we combined all non-fibers into a fragment category. We show examples of recovered microplastic fibers and fragments in Fig. 5.

With ATR-FTIR analysis, we found a distribution of synthetic and semi-synthetic particles by shape across each beach (Fig. 6A). Fibers dominate, with fragments appearing mostly at Two Harbors. Semi-synthetic fibers were exclusively Rayon, while the distribution of plastic fibers analyzed with ATR-FTIR was mostly PET (polyester) (Fig. 6B). We found that the concentration of plastic fibers was dominated by PET ($p < 0.05$) with a t test of the mean concentrations of PET fibers and all non-PET plastic fibers. All fibers recovered except PP (polypropylene) are polymers with densities greater than 1.0, thus negatively buoyant. PP (positively buoyant) makes up less than 10% of the plastic fibers recovered and analyzed with ATR-FTIR. Non-synthetic fibers were mostly found to be cotton and cellulose.

We see fully synthetic and semi-synthetic fibers across all beaches, although the concentration of semi-synthetic fibers is more consistent while we see fewer synthetic fibers at WIP. For the remainder of the analysis, we choose to focus on fully synthetic fibers and fragments to understand the deposition and accumulation of true microplastic particles on these beaches.

Fig. 7 shows that fully synthetic fibers dominate over fully synthetic fragments ($p < 0.05$ using a t test for the mean values of each distribution). Although the mean of the fragment population is not zero, it is small and mostly limited to only one beach from the study. We therefore choose to focus on the fibers for a more robust analysis.

Microplastic distribution

We expected the beach and different samples within each beach to be predictors of microplastic concentrations within sediments. We tested this hypothesis utilizing a 2-way analysis of variation (ANOVA) test with interactions between beaches and samples. We found that the microplastic concentrations across beaches and samples is not linked together for microplastic fibers. Thus, we proceeded with a 2-way ANOVA test between beaches and samples, ignoring any interactions. Here, we found that the variation of the mean concentrations across beaches is significant ($p < 0.05$) although not for variations between sample types. This means that where a sample is taken on the beach is not a statistically significant predictor of microplastic fibers, but the beach itself is a significant factor, consistent with previous studies of surface sediments only (Vermeiren et al., 2021). Many parameters are wrapped up in properties of the beach - proximity to microplastic sources, sediment properties, distance from the start of the beach, primary incident wave angle, and others. Thus, it is not possible to distinguish between these factors in our study.

We examine the microplastic concentration distributions at each beach in Fig. 8. We see that the mean concentrations at MNP and THMN are statistically the same and together different from WIP, consistent with the results of the 2-way ANOVA test. Although the mean concentration for WIP is not zero, it is much smaller than MNP and THMN, and thus appears to be cleaner in regards to microplastic contamination. When looking at the variance of these distributions, we use an f test between each pair. We find that the variance of the concentration distribution is not statistically different between THMN-MNP, but it is for WIP-THMN and WIP-MNP ($p < 0.05$).

To understand the burial of microplastic fibers in beach sediments, we look at the difference between microplastic fibers in the surface and the subsurface at the wrack line. To evaluate the concentration of plastics in the subsurface, we need to compensate

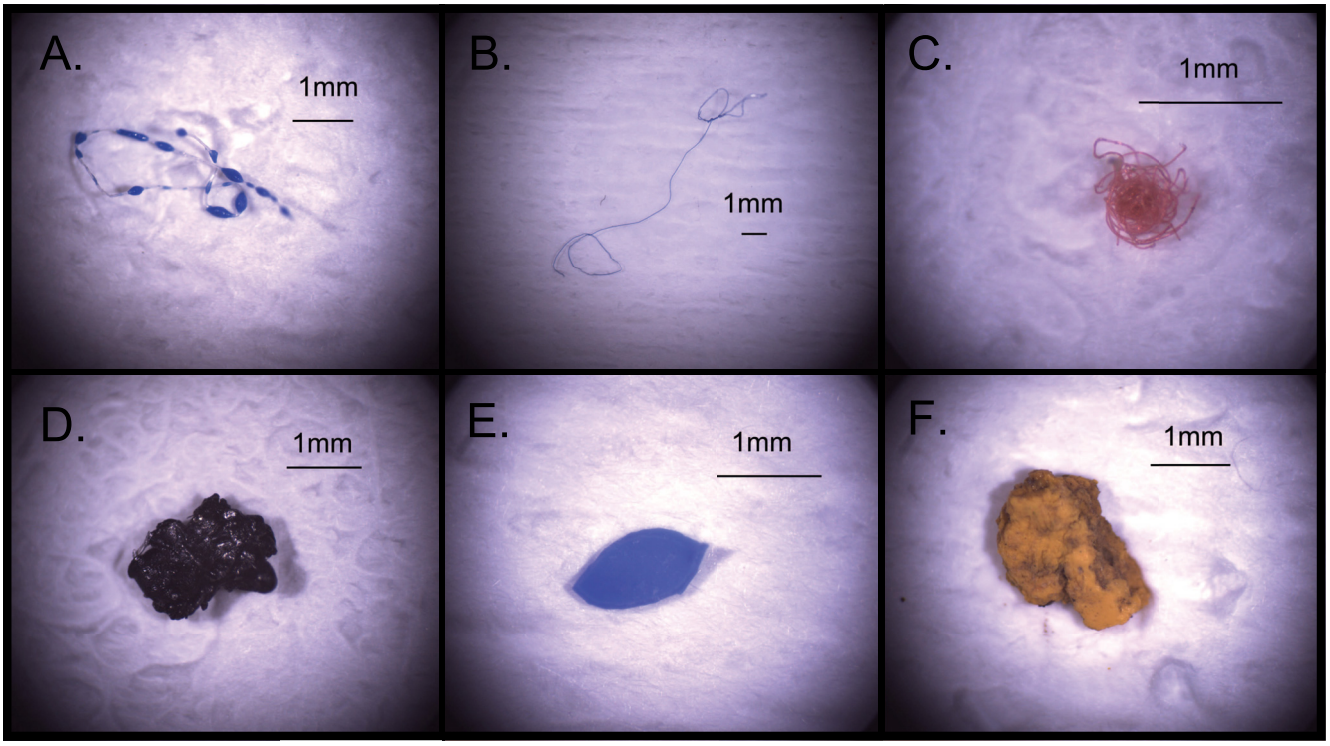

Fig. 5. Example microplastic fibers (A-C) and fragments (D-F) recovered from beach sediments. A) Clear PET fiber covered with blue PMMA dots. B) Blue PP fiber. C) Red PAN fiber bundle. D) Black PE fragment. E) Blue modacrylic fragment. F) Yellow PMMA fragment.

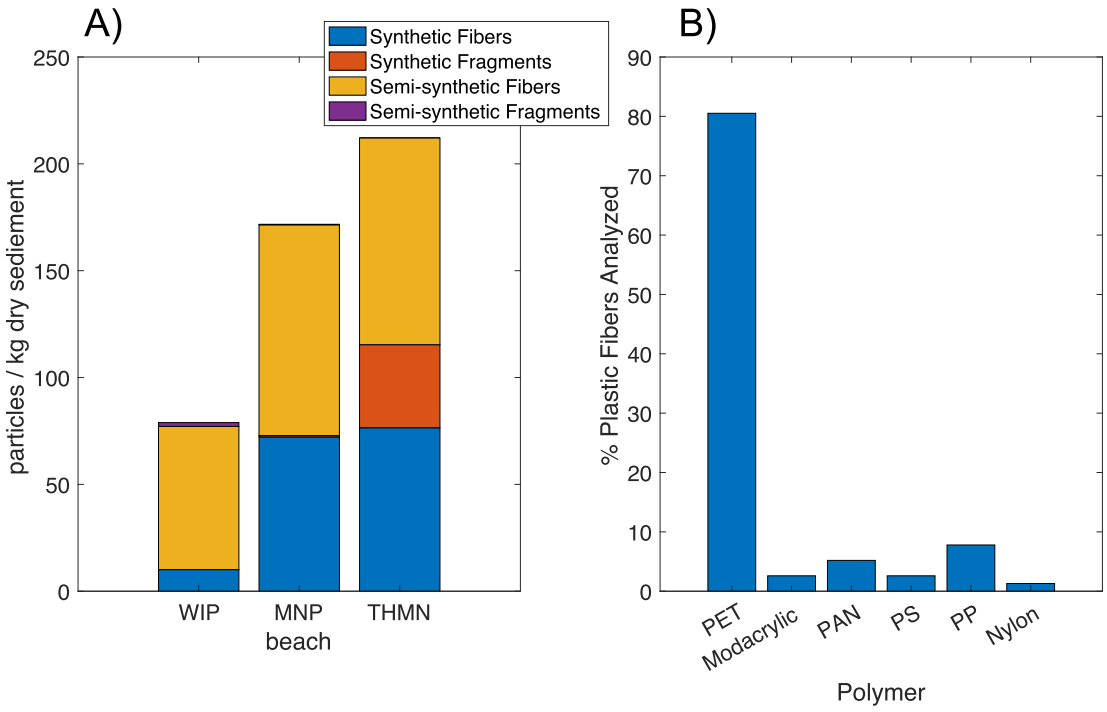

Fig. 6. A) Synthetic and semi-synthetic microplastic particle concentrations by morphology across beaches. B) Percentage of synthetic fibers analyzed in ATR-FTIR for each fully synthetic polymer identified (PET - polyester $\rho = 1.38 - 1.41$ g/cm^3, Modacrylic $\rho = 1.30 - 1.37$ g/cm^3, PAN - polyacrylonitrile $\rho = 1.184$ g/cm^3, PS - polystyrene $\rho = 1.04 - 1.06$ g/cm^3, PP - polypropylene $\rho = 0.85 - 0.92$ g/cm^3, Nylon $\rho = 1.13 - 1.16$ g/cm^3). (Lenaker et al., 2021; Texcoms Textile Solutions, 2019).

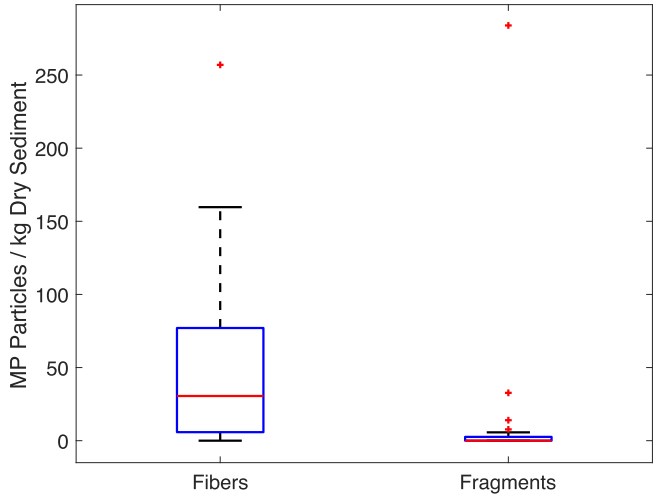

Fig. 7. Compare spread of microplastic concentrations between particle morphology: fiber vs. fragments.

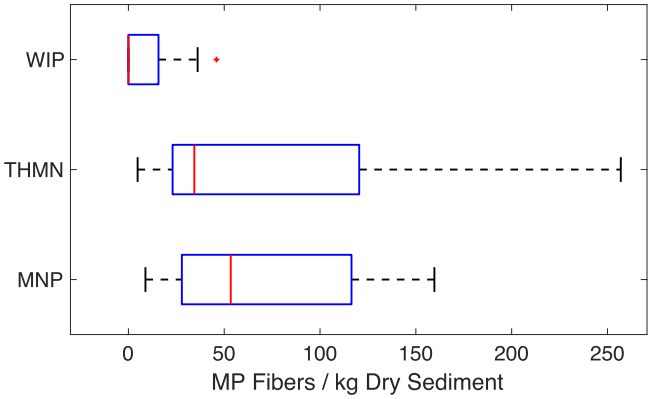

Fig. 8. Spread of fully synthetic microplastic fiber concentrations at each beach sampled.

for the plastics that would be recovered from the surface sediment at the top of the core plug. For this, we find a concentration of microplastic fibers in the surface sample by area. We multiply this by the area of the core sample and then subtract the count of particles from the surface portion of the core sample before we normalize by an adjusted sediment mass. Fig. 9A shows these distributions of the surface and subsurface microplastic fiber concentrations. With a t test, we find that the average concentration between the surface and the subsurface is not significantly different. The variance between these two samples is significantly different ($p < 0.05$) with a larger spread in concentrations for the subsurface than the surface. This difference in variance in the plastic concentrations between the surface and subsurface could be driven by coastal processes, which we discuss further below.

To consider the cross-shore variability of microplastic fiber accumulation on a beach, we compared the core sample from the swash line and the core sample from the wrack line, showing the distribution of plastic concentrations in Fig. 9B. With a t test, we find that the mean microplastic fiber concentrations are not statistically different. Although the wrack core sample appears to have a larger spread, an f test reveals that the variances are also not statistically different across the paired cross-shore core samples. While we see that the microplastic concentrations are not significantly different in the cross-shore direction, we do see a difference in the variance of the sediment d_{50} between these locations ($p < 0.05$). There is a larger spread of sediment sizes in the swash core than in the wrack core, which means that the wrack core sample is more consistently finer sediment particles while the swash core is a larger variety of sediment sizes due to the more extreme hydrodynamic conditions of breaking waves in and near the swash zone that transport sediments of various sizes. It is interesting that

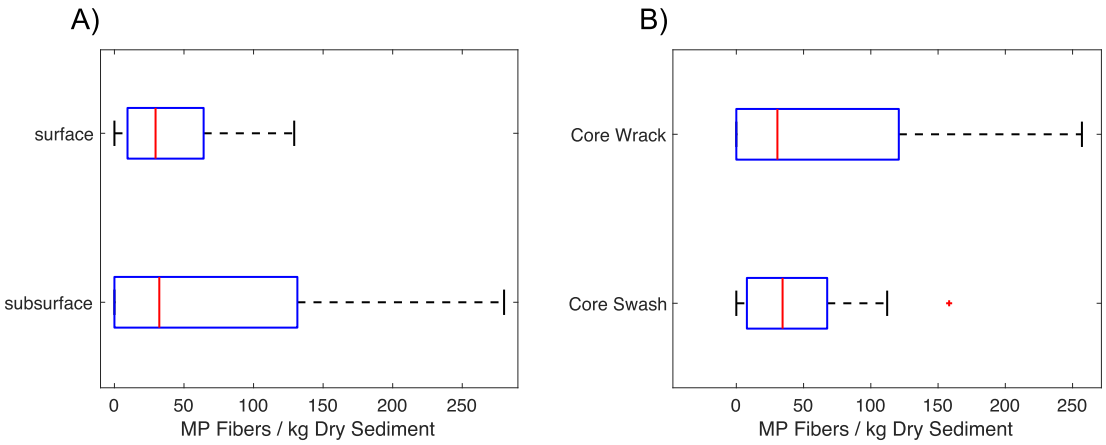

Fig. 9. A) Spread of fully synthetic microplastic fiber concentrations between surface sediments and subsurface sediments. B) Spread of fully synthetic microplastic fiber concentrations between core samples from the wrack line and core samples from the swash line.

although we see a difference in variance of sediment size, indicating some difference in sediment transport between the swash and the wrack, this difference does not appear to affect microplastic accumulation or spread in microplastic concentrations between the wrack and the swash.

Further discussion

This study design was formulated to allow us to capture trends in microplastic distribution across and within the beaches of our study site. We had hypothesized that we would observe trends such as: (1) a higher likelihood of finding more buoyant microplastics in surface sediments compared to subsurface sediments at the wrack line; (2) a higher likelihood of finding larger microplastics in the swash zone compared to the wrack line; (3) a higher likelihood of finding microplastic fibers in THMN due to the proximity to the wastewater treatment plant. Instead, the data paint a picture that is both simpler and more complex than our original hypotheses. In this section, we pick out certain trends from our data and generate new hypotheses regarding how they might be linked to coastal processes and other factors that may determine microplastic pollution of beach sediments.

First, it is quite interesting that the mean microplastic fiber concentrations in surface vs. subsurface sediments were not statistically different, but that we did detect a significant difference in the variances of these distributions. The consistency in the mean value makes sense if we hypothesize that the lake is the primary source of plastics to the beach and the mean concentration within the sediment reflects the average concentration of the plastics in the water. Concerning the variance, intuitively it makes sense that there is a smaller spread in the concentration for the surface microplastics because we expect microplastic accumulation to occur in the surface sediments at the wrack whenever the microplastic contaminated water reaches this level of the beach. In contrast, we only expect microplastics to accumulate in the subsurface when the water is high enough and the hydrodynamic conditions are energetic enough to be mixing up the sediments beneath the surface of the wrack line. Thus, the microplastic fluxes between the water and surface sediments will occur more frequently compared with subsurface sediments. The subsurface sediments will only gain or lose microplastic particles during more energetic hydrodynamic events, and this intermittency is likely the cause for a larger spread in the subsurface concentrations.

Second, no clear trends are observed with regards to the microplastic pollution level on a given beach and that beach's distance from potential sources or sediment size distribution. The significant difference in microplastic fiber concentration between WIP and MNP is particularly surprising. These two sites are located very close together, and should experience similar lake scale dynamics. While MNP previously received a sediment nourishment from benthic sediments that may have been contaminated by microplastics, this was not the case for THMN, which has not received a sediment nourishment, yet still contained a similar level of microplastic pollution. It appears that predicting microplastic concentration on a given beach is more complex than proximity to these potential sources. However, we cannot discount atmospheric deposition as a potential avenue for microplastic accumulation in the sediments. Previous studies have shown significant microplastic deposition from the atmosphere (Dris et al., 2016; Cai et al., 2017; Klein and Fisher, 2019; Huang et al., 2021; Ding et al., 2021), with Dris et al., 2016; Huang et al., 2021; and Ding et al., 2021 finding significant PET fiber deposition, similar to our findings. This could reconcile the trends with microplastic concentrations at the observed beaches in this study. High concentrations at MNP could relate to the close proximity to development and urban center of Duluth, MN, while the WIP peninsula is largely undeveloped and further from the urban center of Superior, WI. When also considering THMN, we can note that the beach sampled was closer to development, more similar to MNP than WIP which is consistent with the increased microplastic concentrations found.

Third, a single class of microplastics dominates over all others: polyester fibers. We know that atmospheric deposition could be one source of these particles to the beach, but they could also be accumulating from the water. Polyester fibers are negatively buoyant, which if the source is the lake water, makes it even more surprising that these fibers are transported all the way to the swash zone and deposited within beach sediments. Sampling of lake waters at the surface and within the water column [Rios Mendoza, private communication, and] (Hendrickson et al., 2018) show a much more varied composition of the microplastic population, i.e., a wider variety of microplastics are found in the waters in western Lake Superior than the beach sediments. The fact that of these varied microplastics in the water, the polyester fibers are selectively transported into the nearshore area is surprising. One possible hypothesis is that the settling velocity of these particles could be lower than those of other plastics that are in the water, making them more likely to stay in suspension long enough to reach the shore. It has been shown that microplastic fibers have significantly smaller settling velocities than microplastic sheets of the same material and length (Jalón-Rojas et al., 2022). Another factor that notably impacts a fiber's individual settling velocity is the flexibility and curvature, especially for fibers longer than

2 mm, as demonstrated by Nguyen et al., 2022. We did notice that many recovered and confirmed microplastic fibers were curved (Fig. 5 A-C), and thus should experience a smaller settling velocity than the equivalent straight fiber. Clearly the relatively simplistic results of microplastic transport in coastal waters (Forsberg et al., 2020; Kerpen et al., 2020) do not explain the trends we observe here and there remain intriguing questions regarding the transport properties of fibers in coastal environments that could be addressed with controlled laboratory studies.

Finally, comparing the results of this study with previous work on microplastic pollution in the Great Lakes, we demonstrate the following expected results: (1) Previous studies of Lake Michigan found surficial benthic sediment to show a significant bias toward fibers (Lenaker et al., 2021) with PET, HDPE, and Rayon fibers dominating the polymer types, which is a similar blend to our results; (2) Previous studies of beaches on Lake Erie (Dean et al., 2018) and Lake Ontario (Ballent et al., 2016) found higher concentrations of microplastics compared to our results and previous results (Minor et al., 2020) of Lake Superior beaches. This is not too surprising as we can expect Lake Superior to be the cleanest of the Great Lakes concerning microplastic pollution since it is the furthest upstream and the least impacted by waste water treatment plants due to the significant volume and low population density in the watershed (Twiss, 2016). Although we expect Lake Erie and Lake Ontario to contain higher concentrations of microplastics compared to Lake Superior, we must note the possibility of overestimation in concentration from each of these studies, as each study either completely excluded or grossly under represented fibers during plastic confirmation and polymer identification. Various methods to separate microparticles, neglecting to confirm recovered microparticles are indeed plastics, and the use of different methods to identify polymer types, are widespread issues making it difficult to compare microplastic concentrations across studies. Hence, we are unable to draw robust conlusions about the spatiotemporal nature of microplastic contamination at Lake Supreior beaches from comapring our results with previous data (Minor et al., 2020; Whitmire et al., 2017).

Conclusions

Sediment sampling from Lake Superior beaches showed a mean microplastic concentration of 65 p kg^{-1} across all samples, although with significant variation between individual beaches. Proximity to suspected microplastic sources are not a significant predictor of pollution levels at a given beach, as WIP had significantly lower concentrations of microplastics than MNP even though the two beaches are adjacent to each other.

The primary microplastic morphology and polymer being polyester fibers is consistent with the idea that textiles are the main source of microplastics into the lake (and therefore beaches). While it is surprising to see these dense particles accumulating on the beach, there is evidence to suggest that the lower settling velocity of microplastic fibers and the possibility of entanglement and curls leading to even lower settling velocities could explain this result. Microplastic deposition from the atmosphere could further contribute to this result.

Additionally, microplastic concentrations do not seem to differ between surface sediments and subsurface sediments, although there is more variation in the concentration of plastic particles in subsurface sediments. We hypothesize that the rarer, highly energetic hydrodynamic conditions cause accumulation in the subsurface, making subsurface microplastic concentrations more variable. Previous laboratory experiments have begun to consider the processes that control the distribution of microplastics in the nearshore and beach environment, but more of these experiments are necessary since we do not yet have a strong understanding of such phenomena.

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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