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A Multicompartment Assessment of Microplastic Contamination in Semi-remote Boreal Lakes

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Abstract: Microplastic contamination is ubiquitous across the globe, even in remote locations. Still, the sources and pathways of microplastics to such locations are largely unknown. To investigate microplastic contamination in a semi-remote location, we measured microplastic concentrations in nine oligotrophic lakes within and around the International Institute for Sustainable Development—Experimental Lakes Area in northwestern Ontario, Canada. Our first objective was to establish ambient concentrations of microplastics in bottom sediments, surface water, and atmospheric deposition in semi-remote boreal lakes. Across all lakes, mean shallow and deep sediment microplastic concentrations, near-surface water microplastic concentrations from in situ filtering, and dry atmospheric microplastic deposition rates were 551 ± 354 particles kg^{-1}, 177 ± 103 particles kg^{-1}, 0.2 ± 0.3 particles L^{-1}, and 0.4 ± 0.2 particles m^{-2} day^{-1}, respectively. Our second objective was to investigate whether microplastic contamination of these lakes is driven by point sources including local runoff and direct anthropogenic inputs or nonpoint sources such as atmospheric deposition. Lakes were selected based on three levels of anthropogenic activity—low, medium, and high—though activity levels were minimal across all study lakes compared with highly populated areas. Whereas a positive correlation would indicate that point sources were a likely pathway, we observed no relationship between the level of anthropogenic activity and microplastic contamination of surface water. Moreover, the composition of microplastics in surface water and atmospheric deposition were similar, comprising mostly polyester and acrylic fibers. Together, these results suggest that atmospheric deposition may be the main pathway of microplastics to these remote boreal lakes. *Environ Toxicol Chem* 2024;00:1–13. © 2024 The Authors. *Environmental Toxicology and Chemistry* published by Wiley Periodicals LLC on behalf of SETAC.

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INTRODUCTION

Plastic pollution is increasingly a concern to policymakers and the public because of its persistence in the environment, potential for long-range transport, and threat to wildlife and human health. Mechanical and chemical abrasion of plastics results in microplastics (<5 mm in size) that come in many shapes, sizes, and polymer types (Browne et al., 2011; Cole et al., 2011). The small size of microplastics facilitates their transport in the atmosphere, in water, and through food webs (Bergmann et al., 2019; Brahney et al., 2020; Setälä et al., 2018).

While research on microplastics has historically focused on marine systems, there has been an increase in studies examining microplastics in freshwater since the 2010s (Lu et al., 2021). Today, there is still a need to understand the fate and effects of microplastics in freshwater (Nava et al., 2023;

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Wang et al., 2021). Differences in salinity, temperature, and water currents between marine and freshwater systems differentially affect the dispersion and transport of microplastics (Eerkes-Medrano et al., 2015). Therefore, what we know from microplastics in marine environments cannot be directly applied to freshwater systems (Eerkes-Medrano et al., 2015). Moreover, freshwater systems are commonly referenced as conduits of plastic between land and sea, rather than a compartment within the larger plastic cycle (Hoellein & Rochman, 2021; Welsh et al., 2022b; Windsor et al., 2019). There is evidence that freshwater lakes and rivers act as dynamic sinks and sources of microplastics (Nava et al., 2023). As such, investigations into freshwater, in addition to other compartments, are important for understanding the full scope of the global plastic cycle (Bank & Hansson, 2019; Hoellein & Rochman, 2021; Welsh et al., 2022b).

Understanding the sources and pathways of microplastics, both locally and globally, is a developing area of research. In recent years, atmospheric microplastics have been cited as a potential pathway to both urban and remote locations (Allen et al., 2019; Brahney et al., 2020; Cai et al., 2017; Dris et al., 2015; Evangeliou et al., 2020; Klein & Fischer, 2019). The small size and low density of microplastics make them suitable for atmospheric transport (Allen et al., 2019; Brahney et al., 2020), and thus microplastics have been observed in remote locations such as the Mongolian Lake Hövsgöl, the Pyrenees mountains, and the Tibetan Plateau (Allen et al., 2019; Free et al., 2014; Jiang et al., 2019). However, atmospheric transport is not the only pathway for microplastics to contaminate remote areas. Direct pollution from litter, surface runoff, boating activities, shedding of swimwear, and footwear abrasion can also contribute to this contamination (Forster et al., 2020; Gao et al., 2021; Horton et al., 2017). Discerning between these direct and indirect pathways as contributors of microplastic contamination in remote areas will further help us understand global transport and fate.

We investigated the quantity and characteristics of microplastics in semi-remote boreal lakes within and around the International Institute for Sustainable Development—Experimental Lakes Area (IISD-ELA) in northwestern Ontario, Canada. In nine freshwater lakes, we measured ambient concentrations of microplastics in sediment, in surface and near-surface waters, and from atmospheric deposition to acquire an understanding of the types, sizes, and shapes of microplastics in the area. We also investigated whether microplastic contamination is driven by direct anthropogenic inputs to the lakes and whether atmospheric pathways may be a potential alternative source. Our study tests the hypothesis that semi-remote boreal lakes will be contaminated with plastic and that lakes that are visited more often for recreation and research will have more plastic than those that are not. Specifically, we predicted that (1) ambient levels of microplastics in surface water and sediment and from atmospheric deposition will be low compared with those in highly populated locations, (2) there will be a positive relationship between anthropogenic activity and microplastic concentrations within the study area, and (3) microplastic compositions in surface water will differ from compositions in atmospheric deposition due to local anthropogenic activity. If the latter two predictions are not supported, the discrepancy may mean that atmospheric transport is a greater pathway of microplastics to the region than direct anthropogenic inputs.

MATERIALS AND METHODS

Study site

The IISD-ELA (49.6603°N, 93.7283°W; Figure 1) in northwestern Ontario contains 58 lakes dedicated to scientific research. The closest population centers include Vermilion Bay (population ~1000) and Kenora (population ~15,000), Ontario, located 31 km northeast and 52 km northwest of the IISD-ELA, respectively. This region is sparsely populated (e.g., population density ranges from 4 to 71 people/km^2 in neighboring municipalities). Within the study area around the IISD-ELA, there is a lack of wastewater-treatment plants, there are few roads, and there is minimal development. However, the field station at the IISD-ELA hosts up to 100 visitors in the summer months, using many of the lakes for scientific activity and recreation. Past waste management at the IISD-ELA involved several decades of on-site incineration. Moreover, the area around the IISD-ELA is a well-known recreational site for camping and canoeing. For these reasons, we have classified this area as a semi-remote system. While the nine study lakes have minimal inputs of pollution overall, we classified them based on three levels of anthropogenic activity: "low," no research activity and no public access (Lake [L] 225, L378, and L628); "medium," extensive research activity but no public access (L114, L373, and L239); and "high," public access for recreational activities (Teggau, Highwind, and Hillock Lakes; Figure 1). All lakes were located within 15 km of the research station (Figure 1).

The study lakes varied in size and order (Table 1). Five are headwater lakes (L114, L239, L373, L225, and L378), while the remainder receive inputs from one (L628), three (Hillock), 34 (Highwind), and 35 (Teggau) upstream lakes or streams. Moreover, the surface areas of the high-activity lakes were larger than the rest, ranging from 7.72 to 13.30 km^2. In comparison, the surface area of the smallest lake, L225, was only 0.039 km^2.

Study design

Sediment, surface water, near-surface water (10-cm depth), and passive dry atmospheric deposition were collected within and around the IISD-ELA in July 2019 (Figure 1). We sampled near-surface water using a peristaltic pump in all nine study lakes. A surface manta trawl was also used on three of the lakes (L239, L373, L378). Sediment was collected at two lakes (L378 and L373), one with no past research activity (L378; low activity) and another with a history of extensive research activity (L373; medium activity). Passive dry atmospheric deposition was sampled at three sites that surround the entire sampling area (on the shores of L240 and L626 and between L164 and L165; Figure 1). Three replicates were taken at every site, resulting in six sediment samples, 27 near-surface water samples by in situ

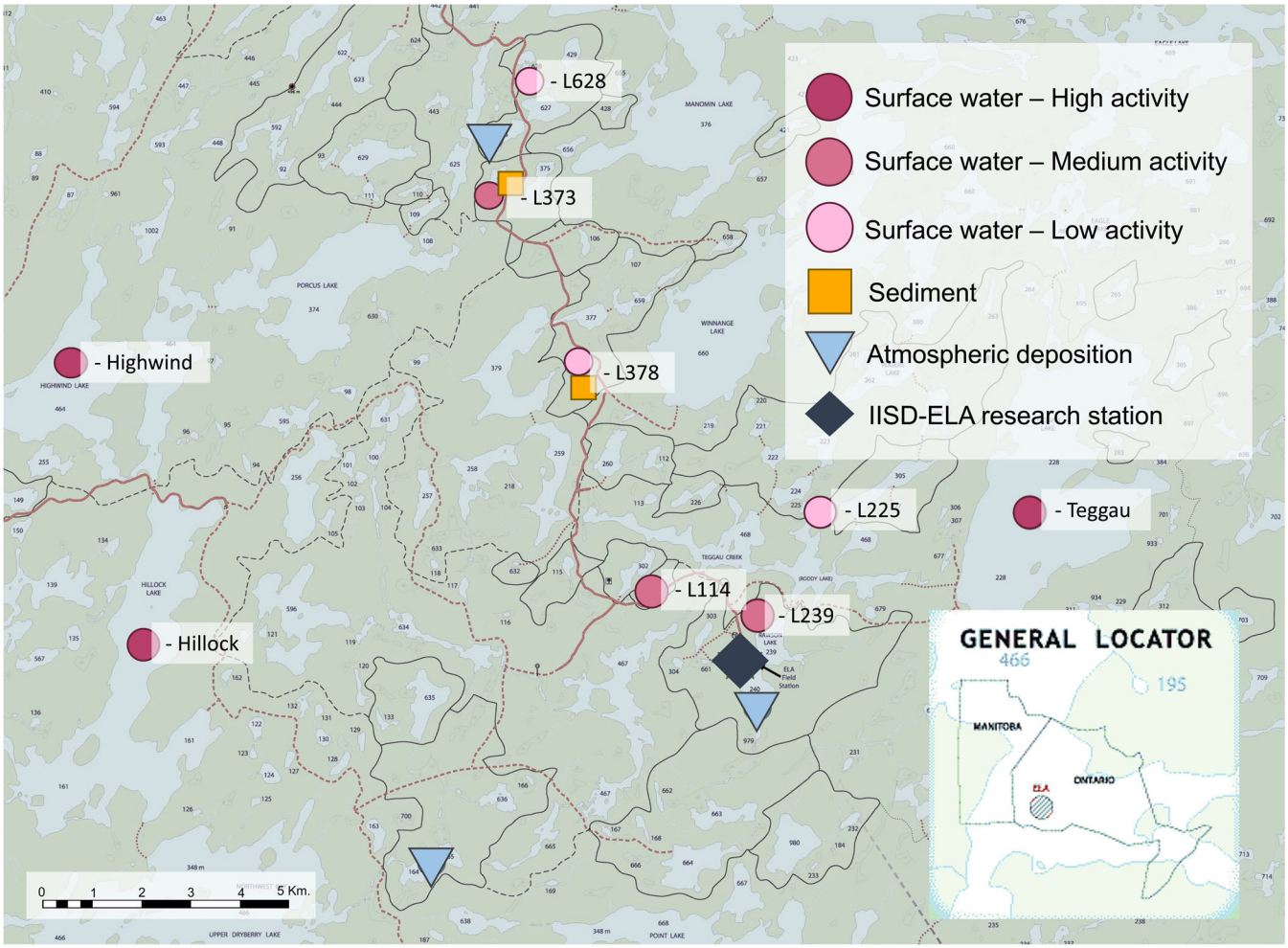

FIGURE 1: Map of sampling sites within and around the International Institute for Sustainable Development—Experimental Lakes Area (IISD-ELA). Three sample types were collected—sediment, surface water, and atmospheric deposition. Basemap from Fisheries and Oceans Canada.

filtering with a peristaltic pump, nine surface water samples by manta trawl, and nine atmospheric deposition samples. To control for procedural contamination, two sediment field blanks (one per lake), four surface water field blanks (one blank sample for each level of anthropogenic activity for the in situ filter with a peristaltic pump sampling method at L628, L239, and Teggau and one for the manta trawl sampling at L239), and one atmospheric deposition field blank (at L240) were sampled.

Field collection

Sediment. Due to resource constraints, we only sampled sediment from two lakes and prioritized near-surface and atmospheric samples to test our predictions. Lakes 378 and 373 were prioritized because we were planning future research within them. Sediment was collected from the deepest point of each lake using a gravity corer (Gravity National Lakes Assessment corer). Maximum lake depth was determined from

TABLE 1: Lake size and order characteristics (data from IISD-ELA)

Lake	Lake area (ha)	Volume (m^3)	Z_{max} (m)	Z_{mean} (m)	Watershed area (ha; including lake area)	Order
Hillock (161)	1007	Not available	117	Not available	2484	4
Highwind (464)	772	Not available	62	Not available	7991	35
Teggau (228)	1330	736,577,119	167	55.4	6481	36
114	12.1	207,486	5	1.7	58	1
239	54.1	6,169,186	31.5	11.4	393	1
373	27.4	3,107,474	21.2	11.3	83	1
225	3.99	47,608	2	1.19	30	1
628	21.8	1,093,565	19.2	5	216	2
378	24.3	1,811,354	18.2	7.5	136	1

Z = water depth.

bathymetric maps and measured in the field with a depth sounder (16.3 and 21.6 m for L378 and L373, respectively). Three 40-cm replicate cores (7 cm diameter) were collected per lake ($n = 3$, $N = 6$). Replicate cores were taken next to each other in both lakes. Cores were extruded in the field, and the top 0 to 5-cm and 30 to 35-cm layers of each core were collected in separate glass jars. These sediment layers were chosen based on Pb-210 radioisotope analysis from Mushet et al. (2018) and Jeziorski et al. (2014), where a 35-cm core represents 300 years for L373 and 204 years for L378. The deeper samples therefore represent preindustrial sediment. All equipment was rinsed three times with reverse osmosis (RO) water between samples or, if not available, with lake water. Field blanks were collected by running RO water through the core tube and extrusion stand and stored in a glass jar. Samples were stored at 4 °C until analysis.

Surface and near-surface water. We used two sampling methods for surface water collection. The first, which sampled the near-surface, was an in situ filtering method comprised of a peristaltic pump and an enclosed filter stack. This method was created to reduce exposure of the samples to external contamination, to increase the capture of smaller microplastics, and because the equipment was compact and therefore easily transportable to remote lakes where we needed to hike in with canoes. Using the in situ filter method meant limiting the amount of water collected in the field (maximum 20 L) and reducing the ability of the method to capture larger items. However, this method increases the likelihood of capturing smaller particles that may otherwise slip through the 333-μm mesh of the manta trawl. In comparison, the manta trawl filters upward of 30,000 L of water and has a large opening, thus is capable of collecting larger items. The manta trawl technique is a popular sampling method in marine and freshwater studies (Cole et al., 2011; Eriksen et al., 2013; Hidalgo-Ruz et al., 2012). However, the manta trawl is large and bulky and requires a motorboat, so this method was not feasible for some of the lakes in the present study. Therefore, we used a manta trawl on three of the nine lakes (L239, L373, and L378) as the second method to allow for comparisons with previous and future studies that use the trawl method.

For the in situ filtering method, we used a Geopump Peristaltic Pump Series II to collect 20 L of surface water per sample. The pump used mechanical peristalsis to pump the sample through Tygon® tubing. This protected the sample from exposure and contact with multiple surfaces. The pump tubing inlet (6 mm in diameter) was submerged 10 cm under the surface of the water, and the outlet was connected to a filter stack that released into a 20-L bucket. Four different filter sizes were used: 500, 213, 104, and 51 μm. We used stainless steel mesh filters that were precleaned in the lab by sonication in RO water. Each filter was enclosed in its own filter holder (Advantec MFS inline filter holder) during sample collection. Filter holders were connected with Tygon tubing. Before collection of each sample, lake water was pumped through the setup without the mesh filters for 1 min to prime the pump and clear any remaining residue from the previous sample. After this cleaning step, 20 L of surface water was pumped through the filter stack. Each filter was then individually placed in a clean Petri dish for later analysis. Three samples were collected per lake ($n = 3$, $N = 27$). Collections of replicate samples were spread across the lake—two on either end of the lake and one in the center—to account for variability in surface microplastic dispersion. For the larger lakes (Teggau, Highwind, and Hillock), this spread of collections was not possible, and samples were collected across one side of the lake. Water volume, collection site coordinates, and total sampling time were recorded for each sample. Field blanks were collected using the same procedure but with 4 L of RO water.

For the manta trawl method, we used a net with a 333-μm mesh size and 61 × 18 cm dimensions. The net was towed at the water surface behind a motorboat for approximately 10 min at a tow speed of 3 to 7 km/h. A flowmeter was attached in the center of the net mouth. After trawling, the outside of the net was rinsed down with RO water, and the sample was collected in the cod end of the net. The sample was then transferred to an amber bottle and preserved in 70% ethanol. Three samples per lake were collected ($n = 3$, $N = 9$) across three separate transects. Flow rate, total tow time, start and end coordinates, and minimum and maximum speeds were recorded. Any large pieces of debris collected by the net were picked out, rinsed with RO water, and placed in a Ziploc bag for later processing. The field blank sample was collected by dipping the outside of the net into the lake, followed by rinsing the outside of the net with RO water. Following the same procedure as the lake samples, the field blank was rinsed into an amber bottle and preserved in 70% ethanol.

Atmospheric deposition. We used a passive sampling method for passive dry atmospheric deposition (Hamilton et al., 2021). Samplers were glass Petri dishes (9 cm diameter) containing a plastic liner with double-sided adhesive tape on the bottom dish, where particles in the atmosphere land and remain trapped on the tape. The atmospheric samplers were placed on the ground in open areas, outside of tree cover. Petri dishes were opened, and the bottom dishes were left out for 5 consecutive days. Sampling surface area was 63.62 cm^2 for each sampler. Three atmospheric samplers were deployed at three sites spaced out 0.5 m apart around the sampling region ($n = 3$, $N = 9$). Collection site coordinates and total sampling time were recorded for every sample. A field blank was placed at one of the three sample collection sites (L240). While the atmospheric samplers were being deployed, the blank Petri dish was opened and handled in the same procedure. Once handling was completed, the blank sampler remained closed for 5 collection days. Researchers worked downwind of the samplers, and clothing worn during handling was recorded.

Laboratory analysis

Sediment samples were processed at Queen's University, and surface water and atmospheric deposition samples were processed at the University of Toronto. All spectroscopy was

performed at the University of Toronto. All particles suspected to be microplastics based on visual observations using a stereomicroscope, and before confirmation of a subsample with spectroscopy, are referred to as "microparticles." When reporting concentrations, microparticle numbers were spectroscopy-corrected for only confirmed microplastics.

Sample processing. Sediment samples (range 144–228 g wet wt) were dried in an oven at 50 to 55 °C, then processed through a wet peroxide oxidation (WPO) extraction based on the 2015 National Oceanic and Atmospheric Administration's protocol (Masura et al., 2015). This method uses 30% H_2O_2 with an iron catalyst and heat. The iron catalyst was made using 500 mL of 15% Fe(II)SO_4 and 3 mL of 98% $H_2SO_{4(aq)}$. On a hot plate on low heat (maximum 65 °C), 20 mL of iron catalyst was added to the sample, followed by 20 mL of 30% H_2O_2. Samples were heated to 40 °C and removed from the hot plate. Once sample temperature reached 45 °C, it was placed in an ice bath. After cooling, the sample was placed on the hot plate again, and another 20 mL of 30% H_2O_2 was added, following the heating and cooling process as before. This was repeated with up to 100 mL of 30% H_2O_2. Samples were then sieved (53 μm) and rinsed with RO water. If samples required further digestion, the WPO process was repeated. After digestion, the samples were sieved and rinsed back into a 600-mL beaker with 10% Alcojet detergent and left overnight. After WPO and the Alcojet soak, samples were sieved through a 53-μm sieve and rinsed with RO water back into beakers, then placed in a drying oven at 30 to 45 °C. After organic matter was digested, a $CaCl_2$ density separation was conducted. Each 600-mL beaker of sediment was filled with 300 mL of 1.4 g/mL $CaCl_{2(aq)}$, stirred for 3 min with a metal spoon, and left covered for 2 h. Any floating material was spooned out, and supernatant was decanted into a 1-L beaker. The remaining sediment went through a second round of density separation. After the second density separation, all floating debris and supernatant were mixed in the 1-L beaker, and a final 12 to 24-h density separation was conducted. Finally, the top layer and supernatant were vacuum-filtered onto a 20-μm cellulose filter.

For surface water collected using the in situ filtering method, each stainless steel mesh filter was sonicated in a 500 mL glass jar of RO water for 1 h, in an ultrasonic water bath. The cleaned filters were removed from the glass jars and rinsed with RO water. The larger size fractions (500 and 213 μm) were then sieved through a metal sieve of a similar pore size (500 and 212 μm, respectively). Sieves were rinsed with RO water into 250-mL glass jars. This sieving process was used to reduce the sample water volume for improved microplastic sorting and counting. The two smallest fractions (104 and 51 μm) were vacuum-filtered onto 47-mm polycarbonate filters with a pore size of 20 μm. This was conducted under a laminar flow hood to minimize procedural contamination. The polycarbonate filters were placed in a clean, glass Petri dish that contained a grid pattern on the bottom. The method of field collection did not pick up any visible pieces of organic matter, so no digestion step to remove organic material was needed.

In contrast, manta trawls pick up all floating debris on the surface water; and as a result, the samples contained more organic material than the in situ filter method. We used a WPO extraction to remove any cellulosic material in the samples similar to that used for sediment. The WPO process was repeated until an adequate level of organic material was digested. After digestion, the samples were sieved (212 μm), rinsed back into a beaker with 10% Alcojet detergent, and left overnight to remove any final residue. Samples were then sieved and separated into two size fractions: 500 and 212 μm. The samples were rinsed into clean 250-mL glass jars with RO water.

Microplastic quantification and sorting. All samples were visually sorted under a stereo zoom microscope. Categories of microparticles were limited to fiber, fiber bundle, film, fragment, foam, and sphere based on the categories described in Rochman et al. (2019). Sediment samples were observed directly on the filter under a Motic SMZ-171 Series microscope, and all microparticles were counted and their color and shape recorded. Particles were left on the filter, and pictures and measurements were taken using ImageJ software (Ver 1.52Q).

For all surface water samples, each size fraction was visually sorted under an Olympus SZ61 (×6.7–×45) microscope. The two smallest fractions were counted and sorted directly on the polycarbonate filter, following a zigzag pattern along the grid. We used a wet sort protocol for the larger fractions. A small portion of the wet sample was spooned onto a clean Petri dish that contained a grid pattern on the bottom, then visually sorted under the microscope following a zigzag pattern. Once a small portion was counted, this was rinsed into a discard jar, and a new portion of the sample was counted. This was repeated until the entire sample was sorted and counted. Microparticles were picked and categorized based on color and shape and placed in a separate clean Petri dish lined with a clear plastic liner and double-sided tape. Every particle was labeled, and the color and category were recorded. Next, all microparticles were photographed and measured using ToupView software (Ver 3.7).

Atmospheric samples were visually sorted directly on the sampler under an Olympus SZ61 (×6.7–×45) microscope. Particles that were identified as suspected microplastic were marked directly in the sampler and their color and category recorded. All microparticles were photographed and measured using ToupView software.

Microplastic identification. Polymer identification of microparticles in sediment was performed by micro–Fourier-transform infrared spectroscopy (micro-FTIR), while that in surface water and atmospheric deposition was performed by micro-Raman spectroscopy.

Micro-FTIR. At least 10% of microparticles in each color/shape combination (e.g., blue fiber, black fragment) from each sediment sample were subsampled for analysis by micro-FTIR, barring some exceptions. Due to difficulties with isolating particles, some could not be characterized. In addition, one sample from L378 was not analyzed for chemical composition.

Thus, a total of 14% ($n = 31$) of all microparticles in sediment were analyzed (after blank subtraction by color and category; see below, section on quality assurance/quality control [*QA/QC*]) with a Nicolet iN10 Infrared Microscope (Thermo Fisher Scientific) in attenuated total reflection (ATR) mode (×15 objective, 0.7 numerical aperture), with a germanium ATR crystal, following the protocol of De Frond et al. (2021). In brief, all spectra were obtained using the cooled mercury cadmium telluride detector with a resolution of 4 cm^{-1} and 32 co-added scans. Prior to analyzing each particle, the crystal was cleaned with ethanol, and a background spectrum was taken. The interferometer was realigned before use at the start of each working day. Spectra were recorded in percentage transmission with a spectral range of 4000 to 675 cm^{-1}. The aperture was manually adjusted to fit the width of each particle. The spectral library used was Opus and the μ-ATR-FTIR Spectral Library of Plastic Particles (Munno et al., 2020). A spectral match generally fell between 80% and 98%, with a few exceptions made based on judgment of spectral features.

Micro-Raman. All surface water and atmospheric deposition samples were blank-subtracted (see below, *QA/QC*) prior to chemical identification by micro-Raman spectroscopy. We did this because we did spectroscopy on only a subsample of particles. Following blank correction, at least 10% of particles in each color/shape category from each sample were randomly selected for chemical identification. This led to a total of 38% ($n = 161$) of microparticles in surface water collected by the in situ filter method, 43% of microparticles ($n = 60$) collected by the manta trawl method, and 62% ($n = 20$) of microparticles in atmospheric deposition being analyzed. Spectra were collected with a Horiba Raman Xplora Plus using LabSpec6 software. We used 785-nm (range 50–2000 cm^{-1}) or 532-nm (range 50–4000 cm^{-1}) lasers with a ×100 long working distance microscope objective. We used a filter ranging from 0.1% to 100%; gratings of 600 or 1200 grooves/mm; up to 8 s for acquisition time; 2, 4, 6, or 8 accumulations; a confocal hole diameter of 100 or 300 mm; and a confocal slit width of 50 or 100 mm. When acquiring spectra, parameters were optimized to inhibit poor resolution, fluorescence, and other issues. This resulted in particles having a different suite of parameters (e.g., the 785 nm would provide a better spectrum for darkly colored particles). The spectral library used was BioRad KnowItAll Raman Spectral Library and the Spectral Library of Plastic Particles—Environmental (Munno et al., 2020). A spectral match generally fell between 80% and 98%, with a few exceptions made based on judgment of spectral features.

Microparticles were assigned the following material categories based on spectroscopy database matches. Confirmed "Plastic" includes all common plastic polymers (e.g., polyethylene [PE], polypropylene, polyamide, polyethylene terephthalate [PET], acrylic, and polystyrene). "Anthropogenic synthetic" includes particles where an additive indicating synthetic origin was identified but the underlying polymer could not be identified (e.g., plasticizers). "Anthropogenic cellulosic" are particles containing cellulose and pigments or additives indicating anthropogenic origin. "Anthropogenic unknown" includes particles where an anthropogenic dye is detected but the underlying material is not, and it is unclear whether the underlying particle is plastic or not. "Unknown cellulosic" includes particles containing cellulose but whose origin could not be determined. Particles in this category may not be anthropogenic. "Unknown" particles could not be identified due to photodegradation, fluorescence, or a lack of Raman signal, and thus also may not be anthropogenic. "Natural" categories include inorganic (e.g., minerals) and organic (e.g., wool, chitin) materials. Reported microplastic concentrations were adjusted based on spectroscopy by multiplying final counts by the percentage of confirmed plastic for each sample type. Shallow and deep sediments were corrected separately. Characteristic results (e.g., shapes, sizes, material types) include all particles identified by visual microscopy and are referred to as "microparticles."

QA/QC. All equipment was washed with soap and triple-rinsed with RO or deionized water, and surfaces were wiped with RO water and/or 70% ethanol. Cotton lab coats were worn in the lab and when handling the samples. High-efficiency particulate air filtration systems were present in all laboratories where samples were analyzed. Samples were covered with lids or aluminum foil when not in use. Field blanks were treated as normal samples and thus followed all laboratory procedures. The total microparticle count of the environmental samples was blank-subtracted based on the average total microparticles of each color and shape found in blank samples. Sediment, in situ filter, manta, and atmospheric blank samples contained a mean of 21 ± 7.1, 25 ± 7.4, 14, and 4 microparticles, respectively. To report conservative numbers, the average number of each microparticle type in blanks was rounded up and subtracted from each sample and size fraction (i.e., 0.3 green fibers rounded up to 1 green fiber). After blank subtraction, the numbers of total microparticles were 225 of 376 particles for sediment, 424 of 891 particles for in situ filtering, 141 of 209 particles for manta trawl, and 32 of 53 particles for air. Blank samples were also analyzed for polymer type. For sediment blanks, 26% ($n = 11$) of particles were analyzed by μ-FTIR; 28% ($n = 21$) of particles from in situ filter blanks, 36% ($n = 5$) of manta trawl blanks, and 25% ($n = 1$) of atmospheric deposition blanks were identified using micro-Raman. See Supporting Information for more details on blank sample results and Supporting Information, Tables S1 and S2.

Data analysis. Microparticle concentrations are reported in particles per kilogram for sediment samples, particles per liter or particles per cubic meter for surface water (depending on collection method), and particles per square meter per day for atmospheric deposition (see Supporting Information, Table S3, for relevant equations).

To test for a difference in microplastic concentration between anthropogenic activity levels in surface water, we performed a two-factor nested analysis of variance (ANOVA; factor 1 = anthropogenic activity level; factor 2 = lake—nested within activity level). Microplastic count data were log-transformed (log10), and a Shapiro-Wilks test for normality ($p = 0.24$) and a Bartlett test for homogeneity ($p = 0.52$) were not significant.

We used Bray-Curtis-based nonmetric multidimensional scaling (NMDS; Vegan package; Oksanen et al., 2019) to assess any similarity among sample types (i.e., between surface water activity levels and atmospheric samples and between the two surface water collection methods and atmospheric samples) in relation to microparticle composition by shape and material type. We did not assess differences among manta trawl samples or among atmospheric deposition sites because there were not enough sites across differing levels of anthropogenic activity and because our aim was to collect ambient concentrations. All analyses were performed in R statistical software (Ver 3.6.1).

RESULTS AND DISCUSSION

Microplastic concentrations in sediment, near-surface water, surface water, and air

Sediment. Microplastics were present in all sediment samples, ranging in concentration from 36 to 971 microplastics kg^{-1} (dry wt) in shallow sediments (0–5 cm depth) and 6 to 320 microplastics kg^{-1} in deep sediments (30–35 cm depth). Mean ± standard deviation (SD) for shallow and deep sediments were 551 ± 354.42 and 177 ± 103 microplastics kg^{-1}, respectively. Shallow sediment in the medium-activity lake, L373, contained 263 ± 213 microplastics kg^{-1}, while deep sediment contained 118 ± 97 microplastics kg^{-1}. Higher concentrations were observed in L378 (low activity) at 839 ± 138 microplastics kg^{-1} in shallow sediment and 236 ± 81 microplastics kg^{-1} in deep sediment (Figure 2A; see Supporting Information, Table S4, for concentrations in each sample).

The microplastic levels observed in the present study are similar to those in freshwater lakes in Canada and the United States, such as Lake Ontario (87.2–615.6 microplastics kg^{-1}; Corcoran et al., 2015) and Lake Mead National Recreation Area (87.5–1010 microplastics kg^{-1}; Baldwin et al., 2020). While these lakes experience higher loads of anthropogenic impacts from tourism or population centers, these studies also had a larger limit of detection (500 and 355 µm, respectively) compared with the one used in our study (53 µm), which would have lowered the number of microplastics reported. The mean microplastic levels of ELA lakes are still comparable to those in remote lakes of the Tibetan Plateau (mean 544.62 ± 297.99 microplastics kg^{-1}), measured using a more comparable lower size limit of 50 µm (Liang et al., 2022). However, the levels of microplastics in the Tibetan Plateau had a much larger range, from 17 to 2643 microplastics kg^{-1} (Liang et al., 2022). Many factors may affect microplastic concentrations among lakes, including sediment type, lake size, microplastic sources, methodology, and sediment deposition rate (Free et al., 2014; Liang et al., 2022; Yang et al., 2021).

Contrary to our expectations, microplastics were present in preindustrial deep sediments of both lakes, although at concentrations lower than those in shallow sediments (Figure 2A). There was a higher proportion of confirmed microplastic relative to overall microparticles in recent sediment (33%) compared with confirmed microplastic in preindustrial sediment (11%; Supporting Information, Figure S1B). A similar pattern was observed in rural lakes of Ontario, Canada, as well as an urban lake in the United Kingdom (Turner et al., 2019; Welsh et al., 2022b). This observation has some possible explanations. First,

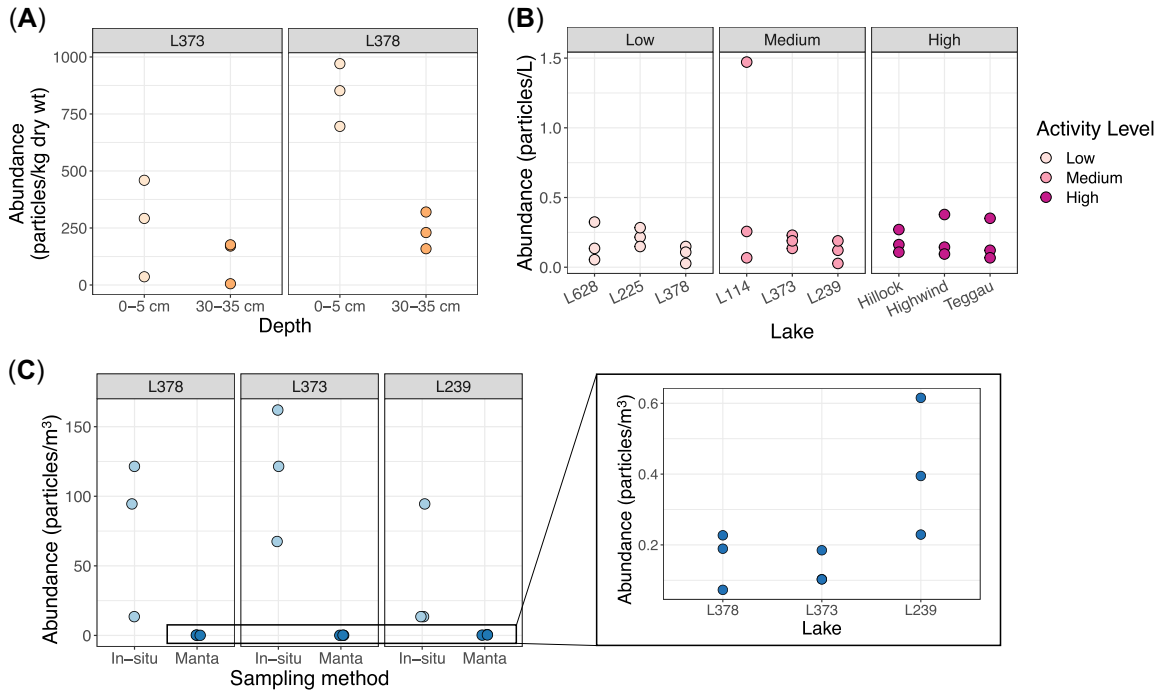

FIGURE 2: Concentration of microplastics (i.e., spectroscopy-corrected values) in sediment and surface water. Sediment was collected in two lakes (**A**) and surface water using the in situ filter across nine lakes of three anthropogenic activity levels (**B**). As well, surface water sampling methodology (manta trawl vs. in situ filter) was compared in three lakes (**C**). For all, each data point represents one replicate sample.

disturbances, both physical (e.g., previous sediment coring) and biological (e.g., bioturbation), may account for the presence of microplastics within sediment layers predating industrial plastic production (Coppock et al., 2021). Second, deep sediments of ELA lakes are soft with a high water content, so downward migration of heavy polymers by gravitational forces is not implausible (O'Connor et al., 2019).

Sizes of microparticles in sediments ranged from 42 to >5000 μm, with the highest proportion of particles in the 200 to 300-μm bin (Figure 3A). Most microparticles in sediment were fibers (76%), followed by fragments (6%), films (6%), fiber bundles (5%), foams (4%), and spheres (3%; Figure 3B; see Supporting Information, Figure S1A, for a breakdown by sediment depth). The large proportion of fibers is consistent with other freshwater sediments, based on a review of 38 studies (Yang et al., 2021). Of all microparticles ($n = 31$) that were identified by μ-FTIR, 35% were unknown, 23% were anthropogenic cellulosic, 20% were unknown cellulosic, 20% were plastic, and 3% were natural (organic; Figure 3C). Six particles were identified as plastic and consisted of one poly(ethylene-co-vinyl acetate) fiber, one PET/polyester fiber bundle, one PE fragment, one ethylene-vinyl acetate fiber, one PET/polyester fiber, and one poly(methyl methacrylate) fiber (Figure 3D). See Supporting Information, Figure S1, for a breakdown of material types between shallow and deep sediment samples.

Near-surface water. Across all sampled lakes, concentrations of microplastics collected through in situ filtering ranged from 0.03 to 1.5 microplastics L^{-1} (mean 0.2 ± 0.3 microplastics L^{-1}; Figure 2B; see Supporting Information, Table S5, for sample breakdown). However, most concentrations were <0.4 microplastics L^{-1} (Figure 2B; Supporting Information, Table S5). The highest concentration was recorded at L114, near an ongoing mesocosm experiment that used several plastic nets and equipment. The mean concentration observed was comparable to concentrations observed in headwater lakes in another semi-remote location in Ontario, within the Muskoka–Haliburton region (1.02–2.39 microplastics L^{-1}; Welsh et al., 2022b). These levels are typical for a site with few to no upstream inputs that does not receive wastewater or sewage overflow effluent (Talbot & Chang, 2022). In comparison, concentrations in lakes in populated areas can reach as high as 25.8 microplastics L^{-1} (Su et al., 2016; Wang et al., 2021).

Fibers were the most common particle shape (78%). Other categories included fragments (18%), films (3%), fiber bundles (0.5%), and spheres (0.5%; Figure 3B). The material types of subsampled particles ($n = 161$) included 31% anthropogenic cellulosic, 27% plastic, 19% unknown cellulosic, 12% unknown, 4% anthropogenic unknown, 4% natural (organic), 2% natural (inorganic), and 1% anthropogenic synthetic. Polyester/PET and acrylic were the most common

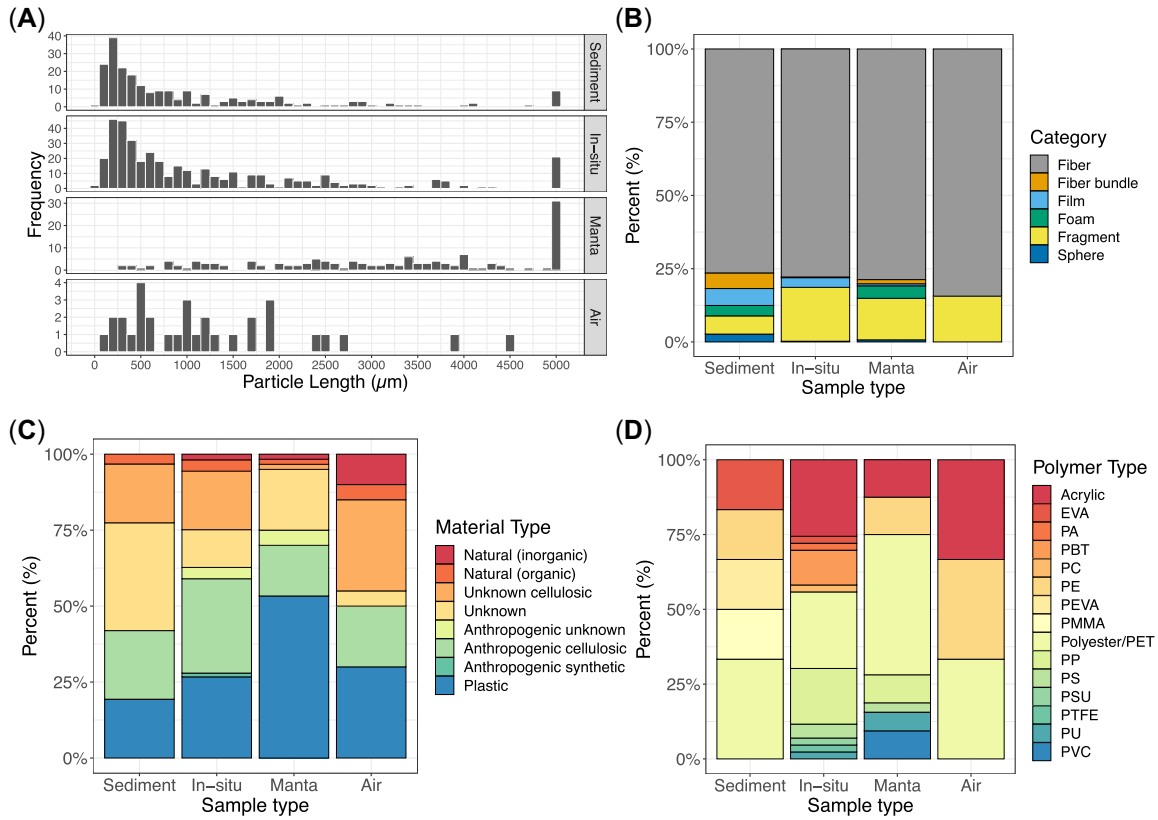

FIGURE 3: Characteristics of microparticles across all studied matrices (sediment, water, air) and sampling methods (i.e., surface water sampled via in situ filter and manta trawl) by size (**A**), shape (**B**), material type (**C**), and plastic polymer type (**D**). See Supporting Information, Figure S1, for a depiction of morphologies and material types between sediment depths. EVA = ethylene-vinyl acetate; PA = polyamide; PBT = polybutylene terephthalate; PC = polycarbonate; PE = polyethylene; PEVA = poly(ethylene-co-vinyl acetate); PMMA = poly(methyl methacrylate); PET = polyethylene terephthalate; PP = polypropylene; PS = polystyrene; PSU = polysulfone; PTFE = polytetrafluoroethylene; PU = polyurethane; PVC = polyvinyl chloride.

polymer types, though nine types were identified overall (Figure 3D).

Surface water. Concentrations of microplastics in surface water collected by manta trawl ranged from 0.1 to 0.6 microplastics m^{-3} (mean 0.2 ± 0.2 microplastics m^{-3} or 0.0002 ± 0.0002 microplastics L^{-1}; Figure 2C; see Supporting Information, Table S6, for sample breakdown). In comparison, in situ filtered samples from the same lakes had concentrations of 27–230 microplastics m^{-3} (mean 131 ± 70 microplastics m^{-3} or 0.1 ± 0.07 microplastics L^{-1}; Figure 2C). The large difference in concentrations between the two collection methods is reflected in the particle sizes. Manta trawl–collected particles ranged from 270 to >5000 μm, and most were >5000 μm. In contrast, the in situ filtering method captured much smaller particles, reaching as low as 39 μm; and most were <1000 μm (Figure 3A). This is in line with previous reports that show increasing concentrations with decreasing particle size and is expected because of the larger mesh size of the manta trawl (333 μm; Hung et al., 2021; Lindeque et al., 2020). When only the upper size fractions (213 and >500 μm) are compared, concentrations from in situ filtered samples were still greater than those of the manta trawl, ranging from 14 to 162 microplastics m^{-3} (mean 78 ± 55 particles m^{-3}). This could be because the in situ method is more efficient at capturing fibers compared with the manta net, where narrow fibers will more easily slip through the higher surface area and pore size of the trawl mesh.

Moreover, the patterns observed among the lakes were not consistent between the two methods. According to the results of the manta trawl method, L239 had more microplastics than the other two lakes, while the in situ filtered method suggests that L373 contained higher levels (Figure 2C). This may be due to the different sizes of microparticles captured and/or the differences in volume sampled, and more work should be done to determine whether both sampling methods were representative (e.g., by taking duplicate samples and assessing the variability between them). Still, microparticle morphologies from manta trawl samples were akin to those from in situ filtered samples. For instance, most were fibers (79%), followed by fragments (14%), films (1%), fiber bundles (1%), and spheres (1%; Figure 3B). Of the 60 analyzed particles, 53% were plastic, and PET/polyester was the most common polymer (47%; Figure 3C,D). Other material types consisted of 20% unknown, 17% anthropogenic cellulosic, 5% anthropogenic unknown, 2% unknown cellulosic, 2% natural (inorganic), and 2% natural (organic; Figure 3D). The higher proportion of plastics in manta trawl samples compared with the in situ filtered samples may be due to the organic matter digestion step, which is known to degrade natural and cellulose-based particles (Treilles et al., 2020).

Atmospheric deposition. The level of microplastics in atmospheric dry deposition ranged from 0 to 0.7 microplastics m^{-2} d^{-1} (mean 0.4 ± 0.2 microplastics m^{-2} d^{-1}; Figure 4; Supporting Information, Table S7). In comparison, low levels of deposition were observed in an area of minimal urbanization in the United Kingdom (1.1–3.2 microplastics m^{-2} d^{-1}; Stanton et al., 2020). Another study in a semi-remote region of Ontario, Canada, reported a mean of seven microplastics m^{-2} d^{-1} (range 4–9 microplastics m^{-2} d^{-1}; Welsh et al., 2022a). Both of these studies sampled bulk deposition. Where Stanton et al. (2019, 2020) suggested no influence from rainfall on particle counts across the year-long sampling campaign, Welsh et al. (2022a) found there was an association between rainfall and particle count. One study in the Arctic that used similar dry deposition samplers to those in the IISD-ELA sites reported 2433 ± 1235 anthropogenic particles m^{-2} d^{-1}, which is many orders of magnitude higher; but these numbers were not corrected for confirmed plastics (Hamilton et al., 2021). Differences in methodology, wind speed, precipitation, elevation, sampling duration, and climate all affect reported microplastic concentrations, making it difficult to compare to other studies (Allen et al., 2019; Leonard et al., 2024; Wright et al., 2020). The IISD-ELA is well characterized by boreal forests, and the presence of dense vegetation can affect particle deposition and emission patterns (Barwise & Kumar, 2020; Leonard et al., 2024). Moreover, we sampled once in the summer season, and deposition rates may vary across time; therefore, these deposition rates may not be representative of long-term trends (Allen, Allen, Le Roux, et al., 2021; Stanton et al., 2020; Sun et al., 2022).

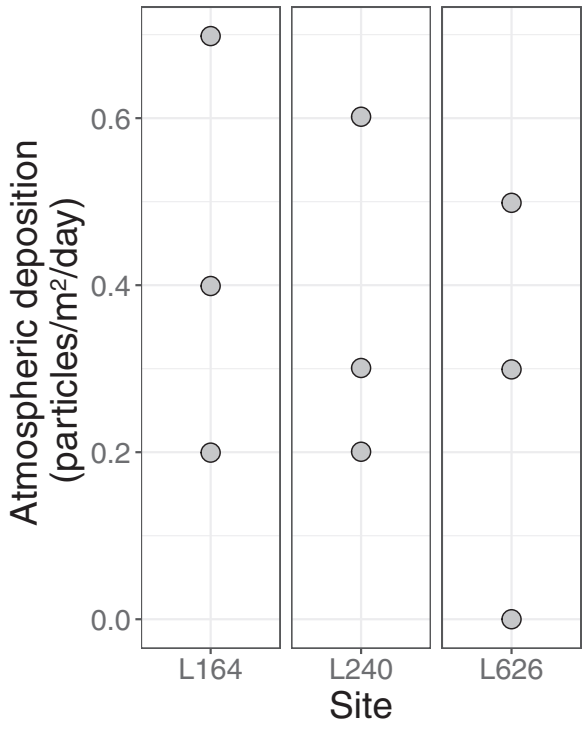

FIGURE 4: Deposition rate of microplastics in atmospheric deposition at three sites sampled across July 20–25, 2019.

The lengths of microparticles ranged from 139 to 4528 μm (Figure 3A). Only fibers (85%) and fragments (16%) were present in atmospheric deposition (Figure 3B), and all fragments were <400 μm. Material types were in similar proportions to surface water. Of the 20 analyzed particles, 30% were plastic and 30% unknown cellulosic; the remaining particles were 20% anthropogenic cellulosic, 10% natural (inorganic), 5% natural

(organic), and 5% unknown (Figure 3C). Three polymer types of equal proportion were identified—acrylic, polyester/PET, and PE (Figure 3D)—all of which are common polymers in clothing (Carr, 2017). Bulk and wet deposition from another area in Ontario, Canada, also had a high proportion of fibers, as did dry deposition from the Canadian Arctic (Hamilton et al., 2021; Welsh et al., 2022a). The large fiber composition is also similar to atmospheric microplastics in high-density urban areas (Cai et al., 2017; Dris et al., 2015; Wright et al., 2020). Notably, a study across the Southern Ocean found that fibers traveled longer distances than fragments (Chen et al., 2023).

Sources and transport of microplastics

Relationship between anthropogenic activity and microplastic concentrations. Microplastic concentrations in surface water did not increase with increasing anthropogenic activity. Low-, medium-, and high-activity lakes had mean ± SD concentrations of 0.2 ± 0.1, 0.3 ± 0.4, and 0.2 ± 0.1 microplastics L^{-1}, respectively. There was no significant difference in microplastic abundance between activity levels ($F_{2,18} = 0.27$, $p = 0.78$), and the variance among lakes within an activity level was not significant ($F_{6,18} = 0.89$, $p = 0.52$; see Supporting Information, Table S8 for ANOVA model results; Figure 2B). It is worth noting that lake size and order varied among the lakes, which may affect particle concentrations (Eerkes-Medrano & Thompson, 2018; Nava et al., 2023; Welsh et al., 2022b). High-activity lakes had greater surface areas and more upstream inputs than lower-activity lakes (Table 1). Nonetheless, the variance in microplastic levels among lakes was minimal. Moreover, Welsh et al. (2022b) also found no correlation between microplastic abundance and lake size. Overall, these results suggest that direct anthropogenic inputs are likely not the main pathway of microplastic contamination to this region and that atmospheric deposition plays a substantial role. Generally, other studies have found that higher microplastic loads correlate with urban centers and human activity (Grbić et al., 2020; Nava et al., 2023). Yet for a semi-remote site that lacks urban centers, industrial sites, and wastewater effluent such as the IISD-ELA, the primary pathway of plastics is likely from nonpoint sources, including the atmosphere. Other studies on lakes with similar land use also concluded that atmospheric transport was the main contributor of microplastics (Dong et al., 2021; Welsh et al., 2022b), highlighting the significance of the atmosphere as a pollution pathway.

Similarities and differences across matrices. Because we sampled near-surface water from all study lakes, we used this matrix to compare microparticle composition with that of atmospheric deposition via NMDS. The types of microparticles in near-surface water and atmospheric deposition were similar in shape but less so in material type (Figures 3B,C and Figure 5A,B). Use of NMDS shows an overlap in microparticle compositions between atmospheric deposition and surface water when categorized by particle shape (Figure 5A) but distinct groups when categorized by material type (Figure 5B). For both, fibers were

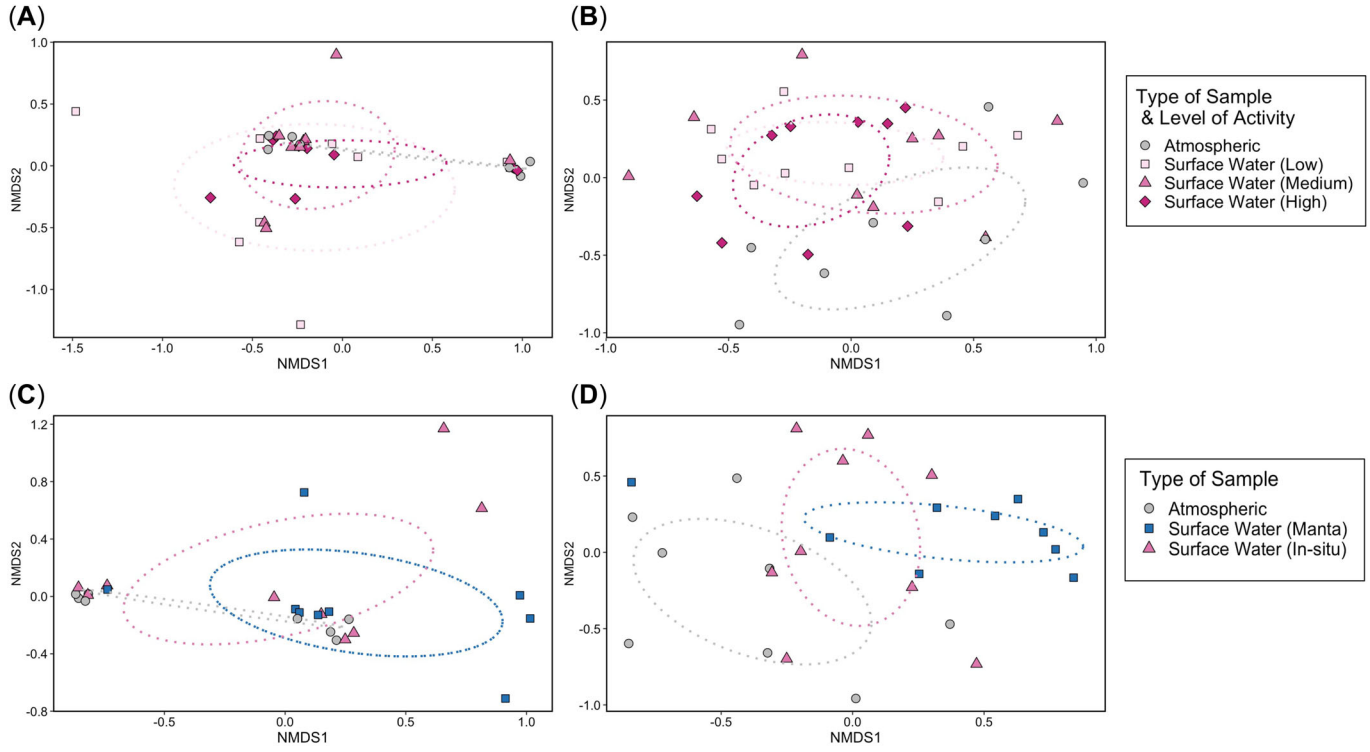

FIGURE 5: Comparison of microparticle composition in different matrices and sampling methods by nonmetric multidimensional scaling using Bray-Curtis dissimilarity. (**A**) Microparticle morphologies (stress = 0.05) and (**B**) material type (stress = 0.19) in near-surface water across differing anthropogenic activity levels and in atmospheric deposition. (**C**) Microparticle morphologies (stress = 0.08) and (**D**) material type (stress = 0.17) in two surface water collection methods and in atmospheric deposition. NMDS, nonmetric multidimensional scaling.

the predominant particle shape (Figure 3A), whereas material type was more diverse in surface water (Figure 3C). In addition, microparticle compositions differed between manta trawl–collected surface water and atmospheric deposition (Figure 5C,D). Because the manta trawl collection method is characterized by larger particles that are unlikely to be transported far distances (Lawrence & Neff, 2009), the larger particles captured are likely coming from elsewhere, such as past and current research activity on the lakes or surface runoff from anthropogenic activities in their watersheds (e.g., all-terrain and motor vehicles, forestry operations, and recreation). Still, these differences may also be due to variation in postdepositional journeys within water, land, and air. Vertical transport (e.g., biofouling, sediment aggregation, egestion in fecal pellets) and horizontal transport (e.g., runoff, winds, waves, currents) both affect particle fate even if atmospheric deposition was the initial source (Cole et al., 2016; Eerkes-Medrano & Thompson, 2018; Kowalski et al., 2016). Lastly, atmospheric samples were collected across a 5-day period, whereas microparticles in surface water could be retained within the lakes over a much longer period.

Despite the differences discussed above between water and atmospheric deposition samples, acrylic, polyester, and cellulose fibers were found to make up the majority of material types for these matrices (Figure 3C). Microfibers often come from the shedding of clothing and other textiles, and laundry washing and drying are a major pathway for fibers to enter the environment (Browne et al., 2011; Kapp & Miller, 2020; Pirc et al., 2016). As a result, fibers comprise a large portion of microplastics in atmospheric samples from urban sites, making urban centers an emission source to farther locations (Dris et al., 2015; Wright et al., 2020). However, the long-range transport of microplastics is largely unknown. Allen et al. (2019) reported airborne microplastics traveling up to 95 km. Microplastics <20 μm can travel through the free troposphere, while larger particles are likely transported from regional sources (Allen, Allen, Baladima, et al., 2021; Brahney et al., 2021). Accordingly, the city of Kenora, located 52 km from the IISD-ELA lakes, could be an atmospheric emission source to these remote lakes; but other areas may also be involved. For instance, the Trans-Canada Highway is located within 30 km from the lakes and could also act as an emission source.

CONCLUSION

In the present study, we show that microplastics are present in the sediment, water, and atmospheric dry deposition of semiremote boreal lakes in northwestern Ontario, Canada. In line with our first prediction, concentrations were relatively low compared with heavily populated regions and similar to those reported in other semi-remote locations. In contrast to our other predictions, microplastic concentrations were not correlated with anthropogenic activity, and morphologies were similar to atmospheric deposition. The lack of association with local levels of anthropogenic activity and the similarity in the morphologies of microplastics in surface water and atmospheric deposition suggest that atmospheric transport is the main pathway of plastic to this region. The present study provides further evidence of atmospheric cycling as an important component of the plastic cycle—transporting anthropogenic particles away from their source. In future, a more holistic view can be gained when multiple environmental matrices are studied because each one can inform sources and pathways of contamination. Moreover, exploring key questions surrounding the global transport of microplastics and the associated risks, such as harm to wildlife and interactions with microbial communities and nutrient cycling, is required to further inform mitigation of this contaminant.

Supporting Information—The Supporting Information is available on the Wiley Online Library at https://doi.org/10.1002/etc.5832.

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Conflict of Interest—The authors declare no conflicts of interest.

Author Contribution Statement—**Hayley K. McIlwraith**: Data curation; Formal analysis; Investigation; Methodology; Visualization; Writing–original draft. **Minoli Dias**: Data curation; Investigation; Methodology; Writing–review & editing. **Diane M. Orihel, Michael D. Rennie, Jennifer F. Provencher, Chelsea M. Rochman**: Conceptualization; Funding acquisition; Investigation; Methodology; Project administration; Supervision; Writing–review & editing. **Anna L. Harrison**: Methodology; Project administration; Supervision; Writing–review & editing. **Matthew J. Hoffman**: Conceptualization; Funding acquisition; Investigation; Methodology; Project administration; Writing–review & editing.

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