Abundance and Distribution of Microplastics in Lake Scugog Catchment, Ontario

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Abstract

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Plastic pollution is a growing concern, owing to its durability, ubiquity, and potential health impacts. The overall objective of this study was to assess the abundance and distribution of microplastics within Lake Scugog catchment, Ontario. This was fulfilled through two tasks (i) the development of a microplastic particle budget for the lake catchment, and (ii) the determination of the dry deposition of atmospheric microplastics in Port Perry, Ontario. The total input of microplastics into Lake Scugog (atmospheric deposition and stream inflow) was 2491 $x10^6$ mp/day, while the output (lake outflow and sedimentation) was 1761 $x10^6$ mp/day, suggesting that 29% of inputs were retained in the lake. The dry deposition of microplastics in Port Perry was 1257 mp/m^2/day, which was high when compared to bulk deposition (37 mp/m^2/day) in the same area. By quantifying the major pathways of microplastics better management techniques can be implemented.

Keywords: Plastic pollution, microplastics, particle budget, catchment, dry deposition, Ontario

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Chapter 1: General Introduction

1.1 Plastics

Plastics are generally defined as synthetic organic polymers derived from petroleum sources (Anderson et al., 2016; Derraik, 2002). The first modern plastic material was developed in 1907, with mass production exponentially increasing since the 1940s (Cole et al., 2011). In 2017, 348 million tonnes of plastic were produced, which is predicted to double by the year 2025 (Ainali et al., 2021). Plastics have become popular for many reasons including their strength, durability, light weight, and low cost (Derraik, 2002; Cole et al., 2011). It is these same characteristics (i.e., strength and durability) that cause plastics to be highly resistant to degradation and to be a potential environmental pollutant. In practice, if plastic was properly managed, at the end of it's life-cycle it would be recycled or combusted to generate energy, resulting in no plastic being released into the environment (Barnes et al., 2009). However, due to improper human behaviour, e.g., lack of infrastructure, inefficient disposal, and littering or loss during transport, plastics are released into the environment.

The many societal benefits of plastic (durability, low cost, etc.), which have driven its popularity, are directly related to the increasing concern of environmental plastic pollution (Cole et al., 2011). A large contributing factor to plastic pollution is the increase of single-use throw-away plastic items such as packaging material. Between 1950 and 2015, 448 million metric tons of plastic was produced, 40% of this was designed for single use (Walker et al., 2021). Some plastics are recycled (9%) or incinerated (12%) after they are no longer in use, however the majority of plastics end up in a landfill or the natural environment (79%), such as water bodies where they can persist for centuries (Cole et al., 2011; Barnes et al., 2009, Leberon, 2018; Walker et al., 2021).

Plastic pollution has been widely observed in both marine and terrestrial environments (Barnes et al., 2009) as well as in the atmosphere (Cai et al., 2017; Anderson et al., 2016; Prata et al., 2020). Further, plastic has buoyancy (owing to its low density), which allows it to be dispersed over long distances with the assistance of wind or wave action (Derraik, 2002; Lebreton et al., 2018). One of the reasons plastics are found commonly across the globe is because of this ease of transport. Ultimately, plastics that are in the natural environment for an extended period of time begin to degrade creating microplastics (Derraik, 2002). Microplastic pollution is one of the many anthropogenic contaminants that will persist in the environment for centuries even if plastic waste is stopped immediately (Barnes et al., 2009).

1.2 Microplastics

A microplastic is generally defined as a plastic particle less than 5 mm (< 5000 µm; Anderson et al., 2016), where primary microplastics are manufactured at a microscopic size for numerous purposes including, personal care products, air-blasting media, medicinal, and pellets used in the initial stage of manufacturing (Cole et al., 2011; Anderson et al., 2016; Ainali et al., 2021; Browne et al., 2007). In contrast, a microplastic that is formed due to the fragmentation of larger plastic particles is defined as a secondary microplastic; fragmentation can be a consequence of physical, biological, or chemical degradation, e.g., plastics can break-down from ultraviolet radiation (sunlight), or under wave action and abrasion in the marine environment (Cole et al., 2011; Browne et al., 2007).

Microplastics can be further classified based on their appearance (e.g., shape, colour, and composition). Particle shape categories generally include fibers, fragments, films, beads, and foams, with fibers and fragments being the most common microplastics observed in environmental samples (see Appendix Table A1.1; Lusher et al., 2020). Fibers are classified as

having a longer length than width (<50–75 µm; 0.05–0.075 mm) and are generally created by the fragmentation of larger textile materials. In contrast, fragments are generally formed by the degradation of larger particles and have an angular structure. Films are sometimes classified as a subcategory of fragments, the difference being films are generally more flexible than fragments and have two dimensions much greater than the third as films are generally very thin (Lusher et al., 2020). The microplastic polymer composition is directly related to the source. Generally, polyester (or polyethylene terephthalate), polyamide (or nylon), polypropylene, and polystyrene are some of the most common types of plastic found in environmental samples (Welsh et al., 2022; Felismino et al., 2021; Wright et al., 2020; Abbasi & Turner, 2021; Allen et al., 2019).

1.3 Effects of Microplastics

In the environment it has been suggested that microplastics can harm organisms both directly and indirectly (Xia et al., 2020). There is limited evidence through lab-based experiments that organisms can be directly affected by ingestion of microplastics, which can lead to intestinal obstruction, and reduced growth and reproductive rates. Indirect effects are associated with the toxic metals and pollutants that bind to microplastics in the environment, potentially causing oxidative stress, inflammation, altered gene expression, and death within organisms (Ma et al., 2019; Chen et al., 2020). Fibers are reported to have a greater impact on freshwater organisms than fragments (Au et al., 2015), which may be due to fibers having a longer duration in the intestinal tract (Ma et al., 2019). Humans are high trophic organisms in the food web, and it has been suggested that microplastics in the environment bio magnify and are present in the food we eat (Xia et al., 2020). Microplastics are also harmful to humans as they can be inhaled and end up in human lungs, generally it is micrometer and nanometer size particles that affect humans (Brander et al., 2020).

1.4 Microplastics in the Environment

Microplastics can travel within and between ecosystems through numerous pathways including via biosolid land amendments, atmospheric transport via wind or coalescence into raindrops, as well as through both the marine and freshwater systems. At wastewater treatment plants, the majority of microplastics leave via biosolids that could then be applied on agricultural fields as a nutrient amendment or end up in the landfill (Anderson et al., 2016; Carr et al., 2016; Crossman et al., 2020). Once in the terrestrial environment, microplastics can persist in the soil or enter aquatic environments through runoff during high rainfall events or erosion via wind (Anderson et al., 2016). Microplastic concentrations can be influenced by a number of factors including, industrialization levels, population density and weather conditions (Brander et al., 2020; Chen et al., 2020).

Microplastics can also become entrained in the atmosphere from multiple sources, e.g., construction sites, landfills, biosolid amended fields, tire wear, or laundry dryer vents. Although, in general the most common microplastic polymer found in the atmosphere are polyester fibers as they easily detach from textiles (Liu et al., 2019; Welsh et al., 2022). The atmospheric settling (dry deposition) of microplastics can be influenced by weather conditions, time of day, as well as wind speed and direction (Chen et al., 2020; Brander et al., 2020). Further, microplastics can adhere to raindrops and then be deposited onto aquatic and terrestrial environments as wet deposition (Chen et al., 2020). Airborne microplastics can potentially travel great distances by being suspended in the air before settling out as dry deposition or being washed out by rainfall onto land (Dris et al., 2015; Brander et al., 2020). Therefore, the atmosphere has been identified as a pathway for microplastics to remote areas (Brander et al., 2020; Roblin et al., 2020); microplastics have been found in the Pyrenees to the Swiss Alps, and in the Arctic (Allen et al., 2019; Bergmann et al., 2019), all remote from microplastics pollution sources.

A number of studies have evaluated the abundance and type of microplastics in wet (or bulk) deposition (Szewc et al., 2021; Klein & Fischer, 2019), but few if any have evaluated dry deposition. To completely understand the dry deposition and fate of atmospheric microplastics, the settling rate needs to be determined. The settling velocity of microplastics has been estimated through models (Wright et al., 2020; Allen et al., 2019), alternatively laboratory-based experiments can be used to directly measure settling velocity following exemplary studies on non-plastic particles (Richards-Thomas & McKenna-Neuman, 2020).

1.5 Catchment Scale Perspective

A lake catchment is the area of land surrounding a lake where water drains from and enters the water system through runoff or streams. The hydrological catchment is a well-defined landscape unit that can provide an integrating scale at which plastic pollution can be investigated and managed (Windsor et al., 2019). At the hydrological catchment scale, microplastics that are deposited on terrestrial ecosystems from the atmosphere, introduced through biosolid amendment, or through poor waste management, will ultimately be transported to surface waters (rivers and lakes) via drains, or runoff. Once microplastics reach the aquatic environment, they can stay within the water column, sink to sediments, get ingested by animals, adhere to plants, get resuspended into the atmosphere, or be washed ashore (Ma et al., 2019; Sarijan et al., 2021). Freshwater ecosystems are considered to be conduits of microplastics (Enamul Kabir et al., 2021) as they receive inputs from the terrestrial and atmospheric environment and contribute microplastics to the marine environment (Horton & Dixon, 2018; Hoellein & Rochman, 2021; Xia et al., 2020). Nonetheless, freshwater ecosystems can act as a transporter, receiver, and sink for microplastic pollution, underpinning the importance of the ‘plastic cycle’ in the fate of microplastics (Hoellein and Rochman, 2021). Once present in a lake, storm activity and turbulence can cause redistribution of microplastics throughout the water column. Although,

sediments have been identified as the ultimate sinks for microplastics in the aquatic environment (Corcoran et al., 2015). Surprisingly, few studies have assessed microplastics in freshwater ecosystems, further many of these studies have only focused on microplastics within rivers; however, lakes can potentially be long-term sinks for microplastics (Horton & Dixon, 2018; Hoellein & Rochman, 2021; Felismino et al., 2021; Anderson et al., 2017).

The transport of microplastics to and from lakes at the hydrological catchment scale integrates terrestrial and atmospheric pathways (Windsor et al., 2019). The terrestrial landscape (urban, rural, agricultural, natural, etc.), the atmosphere, and the different compartments of freshwater ecosystems (rivers, streams, wetlands, ponds, and dams) can affect the flux of microplastics to a lake (Banks, 2022). It is important to estimate these fluxes and pools across catchments as this information can be used to model global budgets (Banks, 2022). Although investigating the hydrological budget of lakes is not uncommon, limited studies have focused on a microplastic particle budget or the 'plastic cycle', i.e., the input and output of microplastics, within a freshwater lake at the catchment scale (Hoellein & Rochman, 2021; Horton & Dixon, 2018). In Ontario, the abundance, sources, and sinks of microplastics in the Great Lakes has been studied extensively (Ballent et al., 2016; Grbić et al., 2020; Eriksen et al., 2013; Corcoran et al., 2015; Dean et al., 2018). In contrast, there are few studies on small inland lakes; one study performed in Lake Simcoe, Ontario, examined microplastics in surface waters and sediments, however atmospheric and stream inputs were not included (Felismino et al., 2021). Further, comparison between studies is hampered by differences in methods, and the size fraction of microplastics that are identified.

1.6 Study Objectives

The overall objective of this thesis was to assess the abundance and distribution of microplastics within Lake Scugog catchment, Ontario. This thesis is comprised of two research (manuscript style) chapters. The first manuscript titled "A microplastic particle budget for Lake Scugog catchment, Ontario" assessed the abundance of microplastics in key watershed components (streams, lake, sediment, atmosphere, etc.) of Lake Scugog during the ice-free period in 2021 (May–October) and quantified atmospheric and riverine pathways of microplastics into and out of the lake. It contributes to the limited knowledge on the fate of microplastics at the lake catchment scale. The inputs that were assessed included stream inflows and precipitation, while the outputs included sediment and lake outflow. Surface water samples were collected from six locations within Lake Scugog, eight inflow streams, and the lake outflow. Sediment samples were collected from the same six locations within the lake. Atmospheric deposition samples were collected with two bulk deposition collectors located on opposite sides of the lake and at nine air biomonitoring sites surrounding the lake. Surface water trawls were also collected at one site, close to the outflow.

The second manuscript titled "Dry deposition of atmospheric microplastics in Port Perry, Ontario" combined measurements of the settling velocity of polyester microfibers with observations of ambient atmospheric microplastics to determine the dry deposition of microplastics. The settling velocity was measured using a drop column and Laser Doppler Anemometer. Ambient atmospheric microplastics were collected daily (every 24 hours) using an active high-volume air sampler in Port Perry, Ontario, during November 2021. Port Perry is located directly beside Lake Scugog, therefore the dry deposition of microplastics found in Port Perry can be compared with atmospheric deposition determined in the microplastic particle budget for Lake Scugog.

The following research chapters were written in manuscript style, therefore some of the text is repeated across chapters, including introductory text, study area description, microplastic identification, extraction methods, spectroscopic analysis, and quality control procedures. This was done in order to facilitate stand alone manuscripts.

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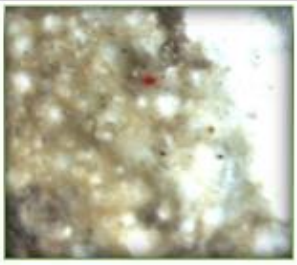
1.8 Appendix

Appendix A- Microplastic Type

Table A1.1- Sample pictures of the most common microplastics including fiber, fragment, and film.

Sample Picture	Microplastic Type
	Fiber
	Fragment
	Film

Chapter 2: A microplastic particle budget for Lake Scugog catchment, Ontario

2.1 Abstract

Globally, microplastic pollution has increased exponentially during the past few decades, which is a concern due to their persistence in the environment. Nonetheless, the knowledge of different sources and pathways of microplastics is limited especially in freshwater ecosystems. In this study, a microplastic particle (input–output) budget was determined for Lake Scugog catchment, Ontario. The inputs assessed included the major stream inflows and atmospheric deposition, while the outputs included sedimentation and the lake outflow. In total 1,181 microplastics (size range: 0.10–5.0 mm) were identified across all sample types. The greatest daily microplastic input flux was precipitation (19.51 mp/m^2/day) and the greatest output flux was lake outflow (3.05 mp/m^2/day). The total input of microplastics to the lake was 2491 x10^6 mp/day, while the output was 1761 x10^6 mp/day, resulting in 29% being retained in the lake. By quantifying the major pathways of microplastics better management techniques for plastic waste can be implemented.

2.2 Introduction

There is growing concern regarding microplastic pollution (particles < 5 mm; 5000 µm) due to its durability, ubiquity within the environment, and potential health impacts (Cole et al., 2011; Anderson et al., 2016). Microplastics have been widely observed in aquatic, terrestrial, and atmospheric environments (Brander et al., 2020; Xia et al., 2020), where it has been suggested that they can harm organisms both directly and indirectly (Ma et al., 2019). Nonetheless, few studies have focused on freshwater ecosystems (Horton & Dixon, 2018; Hoellein & Rochman, 2021).

Knowledge regarding the importance of different sources and pathways of microplastics in freshwater ecosystems is especially limited (Schell et al., 2021). In general, studies that have focused on freshwaters, have typically focused on rivers downstream of anthropogenic point sources (Kay et al., 2018; Sol et al., 2020); pond and lake ecosystems are generally overlooked (Horton & Dixon, 2018; Hoellein & Rochman, 2021). Microplastics can enter lakes via several pathways including surface water run-off, wastewater treatment plants, stream inflows, as well as through atmospheric deposition (Anderson et al., 2016; Carr et al., 2016; Brander et al., 2020; Grbic et al., 2020). Within lakes, microplastics can stay suspended within the water column, sink to the sediments, exit the lake through outflows, be ingested by animals or adhere to plants (Ma et al., 2019; Sarijan et al., 2021). Nonetheless, sediments have been identified as the ultimate sink for microplastics in aquatic environments (Corcoran et al., 2015).

To fully understand the fate of microplastics in freshwaters, the entire catchment must be examined as it integrates all inputs and outputs (the microplastic cycle) at the landscape scale. The microplastic cycle has been defined as a "novel concept and paradigm for understanding plastic pollution and its fluxes across ecosystem reservoirs" (Banks, 2022). Freshwater ecosystems are complex as they have many different compartments including rivers, streams, wetlands, ponds, and lakes (Banks, 2022). Further, freshwater ecosystems are affected by their surrounding landscape and can act as a transporter, receiver, and sink for microplastic pollution. Currently, there are no studies that have determined the microplastic budget (inputs and outputs) for a freshwater lake at the catchment scale.

In Canada only a handful of studies have examined microplastics in freshwater ecosystems (Li et al., 2020; Felismino et al., 2021), even fewer have focused on lakes (Anderson et al., 2017). Within Ontario, studies have primarily focused on microplastics in the Great Lakes

(Yang et al., 2022; Ballent et al., 2016; Grbić et al., 2020; Eriksen et al., 2013; Corcoran et al., 2015; Dean et al., 2018). The few studies that examined smaller lakes have focused only on surface waters and sediments (Felismino et al., 2021; Anderson et al., 2017), but excluded atmospheric and stream inputs. Further, comparison between studies is hampered by differences in methods, and especially the size fraction of microplastics that are identified. To gain a better understanding of the fate of microplastics they need to be quantified at smaller size fractions.

The overall objectives of this study were to determine the abundance of microplastics in Lake Scugog catchment, and to develop a microplastic particle (input–output) budget for the lake catchment. In addition, preliminary estimates of smaller particles typically not included in visual analysis was performed using Nile red dye and blue-light fluorescence to detect microplastics >20 µm (>0.02 mm). The inputs that were assessed included stream inflows and atmospheric deposition, while the outputs included sedimentation and the lake outflow. Surface water samples were collected from six locations within the lake, eight inflow streams, and the lake outflow. Sediment samples were collected from the same six locations within the lake. Atmospheric deposition samples were collected with two bulk deposition collectors placed on opposite sides of the lake, and nine atmospheric bio-monitors surrounding the lake were used to assess variability between atmospheric inputs across the entire lake. Surface water trawls were also collected in triplicate at one site closest to the outflow; all sampling was performed between May–October 2021.

2.3 Methods

2.3.1 Study Area

Lake Scugog catchment is located within south-central Ontario, Canada. The catchment is 529 km^2 with the lake being 68 km^2 (Kawartha Conservation, 2010, 2020); the lake is quite shallow with an average depth of 1.4 m, and 7.6 m at the deepest point (Kawartha Conservation,

2010). The lake's catchment is mainly positioned within the Township of Scugog (Durham Region) and the City of Kawartha Lakes, with Port Perry (population, 9,553 as of 2021) being the largest town (Statistics Canada, 2017). Lake Scugog is a macrophyte dominated ecosystem (Harrow-Lyle & Kirkwood, 2020) and a headwater lake that receives most of its water from headwater streams on the Oak Ridges Moraine. Since it is a headwater lake, it drains northwards to Sturgeon Lake, which leads through the Trent Severn Waterway and onwards to Lake Ontario. Water from the Scugog River is the primary source of drinking water for residents in Lindsay (Kawartha Conservation, 2010). The Lake Scugog watershed is not densely populated; most residents live in small hamlets in the rural portion of the watershed. The catchment is dominated by agricultural/rural landcover (52%), with the rest made up of swamp (14%), open water (12%), treed area (11%) and other (11%) (MNRF, 2020). Long-term annual precipitation to the catchment is 882 mm, mean annual temperature is 6.8°C, and the mean elevation is 275.5 m (MNRF, 2020).

2.3.2 Study Sites

This study focused on the lake catchment and quantified microplastic abundance in the dominant inputs (lake inflows and precipitation) and outputs (lake outflow and sediment) of the lake. In addition, lake water, surface trawls, and atmospheric active bio-monitors (moss bags) were analyzed (Figure 2.1). Water samples were collected monthly from nine stream inflows[1] and the lake outflow (S1–S10), these sites corresponded with Kawartha Conservation's Lake Scugog Environmental Management Plan (LSEMP), which were routinely sampled during the ice-free period (May–October 2021). Five of the inflows and outflow sampled were classified as rural, three as agricultural, and two as urban. Further, six locations within the lake (L1–L6) were

[1] Note: S4 was only sampled once in May 2021.

sampled routinely (May–October 2021), again consistent with Kawartha Conservation's LSEMP monitoring sites. In October 2021, three parallel surface trawls (T1) were performed at the lake site (L1) closest to the outflow of Lake Scugog. In addition, lake sediment (Sd1–Sd6) was collected in October 2021, below the six lake water sites (L1–L6). Precipitation samples were collected monthly at two sites in the Lake Scugog catchment using bulk deposition collectors, which captured wet and a portion of dry deposition; they were placed in downtown Port Perry (R1; collocated with Kawartha Conservation's climate station and Water Survey of Canada [WSC] station) and at Blackstock Creek (R2; collocated with a WSC station). Additionally, nine atmospheric bio-monitors (M1–M9) were placed around Lake Scugog.

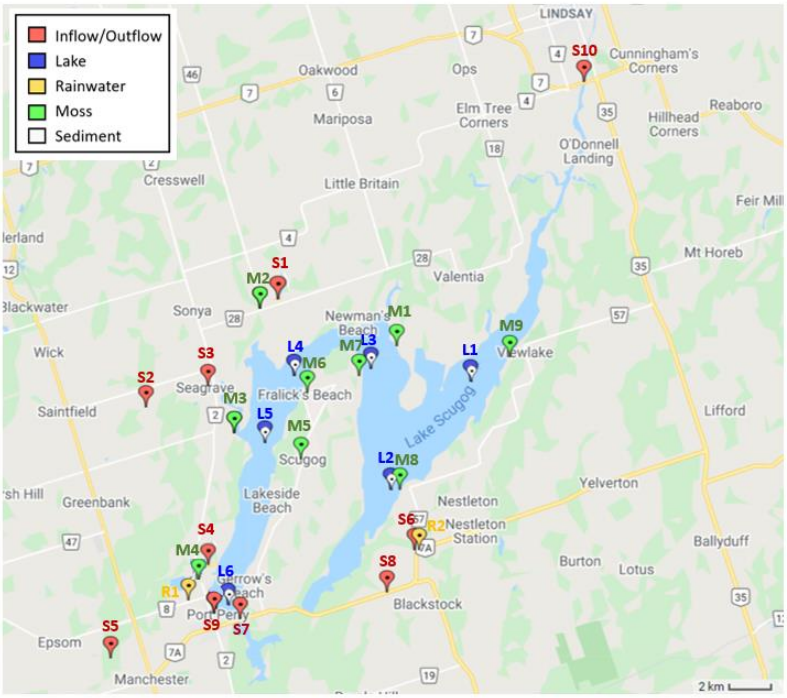


Figure 2.1- Map of Lake Scugog showing the different sampling locations for stream inflows, the lake outflow, rainwater, bio-monitors, and sediment all sampled during May–October 2021 (for site coordinates see Appendix Table A2.1).

2.3.3 Field Sampling

Inflow streams, the lake outflow, and lake water samples were collected monthly by Kawartha Conservation. One-liter samples were taken at each site into a sterilized wide-mouth glass (Mason) jar, which was triple rinsed with the sample water at each site, and filled at a depth of 10 cm. One rotating field blank and duplicate sample were collected monthly for both lake inflows/outflow (n=6) and lake sites (n=6). Duplicates were performed to assess the variability in the sampling procedures and were collected in the same manner as the environmental samples. Blanks were performed to correct for contamination, this was done by pouring one-liter of filtered (to remove any possible microplastics) B-pure water into a one-liter jar during field sampling. All containers used for sample collection were sterilized prior to use.

Three parallel 1 km (approximately 35 m^3 of water) trawls (T1) perpendicular to the lake outflow were performed using a 0.05 mm (50 µm) plankton net, which was trawled beside a boat at approximately 3.7 km/hr in triplicate (back and forth); two trawls were performed north–south and one was south–north. The opening of the net was a circle (diameter of 30 cm), half of which was placed in the water. Once the trawl was complete the cod end was brought onto the boat, and emptied and rinsed into a glass jar. A field blank was performed after the sampling was complete; the net and cod end were rinsed into a stainless-steel bowl with one-liter of filtered B-pure water, which was then transferred to a glass jar.

Sediment was collected at six sites (Sd1–Sd6; Figure 2.1) using an Eckman Grab sampler that collected the top 5 cm. At each site, two samples were taken, one from the starboard side and one from the port side of the boat. The samples were mixed in a stainless-steel bowl, and a 250 mL subsample was kept in a glass jar for analysis. A field blank was performed on the

Eckman Grab sampler by rinsing one-liter of filtered B-pure water over it into a bowl, which was transferred to a glass jar.

Two atmospheric bulk deposition collectors were placed around the lake; in Port Perry, a NILU (PN. 9735) microplastic bulk deposition collector (diameter of 20 cm) was placed on a pole, approximately 2 m above the ground. The Blackstock Creek bulk deposition collector was an aluminum bucket (diameter of 23 cm) installed on the roof of a stream gauge station (WSC ID: 02HG003), approximately 3 m above the ground, both collectors had a basket (stainless steel) on top of the bucket to deter birds. Rainwater was collected at the end of each month (approximately every 31 days) and stored in one-liter glass jars; the collector was then triple-rinsed with filtered B-pure water, which was also stored for microplastic analysis.

Atmospheric bio-monitors were used to assess variability in atmospheric deposition around the lake. These bio-monitors (small metal mesh bags with 1 g of the moss *Pleurozium schreberi* enclosed, "moss bags") were placed on hydro poles (approximately 3 m from the ground) in duplicate at nine locations (close to the six lake sites; Figure 2.1) surrounding the lake perimeter (~500–2000 m from the lake) and were deployed for two-month intervals, June 1st–August 1st and August 1st–October 1st. *Pleurozium schreberi* was picked in May 2021 from Warsaw Caves Conservation Area (44.461904, –78.131332) following ICP vegetation guidelines (ICP Vegetation, 2015) and stored in paper bags; this location was used as it is remote from direct pollution sources. The moss was cleaned of debris, oven-dried at 45–55°C for 24 hours, weighed into 1 g samples and placed into mesh bags, which were stored in aluminum foil until deployment. The moss bags (5 cm^2) were created from 1 mm aluminium mesh and sewn together with 20-gauge wire; the bags were secured with wire onto 'L' brackets attached to the hydro

poles. Field blanks (n=5) were performed by attaching a moss bag to the 'L' bracket on the hydro pole and immediately removing. The moss bags were corrected using unexposed moss.

2.3.4 Microplastic Extraction

In the laboratory, samples were stored at room temperature for up to one month prior to analysis. The lake inflows/outflow and lake water samples were directly vacuum filtered using a Buchner funnel onto 4.2 cm diameter glass fiber filter papers (Fisherbrand™ G6 [09-804-42A] pore size 1.6 µm) using approximately 3 per sample[2] and stored in petri dishes.

The trawl samples were passed through a 200 µm (0.2 mm) sieve, the <200 µm portion was vacuum filtered onto glass fiber filter papers (approximately 14 filter papers per sample). The >200 µm sample was digested using a wet peroxide oxidation, which followed previously published work (Herrera et al., 2018; Masura et al., 2015; Roblin & Aherne, 2020a). The >200 µm contents from each trawl sample were emptied into a 500 mL or 1000 mL glass beaker, 40 mL of 30% hydrogen peroxide (H_2O_2) was added, and it was incubated at 45–55°C for 18–24 hours. Once removed from the heat, 40 mL 0.05M Fe (II) was added and it was left at room temperature for five minutes. The heat of the reaction was monitored and if it exceeded 55°C filtered B-pure water was added. As the reaction slowed down, additional H_2O_2 aliquots were added in 20 mL increments, for a total of at least 100 mL of H_2O_2. Once the digestion was complete the digestate was vacuum filtered onto glass fiber filter papers (approximately 2 filter papers per sample).

The sediment samples were oven dried at 45–55°C for approximately seven days. Once dry, a 5 g sub-sample was analyzed in triplicate (3 × 5 g) from each site using the wet peroxide

[2] When there was a large amount of residual material in the samples, multiple filter papers were used to facilitate easier identification.

oxidation method. The digestate was then passed through a 90 µm (0.09 mm) sieve, the <90 µm material was vacuum filtered (approximately 2 filter papers per sample), the material left on the sieve (most of the sediment was >90 µm) was sequentially density separated three times (once with water and twice with sodium iodide [NaI] solutions). Water and NaI were used as they both have a density higher than the average plastic, resulting in plastic floating on the surface (Nuelle et al., 2014). The density separator was created from a pipe with a ball valve in the middle following Coppock et al. (2017). The sample and solution (water or NaI; approximately 500 mL) was put in the density separator, placed on a stir plate for 5 min and removed, once the sample had settled (2–8 hrs dependent on solution type) the ball valve was rotated, and the bottom and top portions were separated. The initial density separation used water to clean the sample of H_2O_2 and Fe (II); approximately 500 mL of water was added and left to settle for 2–3 hours. The top portion of the density separator was vacuum filtered onto glass fiber filter papers (approximately 2 filter papers per sample) and the bottom portion was sieved (90 µm) to remove the water, which was discarded. The sieved portion was again density separated and left to sit for 6–8 hours using NaI (500 mL, density of 1.8 g/cm^3; Nuelle et al., 2014). The top portion once again was vacuum filtered, and the bottom portion was put through a final NaI density separation; after this the remaining sample was discarded.

The rainwater samples contained organic debris, therefore a 30% H_2O_2 solution was added to each jar (10% by volume) and left to incubate for one week at room temperature to breakdown the organic material. The samples were then vacuum filtered onto glass fiber filter papers (approximately 7 filter papers per sample).

When removed from the field, atmospheric bio-monitors were wrapped in aluminum foil, oven-dried at 45–55°C for 24 hrs and weighed. The contents of each bag were emptied into

separate 500 mL glass beakers and incubated at 45–55°C for 12–24 hrs in 40 mL of 30% H_2O_2 and digested individually using the wet peroxide oxidation method. Once the digestion was complete the sample was vacuum filtered (approximately 4 filter papers per sample).

2.3.5 Identification of Microplastics

Brightfield microscopy is widely used to detect microplastics, which works well when identifying millimeter size particles; however, it becomes challenging when identifying submillimeter size particles, i.e., reportedly <50 µm (0.05 mm) but in practice <100 µm (0.1 mm; Labbe et al., 2020; Erni Cassola et al., 2017). In addition, it is very difficult to identify clear particles with brightfield microscopy. Fluorescent dye can be used to help identify submillimeter and clear particles (Labbe et al., 2020; Erni Cassola et al., 2017). In the current study, brightfield microscopy was performed on all samples, in addition a Nile red dye and blue-light (wavelength 460 nm) procedure was performed on a subset of samples (10%).

Using a stereomicroscope with a digital camera (Amscope with Infinity 2 Teledyne Lumenera camera), all filter papers underwent visual analysis adhering to a widely used identification protocol (Roblin et al., 2020b; Windsor et al., 2019; Norén, 2007) based on five criteria: (i) the particle is unnaturally coloured; (ii) it is homogenous in material and texture with no cell structure or offshoots present; (iii) the particle is not brittle and does not break when poked, tugged or compressed with tweezers; (iv) it is shiny or glossy in appearance; and (v) there is limited fraying with no similarity to natural particles. Suspected plastic particles were photographed and then confirmed using a hot needle test following Roblin et al. (2020b). A hot needle was pressed against the particle, if it melted it was classified as plastic, and the image of the microplastic was then measured using the image processing software, ImageJ. The particles

were measured by converting the number of pixels to a known length in millimeters. The length, width, colour, and type (i.e., fragment, fiber, film, etc.) of each microplastic was recorded.

A subset of samples (minimum of 10% of all samples and blanks, i.e., 10% of inflow/outflow samples, 10% of lake water samples, etc.) were chosen randomly and further analyzed for clear microplastic particles and particles <100 µm (0.1 mm). This subset was dyed using Nile red dye, which takes advantage of the hydrophobic properties of plastics, allowing them to fluoresce under different wavelengths (460–630 nm). To each selected filter paper, 1 mL of Nile red dye (concentration of 0.1 mg/mL) was added to completely cover the sample (Erni Cassola et al., 2017). The filters were incubated in the dark at 50°C for 30 min to improve the adsorption of Nile red (Erni Cassola et al., 2017). Following incubation, they were visually analyzed by microscopy under a blue light with a wavelength of 460 nm. Any particle that fluoresced was then hot needled, if it melted it was counted as a microplastic. These particles were not photographed as they were too small to measure, rather they were categorized as <100 µm.

2.3.6 Polymer Identification

Micro-Raman spectroscopy (WITec, operated by WITec Control) was used to determine the type of plastic polymer following Roblin et al. (2020b). Fragments were analyzed using a 532 nm laser and fibers were analyzed using a 785 nm laser at 100× objective and adjustable power (approximate range, 0–85 mW). Spectra were recorded in the wavenumber range of 0–1,800/cm and analyzed through an open-access library (Open Specy; Cowger et al., 2021) to confirm polymer identity.

2.3.7 Quality Control and Assurance

Field and laboratory blanks were analyzed in the same manner as environmental samples. Open air blanks were performed in the laboratory by placing a clean glass fiber filter paper

(Fisherbrand^{TM} G6 1.6 µm) in a petri dish on the counter for one hour during both the extraction and counting procedures. This was performed twice a month (n =10) and was used to determine potential contamination during laboratory analysis. The duration that samples were exposed through all laboratory procedures (extraction, counting, etc.) was also recorded. Field blanks were used to blank correct the concentration of microplastics across all samples (For blank corrections see Appendix Table A2.2). Duplicates (inflows/outflow, lake water, bio-monitors) were performed to assess sample and environmental accuracy, laboratory triplicates (sediment) assessed the variability within samples and environmental triplicates (trawl) were performed to assess sample and environmental accuracy (see Appendix Table A2.3). All B-pure water and chemical solutions (H_2O_2, Fe (II), Na(I)) were vacuum filtered prior to use to remove any possible microplastics.

In preference, glass and metal material were used throughout field and laboratory work, however some plastic materials needed to be used (petri dishes, B-pure water spray bottle, density separators, etc.). Equipment and sample containers were triple rinsed with filtered B-pure water and glass jars were sterilized at ~100°C for 10–20 minutes. During analysis samples were covered with aluminum foil as much as possible. The Buchner funnel was triple rinsed between samples, the rinse water was poured onto the same filter paper as the sample. The filter was then removed, and the Buchner funnel was triple rinsed again and wiped with a Kimwipe between samples.

2.3.8 Data Analysis and Data Sources

The concentration of microplastics (mp/L) was calculated for all inflows, the lake outflow, lake water, and precipitation by dividing the number of particles (count) observed by the sample volume, i.e., 1 L for all inflow/outflow/lake samples and the volume of rainfall for

each individual deployment period. The concentration of microplastics (mp/m^3; mp/m^2) for all trawl samples was calculated by dividing the number of particles, observed by the sample volume (half the area of net and distance travelled), or distance travelled. The concentration of microplastics (mp/g) for sediment and bio-monitors was calculated by dividing the number of particles observed by the sample weight. The sediment samples were analyzed in 5 g triplicates, which were averaged to determine the concentration at each site. Similarly, the count per moss bag was divided by the final moss weight. The variation in triplicates was estimated using the coefficient of variation, while the variation in duplicates was estimated using the relative percentage difference. Lastly for each sample type the average of their respective field blanks was subtracted from the concentration; no microplastics were found in the laboratory open air blanks.

The daily flux (mp/m^2/day) for inflows/outflow was calculated using the microplastic concentration (mp/L), the liters flowing through each inflow/outflow per day, (L/day; obtained from Lake Scugog Environmental Management Plan; Kawartha Conservation, 2010), and their drainage area (m^2; obtained from Lake Scugog Environmental Management Plan [Kawartha Conservation, 2010] and the Ontario Flow Assessment Tool; MNRF, 2020).

River Flux (mp/m^2/day) = Concentration (mp/L) x Discharge (L/day) ÷ Drainage Area (m^2)

The daily deposition (mp/m^2/day) for precipitation was calculated using the microplastic concentration (mp/L), the precipitation volume (mm; obtained from Kawartha Conservation) measured at each station, and the number of days sampled.

Precipitation Flux (mp/m^2/day) = Concentration (mp/L) x Rainfall (mm) ÷ Number of Days

The microplastic sedimentation flux (mp/m^2/day) was calculated using the microplastic concentration (mp/g) in sediment and the sedimentation rate for the top 5 cm in Lake Scugog (g/cm^2/day; obtained from Wiklund & Hall, 2017).

Sediment Flux (mp/m^2/day) = Concentration (mp/g) x Sedimentation Rate (g/m^2/day)

The particle budget was then determined using the daily number of microplastics entering and leaving Lake Scugog, i.e., the flux multiplied by the contributing area. The total microplastic inflow to Lake Scugog was determined by adding all lake inflows and precipitation directly to the lake surface, while the outflow was determined by adding the lake outflow and sedimentation rate of microplastics. The retention of microplastics in Lake Scugog was estimated by calculating the difference between all lake inputs and outputs. The residence time of microplastics was determined by dividing the total number of microplastics in the lake by the annual retention (daily retention × 365 days).

Residence Time (year) = Total Microplastics in Lake (mp) ÷ Annual Retention (mp/year)

The total microplastics in the lake was calculated using the lake water samples and trawl samples. Microplastics on the surface (depth of 15 cm) of the lake was determined by calculating the average concentration of microplastics in each trawl and upscaling this using the area of the lake to the total number of microplastics on the surface of the entire lake. Microplastics in the lake water was determined in a similar fashion using the average concentration of microplastics at each lake site and the lake volume (less the surface volume). The total number of microplastics on the surface and within the lake were added to determine the overall number of microplastics in Lake Scugog. The percent of microplastics retained in the terrestrial catchment was determined by difference of the total microplastics entering the catchment (precipitation flux

multiplied by the area of the terrestrial catchment) and the total leaving the terrestrial catchment, which was calculated by adding the number of microplastics in all the inflows entering the lake. The particle budget was based on all microplastic types combined rather than individual shapes (i.e., fibers and fragments) due to sampling limitations (for further details see Appendix Table 2.4) Statistical analysis was performed on all sample types to determine if there was a statistical difference between sites and months (where applicable) using a one-way Anova as data were normally distributed (see Appendix Table A2.5). All statistical analysis was carried out in PAST 4.10 (PAST, 2020).

Total microplastics (particles <100 µm plus particles >100 µm) for each sample type (inflows, lake water, sediment, etc.) were estimated using a sample specific scale factor, which upscaled the brightfield microscopy counts to total microplastic counts (see Appendix Table A2.6). The scale factors were determined using the average counts from the samples that underwent both procedures (brightfield and fluorescence microscopy).

2.4 Results and Discussion

2.4.1 Concentration of Microplastics

In total 1,181 microplastics (size range: 100–5000 µm; 0.1–5 mm) were identified across all sample types. In addition, there were 17 macroplastics identified that exceeded a length of 5 mm (5000 µm); the majority of these were in precipitation and sediment samples. Overall, 123 samples were taken and only four of these had zero microplastics; the zero detect samples were either in stream inflow, lake outflow or lake samples and may have been influenced by the low sample volume (1 L) as suggested by the high coefficient of variation for lake and stream samples (72.5–115.3%; Table 2.1; see Appendix B2). Notably, there was no statistical difference in microplastic counts or concentration between sites and seasons (where applicable) for all sample types other than sediment.

The average concentration of microplastics in the stream inflow samples (n=46) was 2.4 mp/L (median of 2.4 mp/L; range of 0–8.4 mp/L), the lake outflow samples (n=4) was 3.1 mp/L (median of 2.4 mp/L; range 0–7.4 mp/L), and in lake water samples (n=36) was 2.9 mp/L (median of 2.4 mp/L; range of 0–7.4 mp/L; Table 2.1; see Appendix Table C2.1; C2.2). Comparison is difficult between studies due to differences in sampling periods, collection methods, and identification techniques. Welsh et al. (2022), who examined three lakes in the Muskoka-Haliburton region, found slightly lower concentrations in lake water, stream inflows, and lake outflows, 0.95–1.51 mp/L, 0.79–1.14 mp/L, and 0.91–2.0 mp/L, respectively across the three lakes, likely due to the lower population density in their study area compared with this study. The lakes examined in the Muskoka-Haliburton region were all remote (3–50 km) from towns, whereas Lake Scugog is directly beside Port Perry. Further, Felismino et al. (2021) found much lower concentrations in lake water in Lake Simcoe (0.04 mp/L); however, this is likely due to the differences in field sampling as Felismino et al. (2021) used a 125 µm mesh screen to collect microplastics.

In the current study, the average surface trawl (n=3) concentration was 3.2 mp/m^3 or 377,778 mp/km^2 (range of 1.9–4.3 mp/m^3; Table 2.1; see Appendix Table C2.3). In contrast, Felismino et al. (2021) found an average of 32,700 mp/km^2 in Lake Simcoe and Eriksen et al. (2013) found an average of 43,157 mp/km^2 in the Laurentian Great Lakes, potentially owing to the larger mesh size in the trawl nets used in these studies (100 µm [Felismino et al., 2021], 333 µm [Eriksen et al., 2013]). Another reason for the different concentrations may be due to the mesh screens used to process samples (125 µm [Felismino et al., 2021], 355 µm [Eriksen et al., 2013]).

The average concentration of microplastic in sediment (n=6) was 2.2 mp/g (range of 0.8–3.6 mp/g; Table 2.1; see Appendix Table C2.4). In contrast, Felismino et al. (2021) observed 0.008–1.07 mp/g in Lake Simcoe, Ballent et al. (2016) found 0.98 mp/g in nearshore sediments in Lake Ontario, and Welsh et al. (2022) observed 3.6 mp/g in three lakes in the Muskoka-Haliburton region. The difference in sediment concentrations is likely owing to the different methods used. Felismino et al. (2021) used different extraction methodology; anything <45 µm was screened out and the sediment samples only underwent one density separation. In the current study it was found that more microplastics were in the second and third density separations (NaI solution) than the first (H_2O solution).

The concentration of microplastics in precipitation (n=10) had an average of 8.1 mp/L (range of 3.4–16.4 mp/L; Table 2.1; see Appendix Table C2.5) across the two sampling sites. In contrast, Welsh et al. (2022) found a lower concentration with a range of 1.47–2.64 mp/L in precipitation samples in the Muskoka-Haliburton region, and Roblin et al. (2020b) found a concentration of 3.5 microfibers/L in Ireland. The higher concentration in the Lake Scugog catchment is most likely due to the population density, both Roblin et al. (2020b) and Welsh et al. (2022) performed their studies in more remote areas away from towns and cities whereas Lake Scugog is next to the town of Port Perry.

Table 2.1- Microplastic counts and concentrations by sample type for particles >100 µm. The particle count is the total across all sites and months. The average concentration, range, and sample variation, i.e., coefficient of variation (CV%) is calculated across all months (where applicable) and sites.

Sample Type	Number of Samples	Number of Sites	Sampling Frequency	Particle Count	Average Concentration	Median Concentration	Range	CV%
Inflow (mp/L)	46	9	Monthly	107	2.4	2.4	0-8.4	87.2
Outflow (mp/L)	4	1	Monthly	12	3.1	2.4	0-7.4	115.3
Lake (mp/L)	36	6	Monthly	108	2.9	2.4	0-7.4	72.5
Trawl (mp/m^3)	3	3	Once	340	3.2	3.4	1.9-4.3	36.6
Sediment (mp/g)	6	6	Once	197	2.2	1.9	0.8-3.6	49.3
Precipitation (mp/L)	10	2	Monthly	328	8.1	8.0	3.4-16.4	48.5
Bio-monitor (mp/g)	18	9	Bi-Monthly	89	3.0	2.8	0.4-6.5	57.8

In general, there was large spatial variability for the inflow and lake sites compared with sediment with an average of 78.3%, 69.7%, and 49.7% respectively (Table 2.2). The inflow and lake sites may have had high variability due to the low sample volume; however, it was clear that the lake site closest to the outflow (L1) had high concentrations compared with the other sites (Table 2.3; see Figure 2.1), which was anticipated due to the flow of water in the lake. The sediment may be more representative of microplastic concentrations across the lake than the inflow and lake sites as there seemed to be a pattern across all samples; there was a statistical difference between the sediment sites (One-way Anova $p < 0.05$) as the east basin near the outflow (Sd1–Sd3) had higher concentrations than the west basin (Table 2.3; see Figure 2.1).

The average concentration of microplastics in the bio-monitors placed around the lake was 3.0 mp/g (range 0.4–6.5 mp/g; Table 2.1; see Appendix Table C2.6). The bio-monitors represent the spatial variability for atmospheric inputs across the entire catchment, and the overall coefficient of variation between samplers was 33.2% (Table 2.2). This low coefficient of variation indicates that the atmospheric inputs across the entire catchment were similar, which suggests that two precipitation stations were representative of the entire catchment. The high variation between inflow sites did not show any relationship with population density, as S9 is directly in Port Perry, yet had the lowest concentration (1.3 mp/L; Table 2.3). Klein et al. (2015) similarly reported no correlation between population and concentration in a river in Germany and rather suggested that hydrodynamic effects may superimpose the correlation with population density. There was a large difference in the average concentrations between precipitation sites, as one collector was in downtown Port Perry (R1; 10.1 mp/L), while the other was in a rural setting near Blackstock Creek (R2; 6.1 mp/L; Table 2.3; Figure 2.1). Similarly, Welsh et al. (2022) found that the concentration of microplastics in bulk deposition increased with anthropogenic activity. The average temporal variability across sampling sites for the inflow and lake were high, at 90.1% and 67.8% respectively (Table 2.3). May and June generally had high concentrations compared to the other months while July and August were generally low (Figure 2.2), suggesting seasonal variability.

Table 2.2- Spatial variability (percent variation in the average concentration between sampling sites) represented by coefficient of variation (CV%) for inflow and lake samples across the six-month sampling period, sediment sampled in October, and the bio-monitors sampled in June–July and August–September.

	Inflow	Lake	Sediment	Bio-monitors
Average (%)	78.3	69.7	49.7	33.2

Table 2.3- Average concentration and particle count (blank-corrected) of microplastics at all sites with the temporal variability (percent variation in the monthly average concentration between sampling months) represented by coefficient of variation (CV%). For stream inflows and lake outflow (S), lake water (L), and precipitation (R), the average concentration (mp/L) across all the months is presented, for sediment (Sd) the average concentration (mp/g) across the triplicates is presented, and for bio-monitors (M) the average concentration (mp/g) across the two deployment periods is presented and relative percent difference (RPD%) is given for variability. Note: S4 was only sampled for one month.

Sample	Site ID	Count	Concentration (mp/L or mp/g)	Temporal Variability (%)
Inflows	S1	18.4	3.1	39.5
	S2	10.9	1.8	102.1
	S3	16.8	3.4	57.3
	S4	2.4	2.4	-
	S5	11.6	1.9	80.8
	S6	7.0	1.4	87.5
	S7	17.4	2.9	89.3
	S8	15.0	3.0	109.5
	S9	8.0	1.3	129.6
Outflow	S10	12.2	3.1	115.3
Inflows and outflow	Average	12.0	2.4	90.1
Lake	L1	21.4	3.6	65.0
	L2	15.9	2.7	87.5
	L3	9.9	1.7	37.1
	L4	17.4	2.9	52.3
	L5	18.0	3.0	83.8
	L6	20.4	3.4	81.1
Lake	Average	17.2	2.9	67.8
Precipitation	R1	164.7	10.1	40.8
	R2	149.8	6.1	45.6
Precipitation	Average	157.3	8.1	43.2
Sediment	Sd1	50.0	3.3	-
	Sd2	54.0	3.6	-
	Sd3	32.0	2.1	-
	Sd4	27.0	1.8	-
	Sd5	12.0	0.8	-
	Sd6	22.0	1.5	-
Sediment	Average	32.8	2.2	-
Bio-monitor	M1	4.2	1.3	47.6
	M2	12.2	3.5	147.5
	M3	12.2	3.5	16.4
	M4	9.2	2.7	43.5
	M5	6.2	1.8	161.3
	M6	10.2	3.0	19.6
	M7	7.2	2.1	55.6
	M8	14.2	4.1	98.6
	M9	13.2	3.8	60.6
Bio-monitor	Average	9.9	2.9	72.3

In general, there is a relationship between the amount of precipitation and the concentration of microplastics (Figure 2.2; Appendix Table C2.5), which is widely observed for precipitation chemistry, i.e., during dry periods with low rainfall volume the concentration of pollutants is higher (Oji & Adamu, 2020; Weli, 2014; Taylor & Nakai, 2012). Similarly, surface water samples were influenced by flow during the drier period (June–August), i.e., the concentrations of microplastics in inflows/outflow and lake water were lower (Figure 2.3).

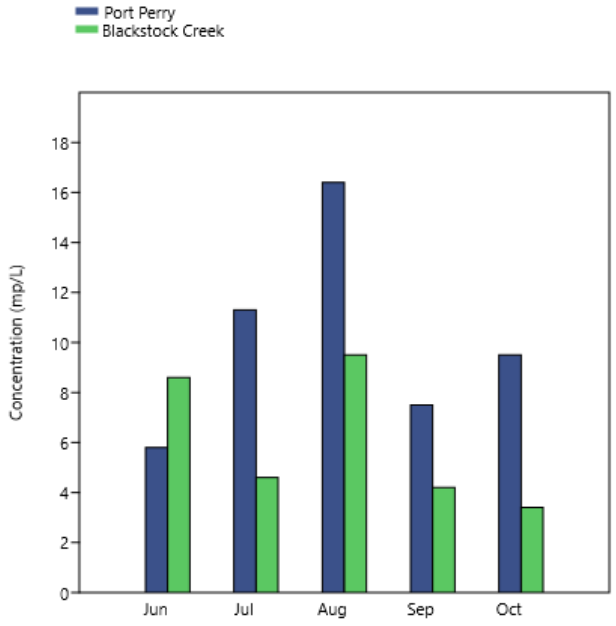


Figure 2.2- Monthly microplastic concentrations in precipitation during June–October 2021 across the Port Perry and Blackstock Creek study sites.

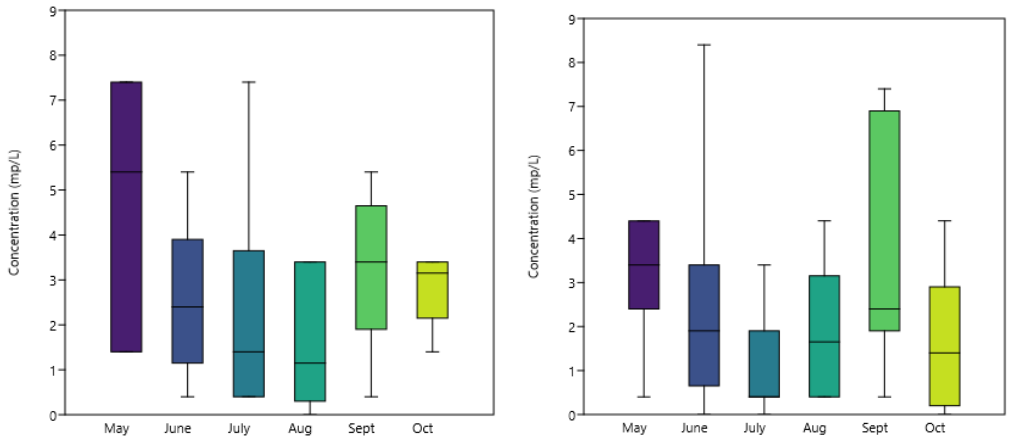


Figure 2.3- Microplastic concentrations during May–October 2021 in (left) lake water by month across six sites, and (right) stream inflows (n=8) and the lake outflow by month. The box represents the 25th and 75th percentile, the horizantal line represents the median and the whiskers represent 1.5 times the interquartile range.

2.4.2 Microplastic Shape

Fibers were the dominant shape (77.4%) in all environmental samples (Table 2.4), which is similar to many other studies (Lenaker et al., 2019; Baldwin et al., 2016; Anderson et al., 2017; Welsh et al., 2022; Felismino et al., 2021). Welsh et al. (2022) found that 71% of microplastics in Muskoka-Haliburton lakes (n=3) were fibers, while Felismino et al (2021) found 82.4% of microplastics in Lake Simcoe were fibers. Sediment was the only sample type that had similar fiber (38.9%), fragment (28.8%) and film (31.3%) counts (Table 2.4), which may be due to fibers leaving the lake more easily through the outflow as they are the dominant shape found in the water column. Dusaucy et al. (2021) also found similar proportions of fibers and fragments in sediment samples across different lakes worldwide. Welsh et al. (2022) found the proportion of fragments to be two times greater than fibers in sediment. In contrast, Felismino et al. (2021)

found that only 5% of microplastics in sediments were fragments, this low percentage may be due to the sampling screening cut-off range of 125 µm.

The largest percentage of microfibers (97.3%) was observed in precipitation (Table 2.4). Welsh et al. (2022), Wright et al. (2020) and Abbasi & Turner (2021) also found that fibers were dominant in atmospheric deposition. Fibers are commonly found in the atmosphere as they easily detach from textiles during wear, washing, and drying (De Falco et al., 2018; Pirc et al., 2016; Napper & Thompson, 2016). Although fibers were dominant in atmospheric bio-monitor samples, these samples also had the second highest percentage of fragments, 14.1% (Table 2.4). The high percentage of fragments in the bio-monitors could be from automotive sources (such as tire wear) as the deployment sites were next to roads. Fragments tend to deposit close to their source, whereas fibers tend to remain suspended in the atmosphere and travel further from their source (Loppi et al., 2021).

Table 2.4- Fibers, fragments, films, and other particle types as a percentage and total count within each sample type.

	Percentage				Count			
	Fiber	Fragment	Film	Other	Fiber	Fragment	Film	Other
Inflows/Outflow	89.1	5.9	5.0	0.0	90	6	5	0
Lake	94.3	3.8	1.9	0.0	99	4	2	0
Trawl	87.6	10.8	1.0	0.5	170	21	2	1
Sediment	38.9	28.8	31.3	1.0	77	57	62	2
Precipitation	97.3	0.0	1.4	1.4	144	0	2	2
Bio-monitor	74.7	14.1	8.1	3.0	74	14	8	3
Percent/Total	77.4	12.1	9.6	0.9	654	102	81	8

2.4.3 Microplastic Composition

Thermoplastics were dominant across all sample media, the most common (52.7%) being polyester or polyethylene terephthalate (PET). The majority of fragments (28.5%) were

polypropylene (PP), while PET was the most common fiber polymer (49.3%). Within water (inflow, outflow, lake, trawl) and precipitation samples PET was 59.0% and 50.0% respectively (Figure 2.4). Whereas nitrile rubber (NR) was commonly found in the bio-monitors (37.5%; Figure 2.4) which could be from automotive sources, such as tire/brake wear (Sommer et al., 2018). Further, the polymers in the sediment samples were quite evenly distributed with the dominant types being PET, PP, polystyrene (PS) and polyvinyl chloride (PVC; Figure 2.4). These polymers have been widely reported by a number of different studies (Welsh et al., 2022; Felismino et al., 2021; Wright et al., 2020; Abbasi & Turner, 2021; Allen et al., 2019). Felismino et al. (2021) found that PET was the most common plastic type in water samples and PS was common in sediment; further, Welsh et al. (2022) found PET was the most common (31%) polymer in precipitation.

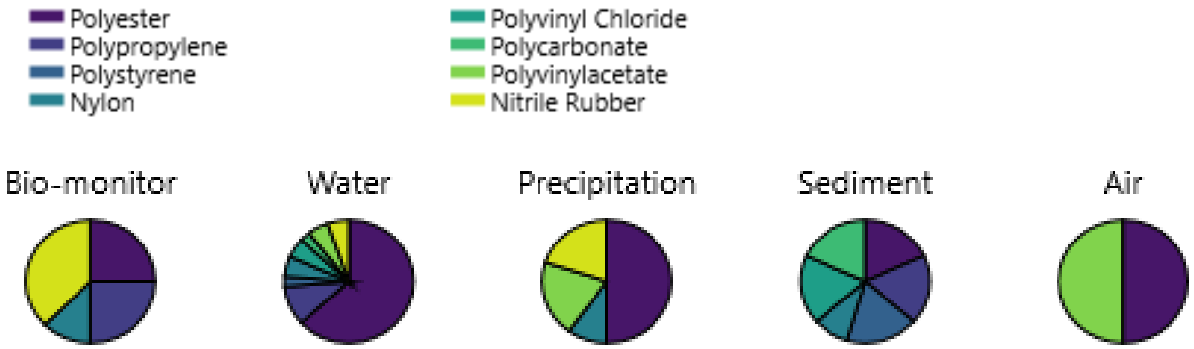


Figure 2.4- Pie charts showing the proportion of different polymers in each sample media type across all shapes for Lake Scugog sampled during May–October 2021 (see Appendix Table C2.7).

2.4.4 Microplastic Size

The majority of microplastics (85%) across all samples were less than 2 mm (2000 µm) in length (Figure 2.5; Figure 2.6). For inflow and outflow samples the average length was 1.15 mm (see Appendix Table C2.8). Welsh et al. (2022) found the average length to be lower in inflow (0.55 mm; 550 µm) and outflow (0.37 mm; 370 µm) samples than the current study. The length of microplastics in the lake samples were similar to the inflow/outflow sample lengths at 1.06 mm (1060 µm; see Appendix Table C2.8); once again longer than Welsh et al. (2022), where the microplastics in lake samples (Muskoka-Haliburton) had an average length of 0.53 mm (530 µm). Microplastics may have been smaller in the Muskoka-Haliburton region because the area is distant from pollution sources resulting in microplastics travelling further; generally smaller microplastics travel greater distances through the atmosphere (Loppi et al., 2021). Whereas Lake Scugog is closer to possible pollution sources and larger microplastics do not travel as far. The length of microplastics in trawl samples was similar to the inflow/outflow and lake samples (0.95 mm; 950 µm; see Appendix Table C2.8). Microplastics in sediment samples had an average length of 1.17 mm (1170 µm; see Appendix Table C2.8); again, longer than Welsh et al. (2022) who found the average length to be 0.54 mm (540 µm). The average length in precipitation samples was 1.43 mm (1430 µm), higher than the length of microplastics found in the Muskoka-Haliburton region's precipitation (0.68 mm; 680 µm; Welsh et al., 2022). Once again most likely owing to the distance from pollution sources. The average length of microplastics found in the bio-monitor samples was 1.14 mm (1140 µm) with the average fiber length being 1.33 mm and the average fragment length being 0.43 mm, smaller than the length found in the precipitation samples. There were also more fragments found in the bio-monitors than precipitation (Table 2.4). The length of microfibers found in bio-monitors were similar to

Roblin et al. (2020a), who found the average fiber length to be 1.02 mm in passive bio-monitor samples in Ireland.

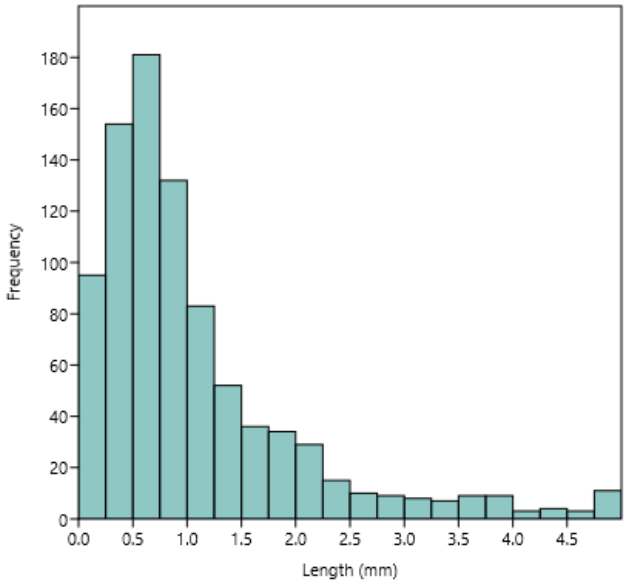


Figure 2.5- Histogram showing the distribution of microplastic lengths across all samples (precipitation, inflow streams, lake water, sediment, lake outflow, etc.) collected from Lake Scugog during May–October 2021.

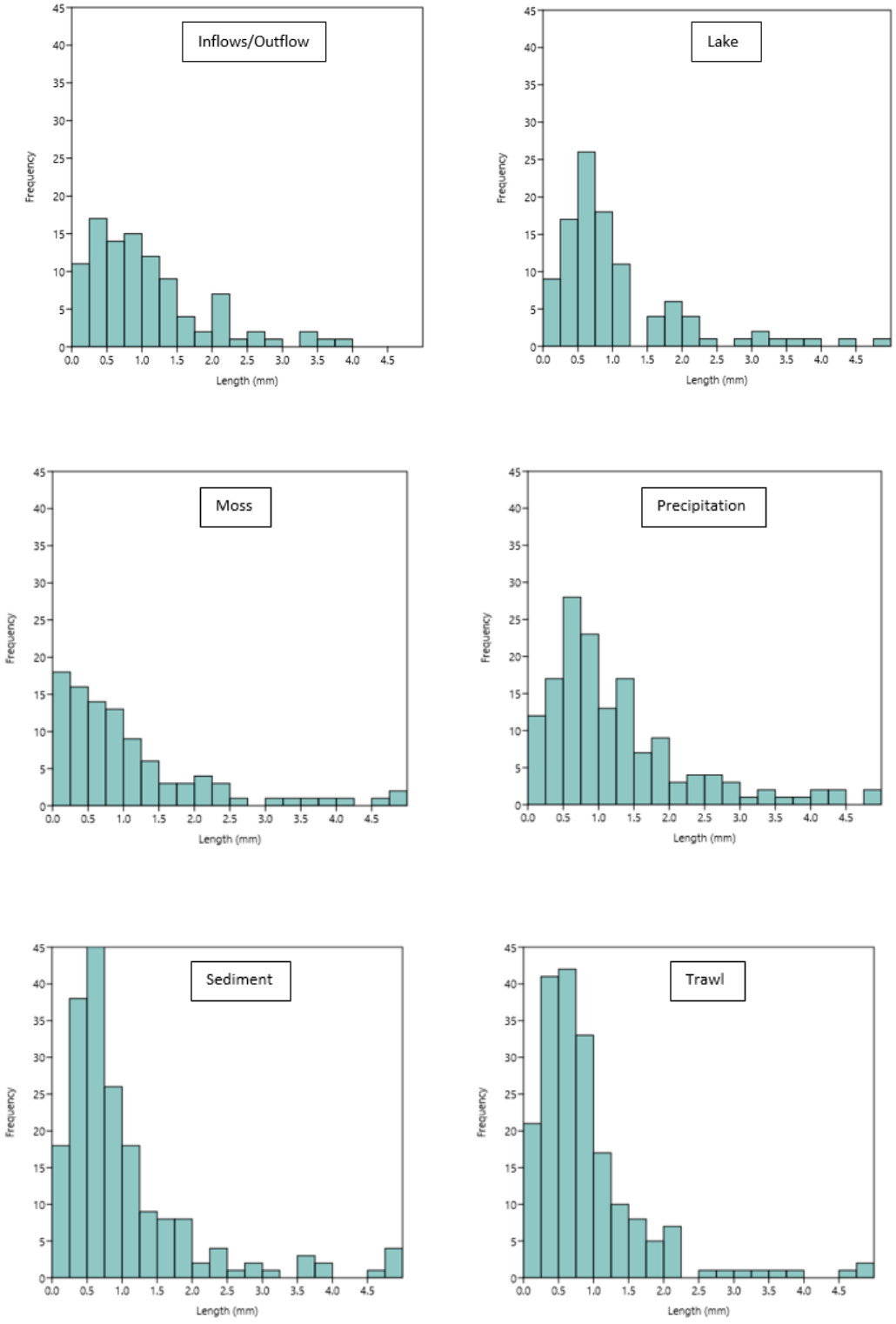


Figure 2.6- Length of microplastics in each sample type collected from Lake Scugog during May–October 2021.

2.4.5 Microplastic Colour

Microplastic colour is generally identified to estimate possible microplastic sources however, this is a very limited approach due to the subjective nature of colour identification. In general, there were a few dominant colours across all shapes; most fibers were black (50.6%), while the dominant colour for fragments/films was white (18.9%), blue was the next most common colour for both shape categories (Figure 2.7). Similarly, Felismino et al. (2021) found blue and black to be the most common colours, and Welsh et al. (2022) found blue to be the dominant colour. Black and blue are the dominant colour of most clothes, e.g., denim jeans are worn by an estimated 46–56% of Canadians every day (Athey et al., 2020). Further, the average Canadian also washes their denim more than necessary, after only two wears, which potentially releases more microplastics into the environment. White and clear may have been abundant in fragments/films due to the loss of colour during environmental degradation, or bleaching during sample digestion (Dris et al., 2015; Baldwin et al., 2020).

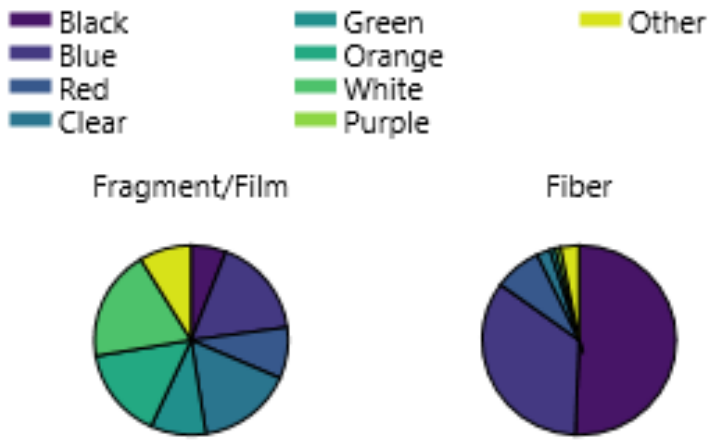


Figure 2.7- Pie charts representing the proportion of different colours found across all samples for fragments/films (left) and fibers (right) for Lake Scugog sampled during May–October 2021.

2.4.6 Microplastic Fluxes and Particle Budget

The daily flux of microplastics into Lake Scugog was highest from atmospheric deposition (19.51 mp/m^2/day), while the daily flux of microplastics out of the lake was highest in the lake outflow (3.05 mp/m^2/day; Table 2.5). The dominant flux within the study area was atmospheric deposition, nearly four times greater than the greatest output. Deposition is important, but only bulk deposition was measured in this study, therefore it is important to quantify the potential contribution from dry deposition. Welsh et al. (2022) found similar results across three lakes in the Muskoka-Haliburton region; atmospheric deposition had the highest daily flux entering the lakes (3.84 mp/m^2/day) and the lake outflow was the highest daily flux exiting the lakes (5.20 mp/m^2/day), however the lake outflow was higher than the atmospheric deposition. Klein & Fischer (2019) found a much higher daily atmospheric deposition flux of 275 mp/m^2/day across six sampling sites in Hamburg, Germany. The differences in concentrations across the three studies is most likely due to different methods and population densities, with the Muskoka Haliburton region having the lowest and Hamburg having the highest.

The inflow's daily flux range for the current study was 0.99–3.67 (S9–S3) mp/m^2/day (Table 2.5). Similarly, Welsh et al. (2022) found the daily flux for inflows into the three lakes in the Muskoka-Haliburton region ranged between 1.24–2.98 mp/m^2/day. In the current study, the S3 inflow is located in the Nonquon river, the wastewater treatment facilities discharge into the Nonquon river which may explain the high concentration of microplastics present in this inflow. Microplastic sedimentation had a daily flux of 2.13 mp/m^2/day, which was similar to the daily flux that was found for sediment in the Muskoka-Haliburton region lakes (average of 1.90 mp/m^2/day; Welsh et al., 2022).

The microplastic particle budget incorporated the dominant inputs (precipitation and stream inflows) and outputs (sedimentation and the lake outflow) of Lake Scugog catchment (Figure 2.8). The total daily input of microplastics into the lake was 2491 $x10^6$ mp/day, while the output was 1761 $x10^6$ mp/day, resulting in 29% being retained in the lake (Table 2.5). The retention rate was similar to three lakes in the Muskoka-Haliburton region, that ranged from 30–45% (Welsh et al., 2022). The total number of microplastics in the lake was 2.62 $x10^{11}$ mp with a residence time of one year in Lake Scugog. Further, 89% of microplastics atmospherically deposited onto the terrestrial catchment was estimated to be retained; 10,602 $x10^6$ mp/day were deposited in precipitation on the terrestrial environment and 1,164 $x10^6$ mp/day left the terrestrial environment via stream inflows.

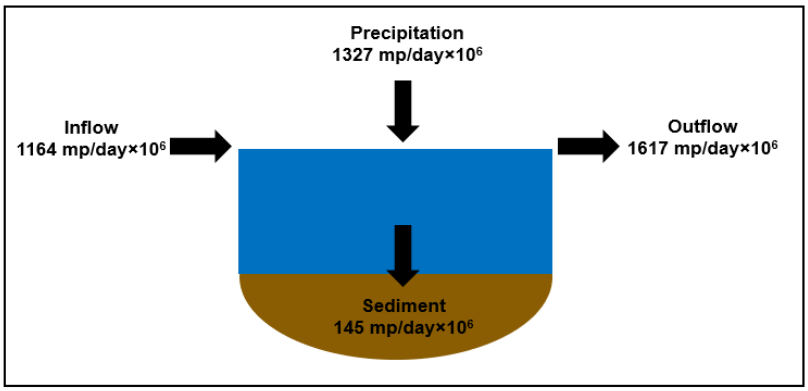


Figure 2.8- The particle budget for Lake Scugog showing the number of microplastics per day entering and leaving the catchment resulting in 29% microplastic retention in the lake.

Table 2.5- Average concentration, drainage area, discharge, daily number, and daily microplastic flux stream inflows, lake outflow, sediment, precipitation for Lake Scugog based on microplastics sampled during May–October 2021.

Sample Type	Site ID	Concentration (mp/L)	Drainage area (ha)	Discharge (m^3/sec)	Number (mp/day×10^6)	Flux (mp/m^2/day)
Precipitation	R1-2	8.08	6800	1.90	1327.0	19.51
Inflow	S1	3.07	1270	0.11	30.2	2.38
Inflow	S3	3.36	18470	2.33	677.6	3.67
Inflow	S4	2.40	1040	0.12	24.5	2.35
Inflow	S6	1.40	3790	0.33	40.4	1.07
Inflow	S7	2.90	2220	0.19	47.8	2.15
Inflow	S8	3.00	1093	0.09	24.3	2.23
Inflow	S9	1.33	139	0.01	1.4	0.99
Inflow*	ungauged	2.36	18148	1.56	317.6	1.75
Lake Outflow	S10	3.05	52970	6.14	1616.7	3.05
Sediment	Sd1-6	2.19	6800	354.40	144.5	2.13
Lake Input					2491	
Lake Output					1761	

* Note: S2 and S5 are not included as they are part of S3; S2 was included in S3 and S5 was part of the same inflow as S3, S3 was used as it is closer to the lake.

2.4.7 Preliminary Estimates of Smaller Size Microplastics

The average concentration and range of microplastics within samples substantially increased once the fluorescent dye procedure was performed (Table 2.6). The majority (68.6–95.3%) of microplastics found in all samples were 20–100 µm (0.02–0.1 mm; Table 2.6). The concentration of microplastics increased the most in sediment samples; 95.3% of microplastics estimated were 20–100 µm (Table 2.6). Trawl and precipitation samples had the lowest increase, 68.6% and 68.8% of microplastics were 20–100 µm respectively (Table 2.6). Further, the majority (91.3%) of small microplastics were fragments (Table 2.7). Similarly, Allen et al. (2019) found that with decreasing microplastic size the amount of microplastics, and in particular fragments, increased; most fibers were >100 µm and most fragments were between 20–100 µm,

with fragments accounting for 88% of the microplastics. While other studies have found fibers to be more dominant (Dris et al., 2015; Felismino et al., 2021), the differences between these studies and the current study are most likely due to different methods, i.e., detection of particles at different cut-off sizes and detection methodology.

The concentration of microplastics >100 µm in sediment was 2.2 mp/g while microplastics >20 µm was 47.1 mp/g (Table 2.6). This was the greatest increase; sediments may have had the highest increase due to more degradation occurring by the time they reach the sediments. Although the concentration of microplastics is higher, the overall mass of microplastics may not have increased that much. Although these are only preliminary estimates of microplastics in the size range 20–100 µm it does show the importance of identification techniques used for counting microplastics. When extracting microplastics the type of equipment used also has an affect on the size of microplastics identified.

Table 2.6- Average concentrations of microplastics >100 µm and estimated concentrations of microplastics >20 µm (up to 5000 µm) across the different sample types

Sample Type	Concentration >100 µm	Concentration >20 µm	20–100 µm (%)
Tributary (mp/L)	2.4	9.8	75.5
Lake (mp/L)	2.9	11.7	75.2
Trawl (mp/m^3)	3.2	10.2	68.6
Sediment (mp/g)	2.2	47.1	95.3
Precipitation (mp/L)	8.1	26.0	68.8
Moss (mp/g)	2.5	26.4	90.5

Table 2.7- Percentages of fibers and fragments found in all samples for both size categories. Note: there were no films/other found in the 20–100 µm category.

	Fiber (%)	Frag (%)
20–100 µm	8.7	91.3
>100 µm	77.4	12.1

2.4.8 Limitations

There are a number of limitations in this study: (1) only dominant inputs and outputs were examined, (2) sampling was only performed for a six-month period, (3) small volume samples (1 L) were collected for stream inflows/outflow and lake water, and (4) there is limited knowledge on sedimentation rates of microplastics. Potential inputs missing from this study included street runoff, and direct human inputs (e.g., cottages, boating, fishing, etc.). The flux presented was based on the known concentrations for the six-month sampling period, and the most recent average discharge data (2005–2008) for the Lake Scugog catchment. It was assumed that the microplastic concentration determined for the six-month period was representative for the entire year; however, we have no knowledge of microplastic pathways and abundance during winter, under ice, and during snow melt. The microplastic sedimentation rate was based on the sedimentation rate of mineral sediment in Lake Scugog (near the Port Perry Harbor) for the top 5 cm (core depth of 52 cm) that was sampled as there is limited knowledge on the microplastic sedimentation rate in freshwater ecosystems. Also, ideally a second outflow sampling location should have been placed directly outside of the lake. Even with the limitations presented here, this study has addressed known knowledge gaps within the literature and is one of the only studies that has determined a microplastic particle budget. This study has attempted to understand the fate of microplastics within a lake and has identified major pathways of microplastics as well as how they are removed or permanently stored (Rochman & Hoellein,

2020; Brahney et al., 2021). Understanding the tendencies of microplastics in a freshwater system allows for better management.

2.5 Conclusion

This study estimated the abundance and distribution of microplastics as well as their daily inputs and outputs in Lake Scugog catchment, Ontario, using a particle balance approach; the inputs included atmospheric deposition and stream inflows, while the outputs included sedimentation and the lake outflow. In addition, microplastics >20 µm were detected using a fluorescent dye procedure. The dominant flux within the study area was atmospheric deposition, nearly four times greater than the greatest output; the lake outflow. The estimated residence time of microplastics in Lake Scugog was one year. However, a greater number of microplastics entered Lake Scugog than left, resulting in 29% being retained. Atmospheric deposition is important, but only bulk deposition was measured in this study, therefore it is important to quantify the contribution from dry deposition. Microplastics <100 µm were the most common across all environmental samples; with the largest proportion of smaller microplastics occurring in sediment samples. Better management techniques for plastic waste can only be implemented by understanding and quantifying the major pathways of microplastics.

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2.7 Appendix

Appendix A- Methods

Table A2.1- Location (latitude, longitude) of study sites in the Lake Scugog catchment, including inflows/outflow (S), lake (L), trawl (T), sediment (Sd), precipitation (R), and bio-monitors (M). Lake and sediment locations were the same and the trawl was performed at L1. Note: S4 was only sampled once (May). See Figure 2.1 for mapped location.

Site ID	Sample Type	Latitude	Longitude
S1	Inflow	44.23293	-78.91892
S2	Inflow	44.19159	-78.98571
S3	Inflow	44.20042	-78.94947
S4	Inflow	44.12035	-78.95384
S5	Inflow	44.08667	-79.00630
S6	Inflow	44.13188	-78.82872
S7	Inflow	44.10288	-78.93047
S8	Inflow	44.11427	-78.84506
S9	Inflow	44.10543	-78.94545
S10	Outflow	44.32772	-78.73026
L1, Sd1, T1	Lake, Sediment, Trawl	44.20234	-78.79567
L2, Sd2	Lake, Sediment	44.15724	-78.84243
L3, Sd3	Lake, Sediment	44.20772	-78.85471
L4, Sd4	Lake, Sediment	44.20477	-78.89927
L5, Sd5	Lake, Sediment	44.17678	-78.91648
L6, Sd6	Lake, Sediment	44.10895	-78.93733
R1	Precipitation	44.11064	-78.96056
R2	Precipitation	44.13183	-78.82906
M1	Bio-Monitor	44.21717	-78.83932
M2	Bio-Monitor	44.23293	-78.91892
M3	Bio-Monitor	44.18070	-78.93415
M4	Bio-Monitor	44.10540	-78.94537
M5	Bio-Monitor	44.16972	-78.89462
M6	Bio-Monitor	44.19776	-78.89171
M7	Bio-Monitor	44.20483	-78.86094
M8	Bio-Monitor	44.15699	-78.83699
M9	Bio-Monitor	44.21294	-78.77307

Table A2.2- The number of microplastics in blanks for all sample types, these counts include both field and laboratory blanks.

Sample Type	Number of Microplastics
Inflow/Outflow	0.6
Lake	0.6
Trawl	3.3
Sediment	2.0
Precipitation	0.6
Bio-monitor	1.2

Table A2.3- The variability for all field duplicate samples and triplicate analytical sub-samples (i.e., sediment). The CV% is presented for all triplicates and the RPD% is presented for all duplicates.

Sample Type	Site ID	Variability Test	Variability (%)
Inflow	Jul S3	Duplicate	200.0
Inflow	Aug S2	Duplicate	40.0
Inflow	Sep S1	Duplicate	0.0
Inflow	Oct S2	Duplicate	0.0
Lake	Jul L6	Duplicate	25.0
Lake	Aug L3	Duplicate	66.7
Lake	Oct L2	Duplicate	28.6
Sediment	Oct Sd1	Triplicate	6.9
Sediment	Oct Sd2	Triplicate	16.7
Sediment	Oct Sd3	Triplicate	23.6
Sediment	Oct Sd4	Triplicate	11.1
Sediment	Oct Sd5	Triplicate	50.0
Sediment	Oct Sd6	Triplicate	28.4
Moss	Jun-Aug M1	Duplicate	0.0
Moss	Jun-Aug M2	Duplicate	100.0
Moss	Jun-Aug M3	Duplicate	22.2
Moss	Jun-Aug M4	Duplicate	50.0
Moss	Jun-Aug M5	Duplicate	66.7
Moss	Jun-Aug M6	Duplicate	28.6
Moss	Jun-Aug M7	Duplicate	120.0
Moss	Jun-Aug M8	Duplicate	138.5

Moss	Jun-Aug M9	Duplicate	90.9
Moss	Aug-Oct M1	Duplicate	40.0
Moss	Aug-Oct M2	Duplicate	76.9
Moss	Aug-Oct M3	Duplicate	0.0
Moss	Aug-Oct M4	Duplicate	66.7
Moss	Aug-Oct M5	Duplicate	0.0
Moss	Aug-Oct M6	Duplicate	50.0
Moss	Aug-Oct M7	Duplicate	85.7
Moss	Aug-Oct M8	Duplicate	0.0
Moss	Aug-Oct M9	Duplicate	85.7

The particle budget was based on all microplastic types combined rather than individual shapes as there were sampling limitations. When separated into a fiber and fragment particle budget, more fragments leave the lake than enter (Table A2.4). This may be for a couple reasons: (1) the fibers may have degraded into fragments, or (2) the 1 L grab samples may not have been a good representation for shape diversity across the samples (see Appendix B).

Table A2.4- Individual particle budget for fragments and fibers, providing an estimate of the amount entering the lake, leaving the lake, and being retained within the lake.

	Fibers	Fragments
Lake Input ($\times 10^6$ mp/day)	2045	128
Lake Output ($\times 10^6$ mp/day)	1723	211
Difference ($\times 10^6$ mp/day)	322	–83
Retained in Lake (%)	16	–65

Table A2.5- The statistical analysis tests performed for all sample types along with their p-values.

Sample Type	Statistical Test	p-value
Inflow/Outflow (Spatial)	One-way Anova	p = 0.66
Inflow/Outflow (Seasonal)	One-way Anova	p = 0.10
Lake (Spatial)	One-way Anova	p = 0.69
Lake (Seasonal)	One-way Anova	p = 0.15
Sediment	One-way Anova	p = 0.000017
Precipitation	One-way Anova	p = 0.11
Bio-monitor	One-way Anova	p = 0.75

Table A2.6- Scale factors for all sample types, the scale factors were multiplied by the >100 µm counts to determine the total (>20–100 µm + >100 µm) counts for all microplastics.

Sample Type	Scale Factor
Inflows/Outflow	4.11
Lake	4.11
Trawl	3.17
Sediment	18.18
Precipitation	3.22
Moss	10.71

Appendix B- High Volume Samples

During October 2021 both low volume (1 L) and high volume (500 L) samples were performed at three lake sites (L2, L4, L6) to compare the concentration of microplastics and sample volumes. The low volume samples were taken into a wide-mouth glass jar, the jar was rinsed with the sample water at each site, and filled at a depth of 10 cm. The high volume samples were taken using a 50 µm plankton net and water pump. The water (500 L) was pumped (approximately 15 min) into the plankton net, the cod end was then emptied and rinsed into a glass jar. A field blank was performed after the sampling was complete, the net and cod end were

rinsed into a stainless-steel bowl with one-liter of filtered B-pure water, which was then transferred to a glass jar.

The average concentration for the low volume sample was 5.1 mp/L and the high volume was 1.1 mp/L (Table B2.1). While the grab sample is limited as the sample volume is low, the high volume sample is also limited as the water needs to go through a 50 µm net resulting in a loss of microplastics. The high volume sample attains a more diverse concentration of shapes, while the low volume sample is limited. The low volume samples all consisted of only fibers, while the high volume samples contained fragments, fibers, films and other; with fibers having the highest concentration.

Table B2.1- Plankton net and grab lake water samples performed in October 2021, comparing the concentration of microplastics in high volume (500 L) and low volume (1 L) samples.

Sample Type	Concentration (mp/500 L)	Concentration (mp/L)
Pump with Plankton Net	269.0	0.5
	527.0	1.1
	895.0	1.8
Average	563.7	1.1
Grab	-	4.4
	-	8.4
	-	2.4
Average	-	5.1

Appendix C- Results

Table C2.1- Concentration (mp/L) of microplastics at every inflow/outflow site for all months sampled. Note: some sites were not sampled every month.

	S1	S2	S3	S4	S5	S6	S7	S8	S9	S10
May	4.4	0.4	-	2.4	3.4	-	4.4	3.4	4.4	-
Jun	2.4	1.4	3.4	-	0	3.4	1.4	8.4	0.4	-
Jul	3.4	0.4	1.2	-	2.4	1.4	0.4	0.4	0.4	0
Aug	4.4	1.9	2.4	-	3.4	0.4	1.4	-	0.4	0.4
Sep	1.4	5.4	6.4	-	2.4	0.4	7.4	2.4	2.4	7.4
Oct	2.4	1.4	3.4	-	0	1.4	2.4	0.4	0	4.4

Table C2.2- Concentration (mp/L) of microplastics at every lake site for all months sampled.

	L1	L2	L3	L4	L5	L6
May	7.4	1.4	1.4	5.4	7.4	5.4
June	3.4	5.4	2.4	2.4	1.4	0.4
July	0.4	0.4	1.4	1.4	2.4	7.4
Aug	3.4	0.4	0.9	1.4	0.0	3.4
Sept	4.4	5.4	2.4	3.4	3.4	0.4
Oct	2.4	2.9	1.4	3.4	3.4	3.4

Table C2.3- Concentration (mp/m^3) of microplastics at the trawl site in triplicate, sampled once in October.

	T1
Trawl 1	4.3
Trawl 2	1.9
Trawl 3	3.4

Table C2.4- Concentration (mp/g) of microplastics at the sediment sites in triplicate analytical sub-samples, sampled once in October.

Sd1 #1	3.6
Sd1 #2	3.2
Sd1 #3	3.2
Sd2 #1	3.6
Sd2 #2	4.2
Sd2 #3	3.0
Sd3 #1	2.2
Sd3 #2	2.6
Sd3 #3	1.6
Sd4 #1	2.0
Sd4 #2	1.8
Sd4 #3	1.6
Sd5 #1	0.8
Sd5 #2	1.2
Sd5 #3	0.4
Sd6 #1	1.8
Sd6 #2	1.0
Sd6 #3	1.6

Table C2.5- Concentration (mp/L) of microplastics and volume (L) of precipitation collected at the precipitation sites for all months sampled.

	Port Perry		Blackstock Creek	
Month	Liters	mp/L	Liters	mp/L
June	4.85	5.77	5.32	8.59
July	4.11	11.30	5.32	4.64
Aug	1.58	16.43	1.48	9.52
Sept	5.07	7.46	9.50	4.22
Oct	2.18	9.46	4.93	3.43

Table C2.6- Concentration (mp/g) of microplastics at the bio-monitor sites for all months sampled.

	Jun–Aug	Aug–Oct
M1	1.0	1.6
M2	1.0	6.0
M3	3.8	3.2
M4	3.2	2.1
M5	0.5	3.2
M6	2.7	3.2
M7	1.6	2.7
M8	6.0	2.1
M9	4.9	2.7

Table C2.7- All polymer types found across all sample media along with the hit rate, which represents the correlation between the sample spectra and polymer library spectra; water includes inflow, outflow, and lake water. The spectrum for each sample were uploaded to an online library of polymer spectra (OpenSpecy: https://openanalysis.org/openspecy/), which reported the correlation (hit rate) between the sample and library entries. The higher the hit rate the greater the likelihood of exact polymer identification.

Media	Shape	Polymer	Hit Rate
Water	Fiber	Polypropylene	0.38
Water	Fragment	Polypropylene	0.44
Water	Fiber	Nitrile Rubber	0.45
Water	Fiber	Polyester	0.32
Water	Fiber	Polyester	0.47
Water	Fiber	Polypropylene	0.95
Water	Fiber	Polyester	0.66
Water	Fiber	Polycarbonate	0.24
Water	Fiber	Nylon	0.45
Water	Fiber	Nylon	0.47
Water	Fiber	Polyvinylchloride	0.32
Water	Fiber	Polyvinylchloride	0.31
Water	Fiber	Polyester	0.21
Water	Fiber	Polyester	0.62

Water	Fiber	Polystyrene	0.27
Water	Fiber	Polyester	0.28
Water	Fiber	Polyester	0.89
Water	Fiber	Polyester	0.95
Water	Fiber	Polyester	0.84
Water	Fiber	Polyester	0.94
Water	Fiber	Polyester	0.95
Water	Fiber	Polyester	0.49
Water	Fiber	Polyester	0.60
Water	Fiber	Polyester	0.91
Water	Fiber	Polyester	0.90
Water	Fiber	Polyester	0.97
Water	Fiber	Polyester	0.88
Water	Fiber	Nylon	0.55
Water	Fiber	Nitrile Rubber	0.23
Water	Fiber	Polyester	0.38
Water	Fiber	Polyester	0.65
Water	Fiber	Polyvinylchloride	0.93
Water	Fiber	Polyester	0.51
Water	Fragment	Polyester	0.85
Trawl	Fiber	Polypropylene	0.92
Trawl	Fiber	Polyvinyl alcohol	0.81
Trawl	Fiber	Polyester	0.53
Trawl	Fiber	Polyester	0.56
Trawl	Fiber	Polyester	0.94
Trawl	Fragment	Polyvinyl acetate	0.86
Trawl	Fiber	Polyester	0.85
Precipitation	Fiber	Nitrile Rubber	0.46
Precipitation	Fiber	Polyvinyl acetate	0.34
Precipitation	Fiber	Polyester	0.98
Precipitation	Fiber	Polyvinyl acetate	0.32
Precipitation	Fiber	Nitrile Rubber	0.60
Precipitation	Fiber	Polyester	0.98
Precipitation	Fiber	Polyester	0.91
Precipitation	Fiber	Nylon	0.40
Precipitation	Fiber	Polyester	0.63
Bio-monitor	Fiber	Polyester	0.29
Bio-monitor	-	Nitrile Rubber	0.77
Bio-monitor	-	Nylon	0.28
Bio-monitor	Fragment	Polypropylene	0.78
Bio-monitor	Fiber	Polypropylene	0.96
Bio-monitor	Fragment	Nitrile Rubber	0.90
Bio-monitor	Fiber	Polyester	0.91
Sediment	Fiber	Polyester	0.61

Sediment	Fragment	Polystyrene	0.80
Sediment	Fiber	Polyester	0.86
Sediment	Fiber	Polystyrene	0.95
Sediment	-	Polyvinylchloride	0.95
Sediment	-	Polycarbonate	0.45
Sediment	-	Polypropylene	0.33
Sediment	-	Polyvinylchloride	0.26
Sediment	-	Polypropylene	0.34
Sediment	-	Polycarbonate	0.39

Table C2.8- Average length of microplastics for each sample type.

Sample Type	Average Length (mm)
Inflow/Outflow	1.15
Lake	1.06
Trawl	0.95
Sediment	1.17
Precipitation	1.43
Bio-monitor	1.14
Total	1.17

Chapter 3: Dry deposition of atmospheric microplastics in Port Perry, Ontario

3.1 Abstract

There is growing recognition of the importance of the atmosphere as a pathway of microplastic deposition to background regions. Atmospheric deposition of microplastics includes both wet and dry deposition, however few studies have focused on dry deposition. In this study, the dry deposition of microplastics in Port Perry, Ontario, was determined using a laboratory measured settling velocity for polyester fibers and observations of ambient air concentrations of microplastics. The average dry deposition rate of microplastics in Port Perry was found to be 1257 mp/m^2/day during November 1–5, 2021. This rate was determined using a measured settling velocity of 0.11 m/s and ambient air concentrations of 0.132 mp/m^3. The dry deposition rate was high when compared to bulk deposition in Port Perry, this may be due to the settling velocity being determined in a laboratory setting with no meteorological conditions applied to it.

3.2 Introduction

Plastics are considered an environmental contaminant owing to their chemical additives, the pollutants that bind to them, and the risk of physical harm when ingested (Cole et al., 2011; Zhao et al., 2014). Plastic production rates have increased drastically during the past seven decades, from two metric tons in 1950 to 380 metric tons in 2015 (Bianco & Passananti, 2020; Allen et al., 2019; Geyer et al., 2017). At the same time, the rate of plastic pollution has also increased, with 55% of all plastic produced in 2015 discarded to landfills or the natural environment (Geyer et al., 2017; Bagaev et al., 2017; Lusher et al., 2015; Ritchie & Roser, 2018).

There has been a growing focus on microplastics (plastic particles <5 mm; 5000 µm) during the last few decades, as they have been widely observed in both aquatic and terrestrial

environments (Cai et al., 2017; Anderson et al., 2016; Felismino et al., 2022). While the majority of studies have focused on microplastics in marine ecosystems, the atmosphere is now recognized as “an enormous microscopic problem” (Bianco & Passananti, 2020; Liu et al., 2019b), as it is a major pathway of microplastics to remote regions (Evangeliou et al., 2020; GOC, 2020) This is evident by observations of microplastics in remote areas of the world, such as the Pyrenees, the Swiss Alps, and the Arctic (Allen et al., 2019; Bergmann et al., 2019). Generally, microplastics in the atmosphere are dominated by fibers, particularly polyester fibers (Welsh et al., 2022; Prata et al., 2020; Dris et al., 2017).

Atmospheric deposition of microplastics includes both wet and dry deposition; wet deposition occurs through rain or snowfall, where microplastics are washed out of the atmosphere by rainwater droplets, while dry deposition occurs through particle settling. A number of studies have reported wet or bulk deposition of microplastics (e.g., Welsh et al., 2022; Klein & Fischer, 2019; Allen et al., 2019; Roblin et al., 2020); however, few if any studies have focused on dry deposition. Ultimately, the quantification of dry deposition provides a more holistic understanding of deposition, transport, and the nature of atmospheric microplastics (Abbasi & Turner, 2021).

Dry deposition is generally determined from observations of air concentrations and estimates of particle settling velocities. A few studies have presented preliminary theoretical estimates for the settling velocity of microplastics (Wright et al., 2020; Allen et al., 2019); alternatively, laboratory-based experiments can be used to directly measure settling velocity following studies on non-plastic particles (Richards-Thomas & McKenna-Neuman, 2020).

The overall objective of this study was to determine the dry deposition of microplastics in Port Perry, Ontario, using a laboratory measured settling velocity and ambient air concentrations

of microplastics. Polyester microfibers were used to determine the settling velocity as they are the most common microplastic found in the atmosphere (Chapter 2; Liu et al., 2019a; Welsh et al., 2022; Wright et al., 2019). The settling velocity was determined using a drop column and a Dantec Laser Doppler Anemometer (LDA). Daily ambient air concentrations of microplastics were measured in Port Perry for five consecutive days in November 2021 using an active high-volume air sampler.

3.3 Methods

3.3.1 Study Area

The town of Port Perry (population 9,553; Statistics Canada, 2021) is located within the Region of Durham, south-central Ontario, Canada. It is approximately 80 km northeast of Toronto (Figure 3.1) and is the main urban center on Lake Scugog (Kawartha Conservation, 2010). The current study measured ambient air concentrations of microplastics at one site (44.10747, –78.94451) in Port Perry (Figure 3.1). Recently, the bulk deposition of microplastics was also measured in Port Perry (44.11064, –78.96056) using a NILU microplastic bulk deposition collector (PN. 9735) for five months (June–October) in 2021; these data provide wet deposition and a portion of dry deposition. The average temperature during the current study period was 2.5°C, the average precipitation was 0.34 mm (see Appendix Table A3.1) and the wind direction was mainly from the south-west with an average speed of 8.2 km/hr (Figure 3.1). Traffic volumes for the area (1.7 km from the study site) during peak travel hours (7–9 am and 4–6 pm) are approximately 74 cars per hour (GHD Consultants, 2019).

Figure 3.1 - Location of the ambient air sampler in Port Perry, next to Lake Scugog. The wind rose shows the wind direction and speed during November 1–5, 2021 mainly coming from the south-west.

3.3.2 Field Sampling

During November 1–5, 2021 (5 days), an active high-volume air sampler (which sampled at 67.96 m^3/hr; Model: Hi-Q HVP-3300BRL) was placed on the roof of a municipal building (44.10747, –78.94451; approximately 3 m above ground) to avoid disturbance and to ensure unimpeded airflow. The sampler contained a 22.5 cm by 17.5 cm cellulose filter paper that was changed daily (~24 hrs; see Appendix Table A3.2). The exposed filter was stored in an aluminum foil container to avoid contamination. A field blank was performed by placing a blank filter paper on the sampler and removing it; the blank was processed in the same manner as the field samples.

3.3.3 Laboratory Analysis

In the laboratory, three circles (analytical sub-samples of 5 cm diameter from three random locations) were cut out of each filter paper including the blank from the high-volume air sampler (n=18). All filter circles underwent visual analysis using a stereomicroscope with a digital camera (Amscope with Infinity 2 Teledyne Lumenera camera). Brightfield microscopy is widely used to detect microplastics >50 µm, although in general the detection size is typically >100 µm (>0.1 mm), and the detection of clear plastic particles is difficult (Erni-Cassola et al., 2017; Labbe et al., 2020). However, clear plastic particles will auto fluoresce under certain wavelengths, such as a blue-light (wavelength 460 nm) source (Stanton et al., 2019). In the current study, brightfield microscopy and blue-light fluorescence were performed on all samples.

Visual analysis of plastic particles adhered to a widely used identification protocol (Roblin et al., 2020; Windsor et al., 2019; Norén, 2007) based on five criteria: (i) the particle is unnaturally coloured; (ii) it is homogenous in material and texture with no cell structure or offshoots present; (iii) the particle is not brittle and does not break when poked, tugged or compressed with tweezers; (iv) it is shiny or glossy in appearance; and (v) there is limited fraying with no similarity to natural particles. Suspected plastic particles were photographed and then confirmed using a hot needle test following Roblin et al. (2020). A hot needle was pressed against the particle, if it melted it was classified as plastic, the image of the microplastic was then measured by converting the number of pixels to a known length in millimeters using the image processing software, ImageJ. The length, width, colour, and type (fiber or fragment) of each microplastic were recorded. Filter papers were then examined under blue-light to identify auto-fluorescing particles; if a particle fluoresced it was hot-needled, these microplastics were not photographed as the fluorescing particles created a halo that made capturing and measuring the image difficult.

3.3.4 Quality Control and Assurance

Glass and metal materials were used where possible throughout field and laboratory work, however some plastic needed to be used (petri dishes, B-pure water spray bottle, etc.). For ambient air, three 5 cm subsamples from each filter (triplicates) were analyzed to assess variability and confirm that the area analyzed (3 × 5 cm circles) was adequate. Open air blanks consisting of a clean glass fiber filter paper (Fisherbrand^{TM} G6 1.6 µm) in a petri dish were exposed for one hour during both the extraction and counting processes (n=4) and used to determine potential in-lab contamination. The period of time that samples were exposed during all laboratory procedures (extraction, counting, etc.) was also recorded. Further, the field blanks were used to determine potential contamination through the entire laboratory procedure. All equipment and sample containers were triple rinsed with filtered B-pure water (to remove any possible microplastics).

3.3.5 Settling Velocity of Polyester Microfibers

The settling velocity was determined for microplastic polyester fibers obtained from a 100% polyester scarf. The material was repeatedly cut with a rotary cutter to obtain particle fibers predominantly <1 mm (<1000 µm). Anti-static gloves were worn while cutting and handling the fibers during all laboratory tests. The polyester fibers were sieved to select microfibers <1 mm; however, due to the rod-like shape of the fibers, some longer fibers passed through the sieve as well. This size category was chosen as microfibers <1 mm are generally the most common lengths observed in the environment (Allen et al., 2019; Roblin et al., 2020; Dris et al., 2016; Wright et al., 2020). Further, this size was within the range of microplastics found in bulk deposition in Port Perry (see Chapter 2).

The length of the laboratory manufactured microfibers was measured to determine the accuracy of the cutting and sieving. A 4 g ± 0.003 g sample was released through a sieve with a

1.5 mm mesh size onto a microscope slide (2.5 cm by 7.5 cm); ten replicates were performed. The subset of fibers that landed on the microscope slide were photographed (Digital Single-Lens Reflex Canon Camera) and measured using ImageJ; approximately 1500 fibers were manually measured (50–350 fibers per slide; see Appendix Figure A3.1 for example photographs). Fibers that extended over the edge of the slide or overlapped other fibers were eliminated from the analysis.

The LDA and fall column (242 cm tall) were placed in an environmentally controlled chamber (constant temperature of 20 ± 1°C and minimal air movement) and used to determine the settling velocity of the polyester microfibers. Other meteorological conditions (wind, humidity, etc.) were not taken into account when determining the settling velocity. The LDA attains high-resolution velocity measurements using four laser beams, without disturbing the air column (Richards-Thomas & McKenna-Neuman, 2020). It measures the doppler shift of light that is scattered from particles passing through the intersection of lasers (Buchhave & George, 1979). The laser beam was split into two beams each with a wavelength of $\lambda = 0.660$ μm, this pair of beams measured a single velocity component, perpendicular to the bisector of the beams (Durst et al., 1981). To determine the particle velocity in another dimension an additional pair of beams was required, these additional beams needed to have a different wavelength ($\lambda = 0.785$ μm) so that the signals for each component could be distinguished (Durst et al., 1981; Drain, 1980; Richards-Thomas & McKenna-Neuman, 2020). The size and shape of a particle affects the amount of beam scattering, because of this, calibration tests were performed on the LDA to attain the correct gain (boosted the signal/noise ratio) and validation (averaged the measurements to avoid collecting "noise" data) settings for polyester microfibers (see Appendix B).

The LDA was positioned in front of the fall column, which was constructed from a Sonotube (inner diameter 29 cm, the top was covered, and a 16 cm opening was cut out to deploy fibers) with a softwood base (height 5 cm by width 10 cm). The four beams from the LDA entered through a Perspex® window (9 cm by 23 cm, 37 cm from the floor), which was cleaned between tests (see Appendix Figure A3.2). The beams were positioned so that their intersection point was directly in the center of the fall column, 182 cm below the point of fiber release and 60 cm from the floor to reduce the detection of upward moving particles. There were two congruent vents (10 cm by 20 cm) approximately 15 cm from the floor, these were used to prevent the development of a pressure gradient within the fall column and decrease the amount of upward moving particles (Figure 3.2). Fibers were released manually at the top of the column (through the 16 cm opening) from a glass jar with a metal screen lid (1.5 mm mesh size) by gently shaking for approximately 20–30 seconds. Each replicate weighed 4 g ± 0.003 g and provided approximately 1000–2500 particle velocity measurements. The lid of the release container was grounded between experiments to reduce static charge. A collection sheet was placed at the bottom for recovery and re-use of the fibers, this was changed between every 10 replicates and the recovered microfibers were stored in glass jars. A typical fall column run lasted between 10–15 minutes, which was ample time for all fibers to reach the collection sheet. After 5–7 minutes the count rate dropped to zero and the beam was no longer visible indicating the absence of particle interception. A total of 20 replicate measurements were carried out.

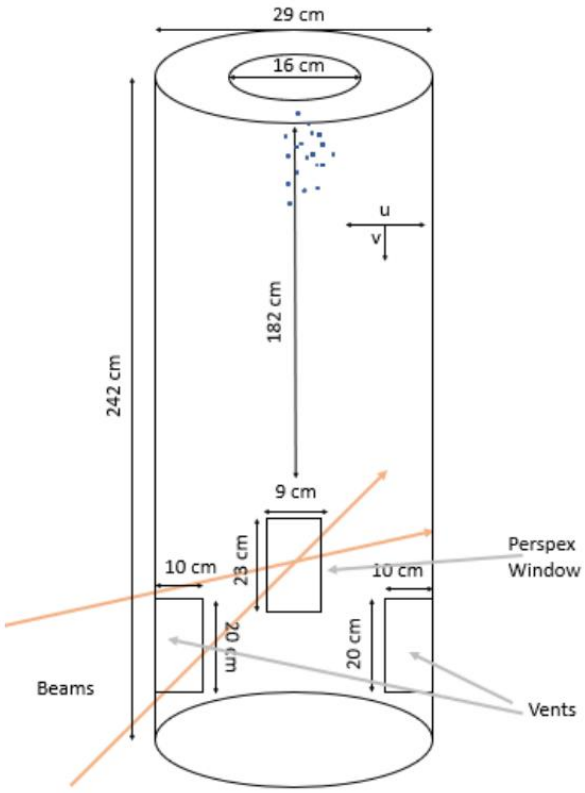

Figure 3.2- Diagram of the fall column apparatus used to measure the settling velocity of fibers. The vertical and horizontal settling velocities were both determined by manually dropping the fibers from the top of the column. Lasers entering through the Perspex window intersected directly in the middle of the column to obtain measurements. Note the diagram is not to scale.

3.3.6 Data Analysis

Daily microplastic counts were estimated by areal upscaling of the cut-out circle filter papers (19.63 cm^2) to determine the count for the entire filter paper (393.75 cm^2). The counts from the three circle cut-outs (triplicate sub-samples) were averaged and scaled up using a factor of 20.05 to attain the daily microplastic count for each entire filter paper. The dry deposition ($mp/m^2/day$) was calculated using the number of microplastics on the entire filter paper (mp), the

volume of air sampled (m^3), and the median deposition velocity determined in the lab study (m/day). It was assumed that fragments had the same settling velocity as fibers. For each sample type the average field blank was subtracted from the concentration; however, no microplastics were found in the laboratory open air blanks.

$$\textit{Dry Deposition (mp/m}^2\textit{/day) = Daily Microplastic Count (mp) ÷ Volume of Air Sampled (m}^3\textit{) x Deposition Velocity (m/day)}$$

The output of the LDA provided both a horizontal and vertical settling velocity component, the horizontal component was used for filtering purposes only. In total 48,707 (46.7%) of the measured settling velocities were screened and removed based on three criteria: (i) if the horizontal component was greater than the vertical component, the measurement was removed, (ii) if the vertical component indicated upward motion, the measurement was removed, and lastly (iii) upper and lower limits were applied to the data distribution to remove any outliers due to signal noise. The upper and lower limits were determined by calculating the average settling velocity plus two times the standard deviation. This resulted in one limit (0.977), which was used as both the upper (positive) and lower (negative) limit.

A wind rose was created to assess microplastic source areas for the five days of field sampling using the wind direction and wind speed (Figure 3.1) this was plotted using the WRPLOT software (Lakes Software, 2022). The wind speed and direction were obtained from the WISKI database (CLIM-MTO-CR26; Government of Ontario, 2022). Statistical analysis testing was performed for ambient air to determine if there was a statistical difference between the days sampled using a t-test as data were normally distributed. All statistical analysis was carried out in PAST 4.10 (PAST, 2020).

3.4 Results and Discussion

3.4.1 Ambient Air Concentrations

Microplastic particles were observed in all atmospheric samples for the five days studied (November 1–5, 2021); the total count across the five days was 1116.2 microplastics (blank corrected; 6.7 mp). The analysis of the circle cut-outs of the filter paper suggested that the area was representative of the entire filter paper, as the variability between the triplicate sub-samples was quite low (11.3%; Table 3.1). The daily microplastic concentrations in the current study ranged from 0.10–0.18 mp/m^3 (Day 5–Day 3); with an average of 0.13 mp/m^3 (Table 3.1). There was no statistical difference (t-test $p > 0.05$) in microplastic count between triplicate sub-samples (5 cm diameter) between the five days sampled.

A number of studies have investigated microplastic concentrations in outdoor ambient air. Dris et al. (2017) in Paris, France, found a concentration range of 0.3–1.5 fibers/m^3; Liu et al. (2019a) in Shanghai, China, found a range of 0–4.18 mp/m^3; and Choi et al. (2022) in Seoul, South Korea, found a range of 0.45–5.16 mp/m^3. Atmospheric microplastic pollution is likely related to population density; Shanghai and Seoul have the highest population (24.9 million and 9.9 million respectively; Major Agglomerations of the World, 2022) and the highest concentrations. While Paris has a lower population (2.2 million; Major Agglomerations of the World, 2022) and lower microplastic concentration. Port Perry has a much lower population (9,553) than the other studies, which explains the much lower air concentration.

Meteorological conditions such as wind speed and direction can affect the ambient air concentrations of microplastics as well as indicate the source region. Microplastics have been found in remote areas indicating that microplastics can travel through the atmosphere far from their source region (Allen et al., 2019; Bergmann et al., 2019); microplastics have been reported to be up to 95 km from potential source regions. The concentration of microplastics in the

environment depends on the source of microplastics for that region. During the sampling period, the wind was mainly from the southwest, i.e., blowing across most of Port Perry (Figure 3.1) and from the Greater Toronto Area (GTA; see Appendix Figure A3.3). However, due to the low concentration of microplastics in the ambient air during the study period it is assumed that Port Perry was the immediate and principal source of pollution.

Table 3.1- Microplastic count, ambient air concentrations (mp/m^3), and coefficient of variation (CV%) between triplicate sub-samples in high-volume air samples, Port Perry, Ontario (November 1–5 2021; see Appendix Table A3.3).

Day	Total Particle Count	Ambient Air Concentration (mp/m^3)	Coefficient of Variation (CV%)
Day 1	247.3	0.161	20.4
Day 2	173.8	0.118	0.0
Day 3	314.2	0.177	6.3
Day 4	153.7	0.104	12.5
Day 5	193.9	0.101	17.3
Average	216.6	0.132	11.3

The majority (97.6%) of microplastics in the ambient air samples were fibers, while the remaining 2.4% were fragments; no films were found (Table 3.2). Similarly, other studies found the majority of atmospheric microplastics to be fibers (Wright et al., 2020; Dris et al., 2017; Liu et al., 2019a). A main source of microplastics (especially fibers) is from textiles; microfibers can enter the atmosphere through the detachment during the wear, washing and drying of clothes (Pirc et al., 2016; De Falco et al., 2018). Another reason fibers tend to be dominant in the atmosphere is they remain suspended longer than other microplastics, whereas fragments tend to deposit closer to their source (Loppi et al., 2021).

Table 3.2- The percentage and count* of fibers and fragments in ambient air concentrations in Port Perry, Ontario (see Appendix Table A3.4).

	Fibers (%)	Fragments (%)	Fibers (count)	Fragments (count)
Total	97.6	2.4	1089.4	26.8

* All counts have been blank corrected and scaled up to represent the entire filter paper.

The observed microfibers generally had a length of <1 mm with an average of 1.13 mm and a median of 0.86 mm (Figure 3.3). The microplastics found in a bulk deposition collector in Port Perry were slightly longer with an average of 1.60 mm and median of 1.04 mm. Liu et al. (2019a) found the size of microplastics in Shanghai to range between 0–9.9 mm, with an average of 0.6 mm. While Dris et al. (2017) found microplastics to be <1.65 mm. The difference in microplastic length between the studies may be due to the different sources of microplastics and different identification techniques. Both Dris et al. (2017) and Liu et al. (2019a) identified microplastics using a stereomicroscope and did not further identify samples under fluorescence microscopy. The dominant colours in Port Perry were black and blue (Figure 3.4), similarly Liu et al. (2019a) found the most common colours in the atmosphere to be black and blue. Black and blue are the dominant colour of most clothes, e.g., denim jeans are worn by an estimated 46–56% of Canadians every day (Athey et al., 2020). Further, the average Canadian washes their denim more than necessary, after only 2 wears, therefore wearing the denim down, which creates easily detachable fibers that end up in the atmosphere (Athey et al., 2020).

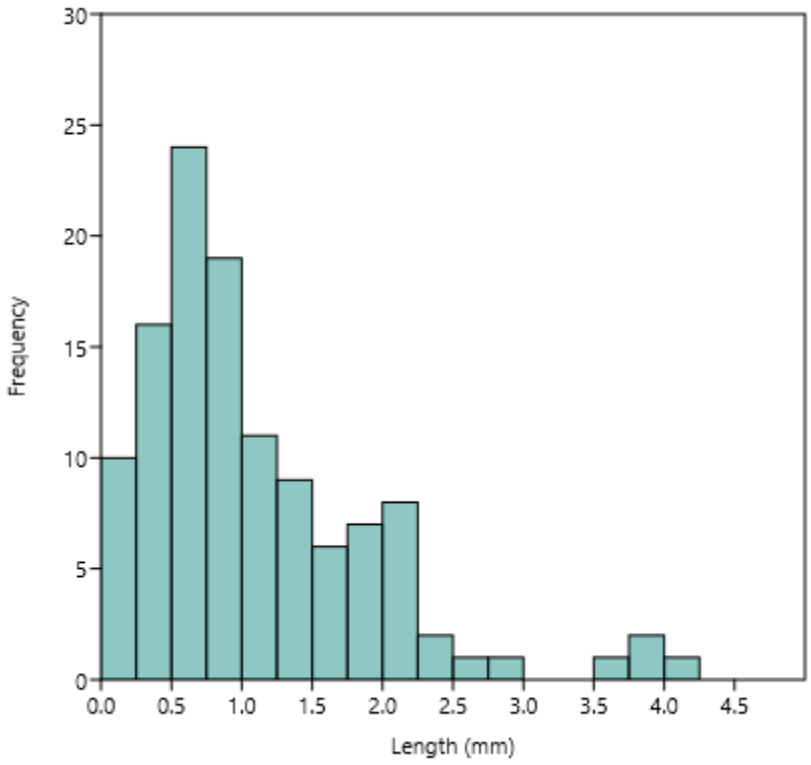

Figure 3.3- Histogram showing the length of microfibers found under brightfield microscopy in ambient air in Port Perry, Ontario, during November 1–5, 2021 (n=119).

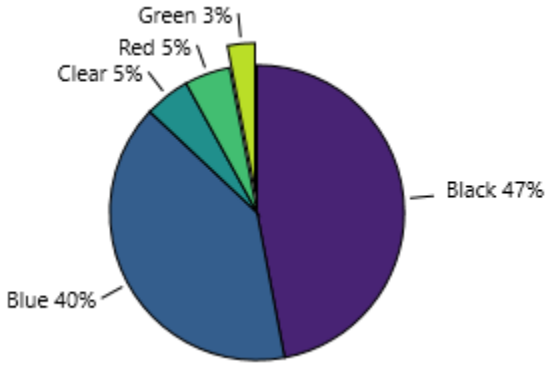

Figure 3.4- Pie chart showing the colours of all microplastics found under brightfield microscopy in ambient air in Port Perry, Ontario, during November 1–5, 2021.

3.4.2 Settling Velocity Analysis

The microfibers used for the settling velocity analysis were mainly <1 mm (81%), with a range of 0.03–2.99 mm, average 0.72 mm, and median 0.61 mm (Table 3.3; Figure 3.5). The kurtosis for the microfiber length (4.74) was quite high; transformations of the data showed that it was lognormal. Therefore, the median is a better measure of the central tendency than the average and the mode is the best representation of frequency. The wide distribution in fiber lengths was due to their cylindrical shape; fibers >1 mm in length passed through the aperture of the sieve vertically due to their small diameter or thickness (~0.07 mm; see Appendix Table A3.5). The size range of the prepared polyester microfibers (Table 3.3) were generally comparable to microfibers observed in the environment. The microplastics in the ambient air concentrations in Port Perry were a bit longer, with a median of 0.86 mm. While Wright et al. (2020) found similar lengths in atmospheric deposition in London, England, with the most abundant fiber lengths between 0.40–0.50 mm and an average of 0.91 ± 0.64 mm. Roblin et al. (2020) also found similar microfiber lengths in deposition across four remote sites in Ireland, i.e., the median length was 0.88 mm. Similarly, in Lake Scugog the median length across two bulk deposition stations during May–November 2021 was 0.79 mm (see Chapter 2).

The median settling velocity was 0.11 m/s, the average was 0.18 m/s, and the range was 0.0005–1.09 m/s (Table 3.3). The distribution of the settling velocities was skewed (Figure 3.5); the data distribution was left aligned, similar to the distribution of fiber lengths (Figure 3.5).

Table 3.3- Summary statistics for the length (mm) of the prepared polyester microfibers (n=1470); the kurtosis is quite high indicating a non-normal distribution (see Figure 3.3). Summary statistics for the measured settling velocity (m/s) of polyester microfibers (n=55,501) determined through the laboratory-based experiments are also given.

	Length (mm)	Settling Velocity (m/s)
Median	0.612	0.11
Average	0.720	0.18
Mode	0.605	0.04
Range	0.03–2.99	0.0005–1.09
5th percentile	0.263	0.02
95th percentile	1.578	0.58
Mean Absolute Deviation	0.302	0.14
Kurtosis	4.736	2.05
Skewness	1.866	1.56

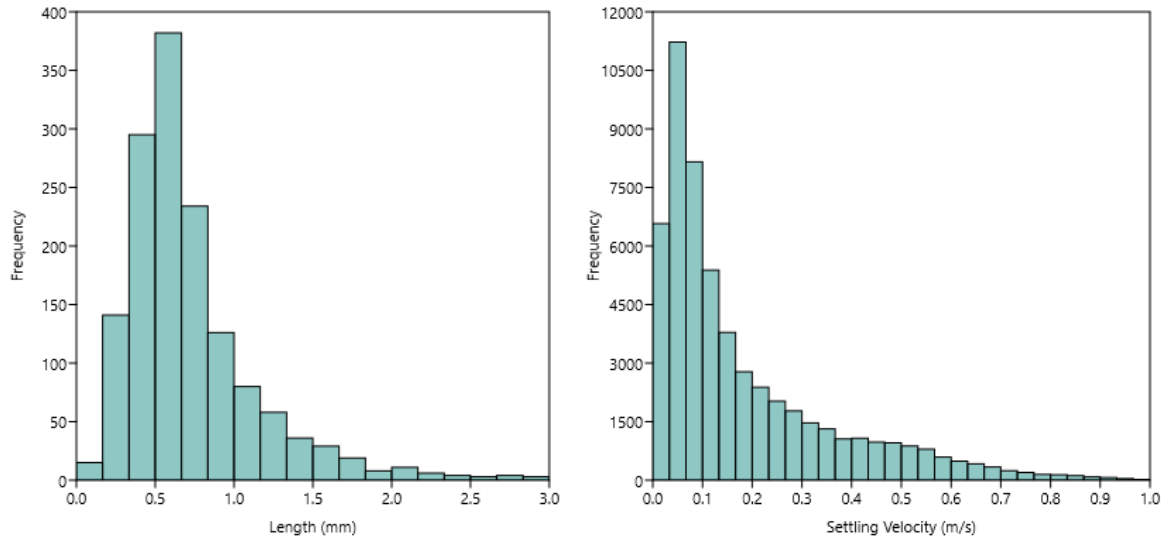

Figure 3.56- Histogram (left) showing the distribution of the length of the laboratory prepared polyester microfibers used to determine the settling velocity; measured across ten replicates (n=1470). Histogram (right) showing the distribution of the settling velocity (m/s) for polyester microfibers (n=55,501); the distribution is left skewed (lognormal).

Currently, there are no published laboratory-based atmospheric settling velocity measurements in the peer-reviewed literature. However, a few studies did carry out theoretical calculations for settling velocities of microplastics (Allen et al., 2019; Wright et al., 2020). Allen et al. (2019) estimated a settling velocity of 0.10 m/s based on non-fibrous <0.025 mm Saharan dust particles (with a density of approximately 2.65 g/cm^3; Rocha-Lima et al., 2018). In contrast, Wright et al. (2020) estimated a settling velocity for fibrous microplastics of 0.06 m/s based on an assumed fiber length of 0.4 mm and density of 1.184 g/cm^3.

The settling velocities determined by Wright et al. (2020) and Allen et al. (2019) are generally consistent with the mode and median settling velocities determined in the current study (Table 3.3); the slight differences may be related to their theoretical approach. However, the density and lengths of the microplastics were different between all studies; polyester was used in the current study, which has a density of 1.38 g/cm^3 (Wu et al., 2015), and the median fiber length was 0.61 mm (Table 3.3).

3.4.3 Dry Deposition

The average daily dry deposition across the five days was 1257 mp/m^2/day, with the range being 963–1678 mp/m^2/day (Table 3.4). Bulk deposition (wet and a portion of dry deposition) in Port Perry (approximately 2 km from the current study site) was recently determined for five consecutive months (June–October 2021); the average was 37 mp/m^2/day (see Appendix Table A3.6). The measurements for bulk deposition in Port Perry were comparable to other studies (Welsh et al., 2022) and reasonable based on the surrounding population density. In contrast, the calculated dry deposition for microplastics in Port Perry seemed very high; more than 30 times higher than the bulk deposition. There was very little rain during the sampling period (average rainfall of 0.34 mm; see Appendix Table A3.1), therefore

dry deposition would have been the dominant removal process; however, it still seems unreasonably large. One of the reasons for the uncertainty in the dry deposition rate may be due to the limited period of observation for ambient air sampled. Further the laboratory-based settling velocity does not account for potential influences that occur in the natural environment.

The laboratory settling velocity determined for this study is a valid estimate based on ideal conditions. However, in an environmental context it may need to be modified for meteorological conditions. Lo et al. (1999) found that with increasing wind speed the deposition velocity of a particle also increases by a factor of 2–5, depending on the size; the larger the particle the higher the increase. Chen et al. (2012) also found a positive correlation between dry deposition velocity and wind speed; a high wind speed tends to increase the friction velocity, which results in accelerating the transport of the particle. A limitation within this study is that wind speed was not taken into account; the current study solely focused on a laboratory-based determination of the settling velocity of polyester microfibers.

Table 3.4- Dry deposition of microplastics in Port Perry, Ontario, during November 1–5, 2021 based on the median laboratory estimated settling velocity (Table 3.3) and observed ambient air concentrations (Table 3.1).

	Dry Deposition ($mp/m^2/day$)
Day 1	1530
Day 2	1123
Day 3	1678
Day 4	991
Day 5	963
Average	1257

3.5 Conclusion

This study determined the dry deposition of microplastics in Port Perry, Ontario, using a laboratory measured settling velocity of polyester microfibers and measured ambient air concentrations. The dry deposition of microplastics in Port Perry was found to be 1257 mp/m^2/day. The average ambient air concentration was 0.132 mp/m^3 and the median settling velocity of polyester microfibers was 0.11 m/s. During the study period, the wind direction was south-west, i.e., blowing from downtown Port Perry, which was mostly likely the dominant source of microplastics. Results suggest that dry deposition is the dominant removal process for atmospheric microfibers compared with wet or bulk deposition; however, there were limitations within this study, and it is recommended that settling velocities under different meteorological conditions be further examined and used to estimate dry deposition.

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3.7 Appendix

Appendix A- Ambient Air Sampling and Settling Velocity Experiments

Table A3.1- The temperature and precipitation in Port Perry during the sample period (November 1–5, 2021), the data was obtained from Kawartha Conservation weather gauges (Kawartha Conservation, 2020).

Date	Temperature (°C)	Precipitation (mm)
Nov 1	5.56	0.17
Nov 2	2.96	0.00
Nov 3	1.41	0.12
Nov 4	1.25	0.96
Nov 5	1.93	0.47
Average	2.62	0.34

Table A3.2- The sampling period (start and end times, and exposure times) for ambient air concentrations and the volume (m^3) of air sampled in Port Perry.

Day	Start Date	Start Time	End Date	End Time	Exposure (hrs)	Volume of air (m^3)
Day 1	11/1/2021	12:30	11/2/2021	11:05	22.58	1534.77
Day 2	11/2/2021	11:05	11/3/2021	8:42	21.62	1469.08
Day 3	11/3/2021	8:42	11/4/2021	10:52	26.17	1778.30
Day 4	11/4/2021	10:52	11/5/2021	8:32	21.67	1472.48
Day 5	11/5/2021	8:32	11/6/2021	12:40	28.13	1911.95

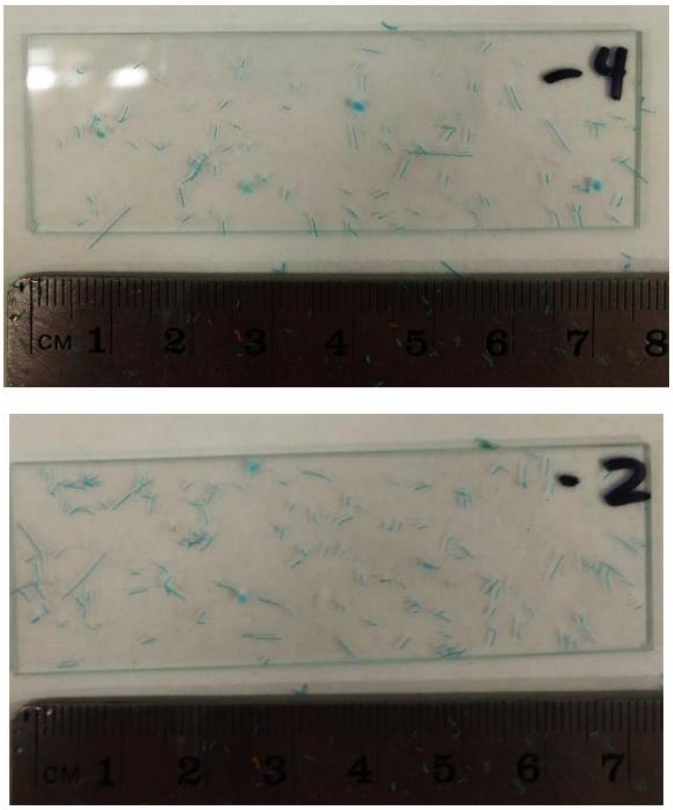

Figure A3.1- A sample of the images of slides analyzed to determine the length of the laboratory manufactured polyester microfibers used for the settling velocity measurements.

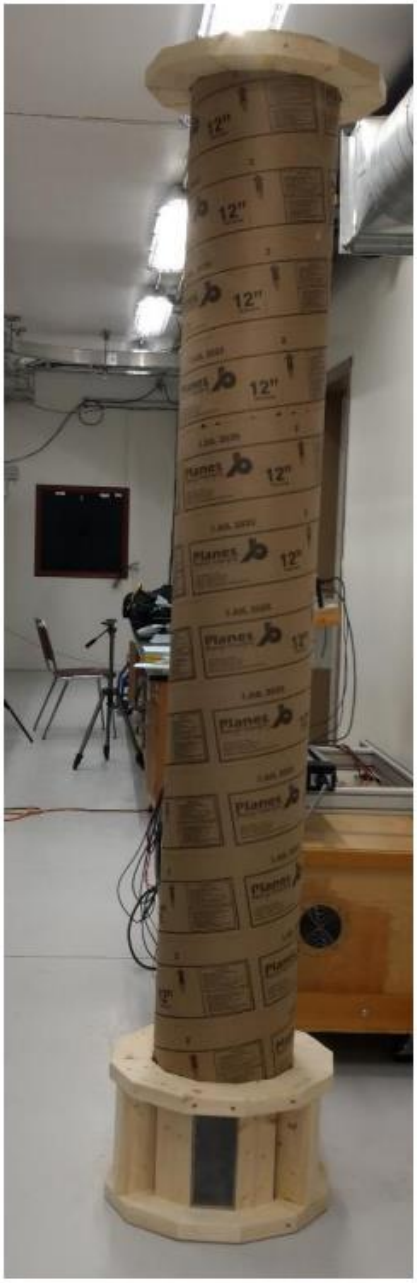

Figure A3.2- Photograph of the fall column constructed to determine the settling velocity of polyester microfibers, showing the base and top cover of the fall column (see Figure 3.2).

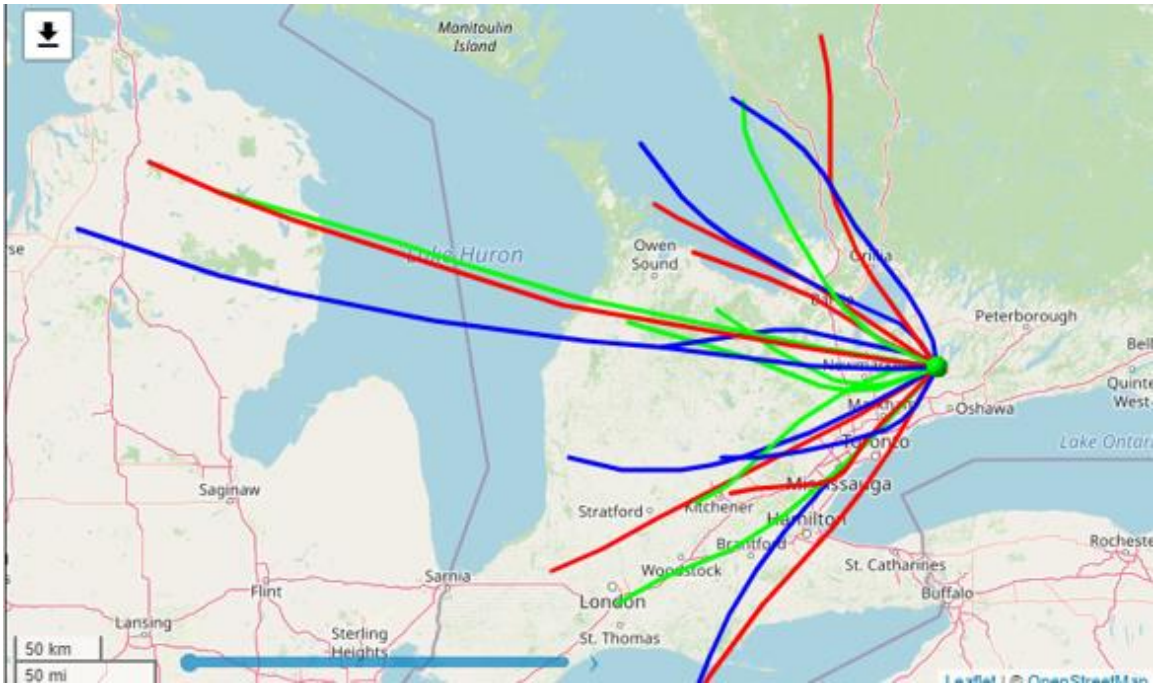

Figure A3.3- Back trajectory plot representing the wind direction during the five days ambient air was sampled in Port Perry, November 1–5 2021. Note: Colours represent individual trajectories.

Table A3.3- Count of microplastics under brightfield microscopy, blue-light fluorescence, and the total particle count* for ambient air sampled in Port Perry, Ontario (November 1–5 2021).

Day	Brightfield Microscopy Count	Blue-light Fluorescence Count	Total Particle Count
Day 1	9.7	2.7	247.3
Day 2	6.7	2.3	173.8
Day 3	12.3	3.7	314.2
Day 4	6.0	2.0	153.7
Day 5	6.7	3.3	193.9
Average	8.3	2.8	216.6

*The total particle count (blank corrected) includes the entire filter paper count, which was scaled-up from the three circle cut-outs, while the brightfield microscopy and blue-light fluorescence count is the average of the three circle cut-outs.

Table A3.4- The percentage and count* of fibers and fragments in ambient air concentrations in Port Perry, Ontario, found under brightfield microscopy and blue-light fluorescence.

	Fibers (%)	Fragments (%)	Fibers (count)	Fragments (count)
Brightfield Microscopy	98.4	1.6	815.4	13.4
Blue-light Fluorescence	95.3	4.7	274.0	13.4

* All counts have been blank corrected and scaled up to represent the entire filter paper.

Table A3.5- The diameter (mm) of the polyester fibers (n = 1470) used to determine the settling velocity, measured manually through Image J open- source software.

Variable	Diameter (mm)
Average	0.076
Median	0.070
Mode	0.070

Table A3.6- The count, concentration, and daily flux over five months (June–October 2021) for bulk deposition in Port Perry, Ontario.

Month	Microplastic Count	Volume (L)	Concentration (mp/L)	Daily Flux (mp/m/day)
Jun	31	4.8	5.8	29.7
Jul	49	4.1	11.3	47.7
Aug	27	1.6	16.4	26.7
Sep	41	5.1	7.5	40.1
Oct	22	2.2	9.5	21.2
Overall Average	34	3.6	10.1	37.3

Appendix B- Pilot Testing for the LDA

Initial pilot testing was performed to determine the settings (gain, validation and sensitivity) for the LDA. The two main settings that need to be adjusted are gain and validation. Gain boosts the signal/noise ratio for the LDA to receive and validation averages the obtained measurements to avoid collecting "noise" data. A poor validation rate can record many particles, however the results from this may not be accurate. On the other hand, a high validation rate may be very accurate, but there are not many counts. A validation rate had to be determined that had a high count while still being accurate. There are two sensitivity rates, vertical and horizontal, when the sensitivity threshold is exceeded the particle is then measurable. This optimization was carried out by releasing multiple replicates into the fall column to determine which settings would work best for polyester. A total of 18 different combinations of settings were adjusted, at each different combination three replicates were performed. After performing these pilot tests, the following settings were determined for polyester: sensitivity: 1000_1200, gain: 14, validation: 4.

Calibration tests were performed to confirm the calibration of the LDA, using 0.22 mm glass beads (see Appendix Table B3.1). These tests were performed to determine if the air was still in the room. The tests were also done to confirm that the LDA was measuring settling velocities correctly. The measurements from the calibration tests were confirmed through published literature (Kliafas, 1989). Glass beads needed to be used for this as there is a known settling velocity for them. The tests consisted of five replicates with each replicate having a particle count of 2000. The replicates were dropped by hand down the fall column. The size of the glass beads was confirmed using a Horiba LA-950V2 particle size analyzer, three replicates were performed, and each replicate was tested three times (Table B3.1).

Table B3.1- The medians of nine replicates of the glass bead reference material for size confirmation. The size of the glass beads was measured using a Horiba LA-950V2 particle size analyzer.

Replicate	Median (mm)
Rep 1	0.2177
Rep 2	0.2176
Rep 3	0.2175
Rep 4	0.2197
Rep 5	0.2197
Rep 6	0.2197
Rep 7	0.2199
Rep 8	0.2198
Rep 9	0.2200
Average	0.2191

4.0 Conclusion

4.1 General Conclusion

It is well established that biogeochemical and nutrient budgets can be used to effectively integrate all inputs and outputs at the catchment scale, however this is still quite a novel concept for the study of the microplastics cycle (Hoellein & Rochman, 2021; Horton & Dixon, 2018). By studying the entire catchment, the terrestrial and atmospheric transport of microplastics is also integrated in this analysis and the overall fate of microplastics can be better understood. Freshwater ecosystems are considered conduits for microplastics as they act as transporters, receivers, and sinks for microplastics (Enamul Kabir et al., 2021; Horton & Dixon, 2018; Hoellein & Rochman, 2021; Xia et al., 2020). There has been growing recognition of the importance of the atmosphere as a pathway of microplastics into remote environments (Allen et al., 2019; Bergmann et al., 2019). In general studies have quantified microplastics in wet-only or bulk deposition (Szewc et al., 2021; Klein & Fischer, 2019); however, to gain a more holistic understanding of the deposition and transport of atmospheric microplastics, dry deposition needs to be studied. The overall objective of this thesis was to assess the abundance and distribution of microplastics within Lake Scugog catchment, Ontario. This was achieved through two secondary objectives to (1) determine the abundance of microplastics in Lake Scugog, and to develop a microplastic particle (input–output) budget for the lake catchment, and (2) determine the dry deposition of microplastics in Port Perry, Ontario, based on ambient air concentrations of microplastics and a laboratory measured settling velocity for polyester fibers. In addition, a preliminary assessment of smaller particles (in the range of 20–100 µm) was carried out, which is typically not included in visual analysis.

Chapter 2 focused on developing a microplastic particle budget for Lake Scugog catchment. This was determined by quantifying the inputs (precipitation and inflows) as well as the outputs (sedimentation and the lake outflow) of microplastics. Stream inflows, the lake outflow, and lake water samples were collected for six months (May–October), and precipitation was collected for five months (June–October), while sediment samples were collected once in October. Microplastics were extracted from each sample and identified under brightfield microscopy and blue-light fluorescence to identify particles <20 µm. Raman spectroscopy was used to determine plastic polymer composition. The total number of microplastics in Lake Scugog was 2.62 $x10^{11}$ mp with a residence time of one year. The total daily input of microplastics into the lake was 2491 $x10^{6}$ mp/day, while the output was 1761 $x10^{6}$ mp/day, resulting in 29% being retained in the lake. The largest daily microplastic flux into Lake Scugog was precipitation (19.51 mp/m^2/day), which was nearly four times greater than the main output flux through the lake outflow (3.05 mp/m^2/day). This suggests that atmospheric deposition is the main pathway of microplastics in background regions. Further, it may play a dominant role in other systems, but has had limited attention as a source pathway in aquatic microplastics research. Atmospheric deposition is composed of wet and dry inputs, but only bulk (wet plus a fraction of dry) deposition was measured in the particle budget, therefore, to gain a more holistic understanding of atmospheric microplastics the dry deposition should be examined.

Chapter 3 examined the dry deposition of microplastics in Port Perry, Ontario, Canada, based on field measured ambient air concentrations of microplastics and a laboratory settling velocity for polyester microfibers. Ambient air concentrations were collected for five consecutive days in November 2021 using an active air sampler, and settling velocity was measured using a Laser Doppler Anemometer and a drop column. Microplastics were analyzed

under brightfield microscopy and blue-light fluorescence. The dry deposition was estimated to be 1257 mp/m^2/day based on an observed average ambient air concentration of 0.132 mp/m^3 and a settling velocity of 0.11 m/s. The dry deposition was considered unreliable as there was a large difference between observed bulk deposition in the same area. One potential reason for the high dry deposition may be the settling velocity, as it was determined in a laboratory setting rather than the natural environment where meteorological conditions influence (potentially reduce) the settling velocity of particles.

4.2 Contributions to Research

Chapter 2 is one of the first studies in Canada (if not globally) to quantify the microplastic particle budget in a freshwater lake catchment. This study is the first to estimate the abundance of microplastics in stream inflows, precipitation, bio-monitors, the lake outflow, sediment, and lake water in a freshwater lake during the ice-free period. Many studies in Canada have focused on the Great Lakes (Yang et al., 2022; Ballent et al., 2016; Grbić et al., 2020; Eriksen et al., 2013; Corcoran et al., 2015; Dean et al., 2018), and the few that have focused on smaller lakes, generally only quantify microplastics in surface waters and sediment (e.g., Felismino et al., 2021; Anderson et al., 2017) and have excluded atmospheric and stream inputs.

Chapter 3 provides a more holistic understanding of atmospheric microplastics. Many studies have focused on bulk or wet only deposition (Szewc et al., 2021; Klein & Fischer, 2019), few if any studies have focused on the dry deposition of microplastics which is necessary to gain a more holistic understanding of atmospheric microplastics. This is also the first study to determine the atmospheric settling velocity of polyester microfibers, all other studies to date have based microplastic settling velocities on theoretical calculations (Wright et al., 2020; Allen et al., 2019).

4.3 Recommendations for Future Work

Comparison between studies is difficult as methodologies and units are not standardized. It is recommended that harmonized methodologies be used during both sampling and laboratory techniques when studying microplastics (ECCC, 2020). Further, units used to present microplastic concentrations should be standardized. Future studies should determine an appropriate sample volume for water as one-liter samples can be highly variable for low levels of pollution and the methodologies for sampling large volumes have the risk of losing microplastics. Preliminary tests regarding sample volumes were performed in the current study, 1 L and 500 L samples were taken, and both had limitations; it is suggested that a 10 L grab sample be performed for water samples (see Appendix Table B2.1).

Although this study did fill a knowledge gap regarding the microplastic particle budget in a lake, there were limitations. The lake catchment was only sampled during the ice-free period (May–October), in future studies, samples should be taken over the entire year. Furthermore, microplastic concentrations in soil in the terrestrial catchment should be examined to determine the amount of microplastics retained in the soil from atmospheric deposition and the amount of microplastics that enter the lake from the terrestrial catchment. Another limitation in this study were the effects of meteorological conditions on the settling velocity of microplastics, future studies should examine this. Ultimately, the identification of the microplastic budget in Lake Scugog proved that microplastics should be examined at a catchment scale to understand their fate in multiple environmental compartments. Through determining the microplastic budget better management strategies can be implemented to assist in controlling microplastic pollution. The atmosphere was a major pathway for microplastics into Lake Scugog catchment through both wet and dry deposition and should be a key component when studying microplastics in the environment.

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