STORM-DRIVEN TRANSPORT AND DEPOSITION OF ATMOSPHERIC MICROPLASTICS IN REMOTE NEWFOUNDLAND, CANADA

by

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ABSTRACT

Microplastic pollution is an increasing global concern due to its pervasiveness in all types of environments around the world, and the potential risk it can pose to all living organisms, including humans. In recent years, the study of how microplastic particles travel across the atmosphere has attracted increasing attention, and preliminary results have shown that rain and wind can transport and deposit large quantities of microplastics in remote regions. However, how intense meteorological events such as hurricanes can influence microplastic pollution is still unknown.

In this MSc thesis, I present two studies that investigate the dynamics of atmospheric microplastic transport and deposition in Newfoundland, Canada, a relatively remote region with low population density and susceptible to intense storms. In the first study, the passage of Hurricane Larry over Newfoundland in 2021 was used as a natural laboratory to quantify storm-driven transport and deposition of microplastics. I collected atmospheric samples before, during, and after the storm to quantify microplastic deposition. I used chemical analysis of polymer types and back-trajectory modelling to determine that the majority of microplastics collected were likely sourced from the ocean. To quantify whether tropical cyclones and nor'easters can have a dominant role in microplastic pollution in the region, I collected remote lake bottom sediment samples across Newfoundland. Assuming that the lakes are fed by rain or groundwater and human impact is negligible, I hypothesized that the primary source of microplastics at this location is the atmosphere. The results show microplastics were present in all samples and had a similar size distribution to the particles deposited during Hurricane Larry, identifying the possible link of microplastic deposition to storm events. This research provides quantitative data for the first time on how hurricanes may influence microplastic pollution in remote regions of North America. Additionally, by quantifying the level of plastic pollution in many lakes across the province, I provided a baseline dataset that can support future works aimed at investigating fresh-water environments, in Newfoundland and elsewhere. This research contributes to a comprehensive understanding of the fate and impact of atmospheric microplastics in remote environments.

LIST OF ABBREVIATIONS USED

MP	Microplastic
PE	Polyethylene
PVC	Polyvinyl chloride
PP	Polypropylene
PS	Polystyrene
PET	Polyethylene terephthalate
LDPE	Low-density polyethylene
HDPE	High-density polyethylene
TWPs	Tire wear particles
HYSPLIT	Hybrid single-particle lagrangian integrated trajectory
PMMA	Polymethyl methacrylate
PVA	Polyethylene vinyl acetate
PES	Polyester
PA	Polyamide
PLS	Polysulfone
PU	Polyurethane
w.w.	Wet weight
ATV	All-terrain vehicle

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Chapter 1 INTRODUCTION

1.1 Statement of problem

Since 1950, plastic products have steadily become more and more prevalent in our daily lives with plastic production increasing every year (Figure 1.1), reaching an estimated 390.7 million metric tonnes in 2021 (Statista, 2023). Of all the plastic ever produced globally, studies report that less than 10% is recycled (Geyer et al., 2017). The remaining plastic ends up in landfills or is littered in oceans, rivers, and across landscapes (Geyer et al., 2017). Plastics are highly resistant to degradation meaning they can persist in nature, including aqueous environments, for hundreds of years (Liu et al., 2019). Though they do not degrade completely, plastics can easily be broken down into smaller pieces called microplastics (MPs), particles 1 – 5000 μm in size (Thompson et al., 2004; Arthur et al., 2009). These tiny particles are more easily ingested, entrained, and transported around the globe (Evangeliou et al., 2020).

MP pollution has been detected in all types of environments and thus is a growing environmental and public health concern (Thompson et al., 2004; Browne et al., 2011; Van Cauwenberghe et al., 2013; Cózar et al., 2014; Dris et al., 2015; Peng et al., 2018; Allen et al., 2019; Bergmann et al., 2019; Kiu et al., 2019; Zhang et al., 2019; Napper et al., 2020). MP particles can cause significant harm to wildlife through ingestion or inhalation, introducing contaminants into an organism and interfering with biological processes, as well as overall ecosystem health by transporting toxins including heavy metals and viruses, or leaching toxic chemicals (Cole et al., 2013; Karbalaei et al., 2019; Walker et al., 2022; Marn et al., 2020). Similarly, MPs also pose a risk to humans: when

inhaled or ingested MPs can transport toxins into the human body and, at a cellular level, can affect cellular viability and oxidative stress (Schirinzi et al., 2017). Studies have even shown the risk to climate processes, as MPs in the atmosphere can affect radiative forcing and cloud formation (Revell et al., 2021; Aeschlimann et al., 2022).

Plastics, and therefore MPs, are generally sourced in continental regions with high levels of anthropogenic activity and then are transported, primarily by rivers, to oceans (Schmidt et al., 2017; Lebreton et al., 2017; Meijer et al., 2021). Therefore, most MP research during the last decade has focused on describing the abundance, composition, suspected sources, and impacts of MPs in such environments (Allen et al., 2022a; Andrady, 2011; Cole et al., 2011; Dubaish & Liebezeit, 2013; Nyberg et al., 2023). In the past, fate and transport processes in the atmosphere were not as thoroughly investigated.

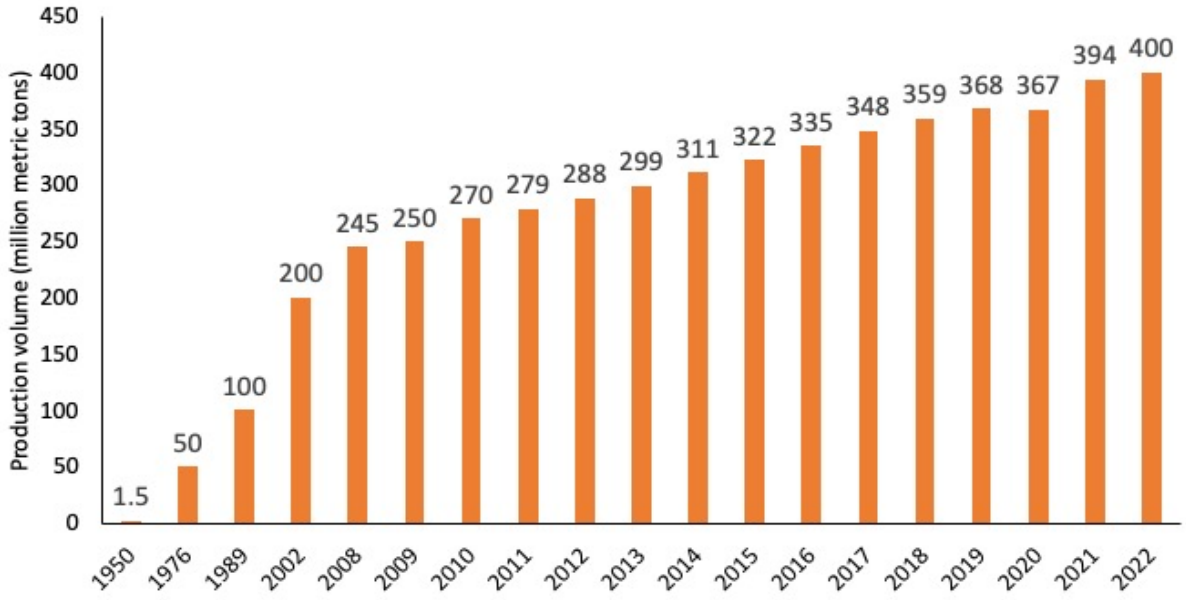


Figure 1.1: Annual global plastic production in million metric tons from 1950-2022 (Statista, 2023).

The atmosphere has the potential to move large quantities of MPs for further distances over a shorter time period than via aquatic transport. Studies have also shown that the atmosphere deposits MPs to remote areas that have no direct source and would have previously been considered pristine (Brahney et al., 2020). This may be especially true during major meteorological events, such as hurricanes, that are characterised by sustained high winds and heavy rainfall. Indeed, precipitation has been shown to increase atmospheric deposition of MPs (Dris et al., 2016), while high winds mean larger particles can be entrained and transported than in fair-weather conditions (Allen et al., 2019; Szewc et al., 2021). Hurricanes form over the ocean and travel long distances, potentially entraining MPs from polluted surface waters or coastal urban centres and depositing them in remote locations that would otherwise have very little MP input (Figure 1.2).

In this MSc project, I used newly acquired data from Newfoundland, Canada to evaluate whether tropical and extra-tropical cyclones influence the transport of MP to remote locations in the North Atlantic and to answer the following key questions: What is the impact of major atmospheric events on the transport and deposition of MP in Atlantic Canada and what are the sources?

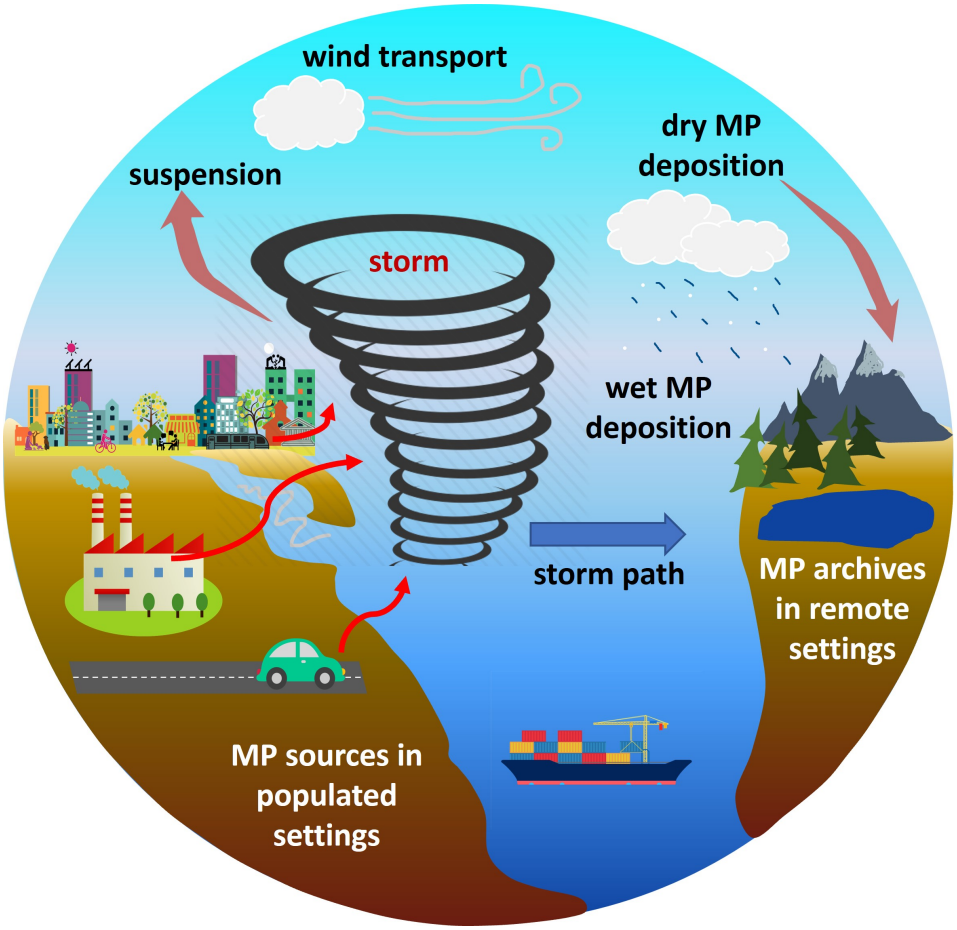


Figure 1.2: Conceptual model for MP transport from source in urban or marine environments to deposition in remote areas via the atmosphere

1.2 Objectives

To answer these questions, I conducted two field work campaigns in Newfoundland, Canada, a province with low population density, surrounded by Northwest Atlantic Ocean waters, and impacted by severe storms. I collected and analyzed samples of atmospheric MP deposition before, during, and after the passage of Hurricane Larry, a category 1 hurricane, as it made landfall in September 2021. In addition, I collected and analyzed sediment samples from various remote endorheic lake beds across Newfoundland, considered to be sites with no local inputs of MP and where MPs

accumulate primarily via atmospheric fallout. The expected outcome of this work was to quantify MP fallout during a storm and define possible sources.

It is important to get a better understanding of how MPs are transported through the atmosphere, as well as be able to quantify the amount of MPs deposited during a hurricane, in order to know which areas are more at risk of receiving large MP deposits. Improving our understanding of how MPs move around the world helps policy makers and remediators know where to focus protection and mitigation efforts. Though this is only a snapshot of global atmospheric MP transport and deposition during storms, quantifying the amount of plastic in one single event provides awareness and encourages changes to reduce the amount of plastic being produced and managing how much gets released into the environment.

CHAPTER 2 BACKGROUND

2.1 Microplastics: composition, sources, and distribution in the North Atlantic

Microplastics (MPs) are defined as plastic particles ranging from 1 – 5000 μm in size and may vary in shape, colour, and composition (Thompson et al., 2004; Arthur et al., 2009). The characteristics of a MP particle are largely dependent on the intended purpose of the original plastic. Primary MPs, such as microbeads, are intentionally manufactured for use in products such as beauty products (Boucher & Friot, 2017). Most MPs are secondary, meaning they were created when larger plastics fragmented into smaller pieces via photodegradation, physical abrasion, biodegradation, or hydrolysis (Zhang et al., 2020; Evangeliou et al., 2020). The most common MP shapes are fragments and fibres, as well as films, foams, and beads. Plastics come in many different colours, though MPs may lose much of their original colouring due to weathering (Pickett, 2018).

The best way to identify MP particles is by identifying their composition. There are many different plastic polymers manufactured, with the most abundant types including polyethylene (PE), polyvinyl chloride (PVC), polypropylene (PP), polystyrene (PS), and polyethylene terephthalate (PET) (Nerland et al., 2014). Chemical additives such as phthalates or plasticizers may also be added to improve certain qualities of the plastic like durability, malleability, and heat resistance (Nerland et al., 2014). There are also variations of polymers that affect the intended usage of a plastic. For example, PE has a low-density (LDPE) variety that is light and flexible making it an ideal material for plastic bags, and a high-density (HDPE) variety that is harder and stronger (Yashoda, 2016).

MPs are sourced in areas with anthropogenic activity, mostly urban centres, agricultural areas, or industrial areas. Major sources for MPs include city dust, factories, wastewater treatment plants, agricultural areas, and busy highways (Browne et al., 2011; Boucher & Friot, 2017). Polymer type can sometimes be indicative of a particular MP's source. For example, nylon fibres often come from fishing nets, while tire wear particles (TWPs) are released by car tires on roadways. However, most polymer types have several different applications and are used in many industries making it hard to pinpoint an exact source for any particular MP particle. Additionally, MPs that were transported away from their initial source and deposited in the environment may be remobilized and further transported meaning places that are generally considered sinks for MPs, namely terrestrial soils and the ocean, may also act as sources (Brahney et al., 2021). In these cases, polymer type indicates the original source of plastic but is not a reliable indicator of where the MPs first entered the environment.

Though there are marine sources of MP such as offshore petroleum operations, fishing (both commercial and recreational), and shipping, the majority of MPs are sourced on land (Geyer et al., 2017) and a large proportion of land-sourced MPs end up in the ocean. The North Atlantic is bordered by many densely populated urban areas of the Eastern United States and Western Europe. Rivers, coastal runoff, and the atmosphere are all potential transport pathways for bringing large amounts of MPs from these centres to the ocean (Geyer et al., 2017).

The distribution of MPs in the North Atlantic is not homogenous, with a large proportion of MPs in surface waters concentrating in sub-tropical ocean gyres (Eriksen et al., 2014). The Sargasso Sea in the North Atlantic is the site of a garbage patch, with previous estimates of up to 580,000 MP particles/km^2 in surface waters (Law et al., 2010). Since so much MP is transported to the ocean, it is generally thought of as a sink for MPs. However, recent studies suggest that plastic particles in surface waters may be resuspended into the atmosphere and further transported (Allen et al., 2020). This means instead of following a source to sink ideology, MPs may follow a cyclical path of being entrained into the atmosphere from ocean or land, transported and deposited, then resuspended and further transported allowing them to spread across the entire globe.

2.2 Microplastic transport mechanisms

2.2.1 River transport

Since all plastic is manufactured on land and, excluding maritime or fishing uses, all consumer uses for plastic take place on land, rivers are a major transport mechanism for carrying MPs from the terrestrial environment to the marine environment. It is estimated that rivers may export up to 0.5 – 4 million tons of plastic globally to the ocean each year (Schmidt et al., 2017; Lebreton et al., 2017; Meijer et al., 2021; Strokal et al., 2023). Though rivers have been generally considered as conduits for MPs to estuaries, coastal areas, and the ocean, they are now being recognized as sinks as well, retaining a portion of the MPs that enter them (Castaneda et al., 2014). As with sediments, the distance and speed of MP transport is dependent on river flow and particle density (He et al., 2021). Higher energy rivers will entrain more MPs and larger MPs than low energy rivers. On

average, MPs are lower density than sediments (Wypych, 2022) and therefore are more buoyant. However, there are higher density polymers that are more prone to sinking. Additionally, buoyancy is also dependent on particle shape and biofilm accumulation (Liu et al., 2022).

The primary source of MPs in rivers is mismanaged waste, both municipal and agricultural, with other important sources including car tires, laundry fibres, and personal care products (van Wijnen et al., 2019; Weinstein et al., 2016). However, types of sources for plastic pollution in rivers varies by region. River basins in Europe, North America, and Oceania are dominated by MP pollution from point sources (i.e., sewage systems) while river basins in Asia, Africa, and South America are dominated by MP pollution from diffuse sources (i.e. mismanaged solid waste; Strokal et al., 2023). In both scenarios, MP contamination in rivers is largely due to mishandled treatment of waste, whether that be solid or liquid.

The amount of MP transported by rivers can be difficult to quantify since it varies seasonally and is affected by events such as floods (Strokal et al., 2023). Snowmelt, heavy rains, or monsoon season all increase river flow and therefore the amount of MPs transported via rivers (Lebreton et al., 2017). Increased runoff also promotes the deposition of terrestrial MPs into waterways, especially in urban or agricultural centres (Moore et al., 2011). Erosion can also contribute to MP load in rivers, as increased erosion, such as in times of heavy rainfall, can release MPs stored in the soils of riverbanks into the water (Moses et al., 2023).

2.2.2 Oceanic transport

MPs have been detected in all marine environments, from shorelines to surface waters of gyres to the bottom of deep-sea trenches (Van Cauwenberghe et al., 2013; Vianello et al., 2013; Cózar et al., 2014; Turra et al., 2014; Peng et al., 2018). MPs in the ocean may be sourced from marine activities, however as previously stated, the vast majority originates on land (Jambeck et al., 2015; Geyer et al., 2017). This includes the release of MPs from coastal activities, inputs from rivers, and breakdown of macroplastics sourced from land. Suspended MPs in the ocean may be transported to remote areas via surface or deep-water currents (Li et al., 2020; Kane et al., 2020) or accumulate in central ocean gyres creating garbage patches. The most well-known of these is the Great Pacific Garbage Patch, located in the northeastern Pacific Ocean between California and Hawaii; however, there are also significant garbage patches in the northwestern Pacific, the Indian Ocean, the south Pacific, and both in the north and south Atlantic (Eriksen et al., 2014; Figure 2.1). It is estimated that due to gyre circulation the Pacific Ocean contains 33 – 35% of the global ocean plastic load (Cózar et al., 2014); however, gyres are the most well-sampled regions of the ocean while other areas are significantly underrepresented, introducing potential for error in that estimate.

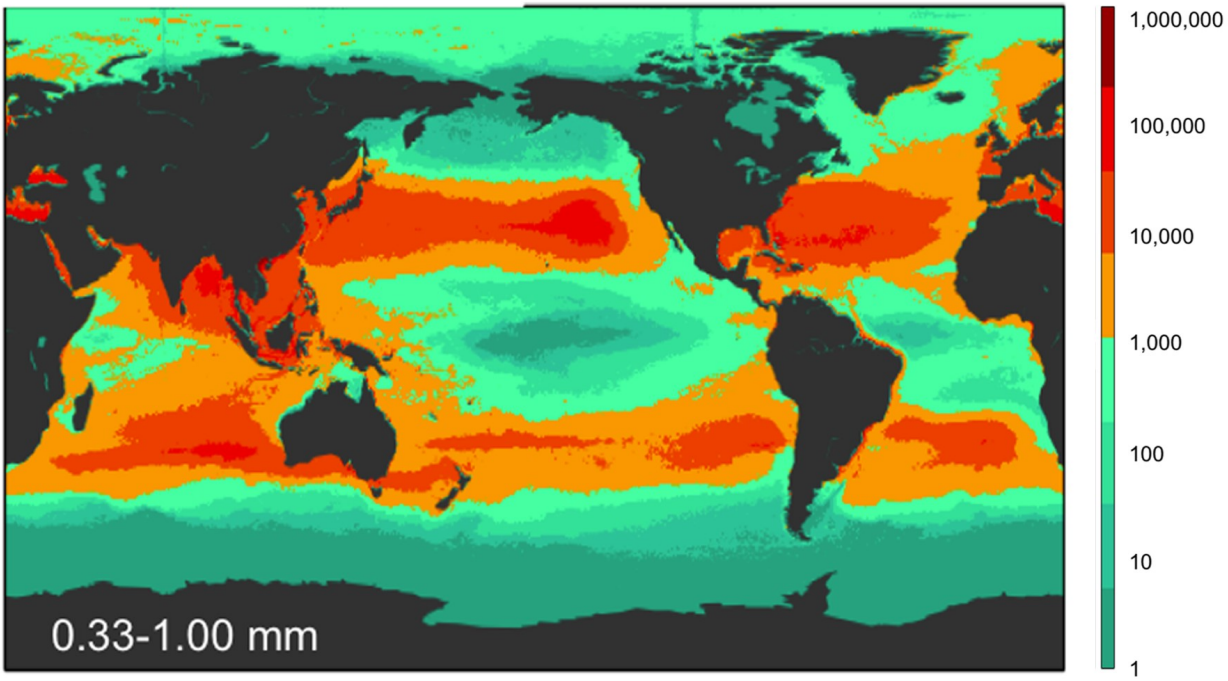


Figure 2.1: Model estimate of global MP density for particles between 0.33 – 1 mm in ocean surface waters showing concentration of MPs in subtropical gyres (from Eriksen et al., 2014).

Not all plastic that enters the ocean remains at the surface. Similarly to river transport, there is export of MPs to the deep ocean and seafloor. Previous studies show that only approximately 1% of plastics in the ocean are in the surface waters (Koelmans et al., 2017) with the highest concentration of plastic particles occurring between 200 – 600 m depth (Choy et al., 2019). The likelihood of MPs sinking below the ocean surface layer is largely dependent on density, and therefore polymer type. Over half of plastics produced (by weight) have a density lower than average surface seawater (1027 kg m^{-3}; Kukulka et al., 2012), meaning they would be assumed to stay in the surface waters. However. the export of MPs to depth is also influenced by biological interactions, as MPs may be ingested by zooplankton or filter feeding organisms and either transported to deeper waters at the organism's death or potentially bioaccumulated through the food web (Miller et al., 2020). Additionally, biofilms may form on the plastic's surface which can

increase the density and more readily transport the plastic particle to deeper waters (Liu et al., 2022). Oceanic processes such as vertical mixing can also play a role in moving MPs from surface waters to depth (Enders et al., 2015).

MPs at depth can be brought back to the surface via upwelling while deep-water currents can transport these particles long distances or can create zones of accumulation similar to the garbage patches found in surface gyres (Kane et al., 2020). At the surface, MPs are subject to wind forcing and geostrophic circulation (Li et al., 2020). Wind waves and bubble injection support the suspension of MPs in surface waters into the atmosphere (Allen et al., 2020). Alternatively, MPs may be stuck within the water column, rising and falling due to turbulent mixing, and never make it to the sediments or back to the surface waters (Kanhai et al., 2018; Li et al., 2020).

2.2.3 Atmospheric transport

The atmosphere is another crucial part of the plastic cycle, providing a transport and deposition mechanism to remote areas, both terrestrial and aquatic, that would otherwise receive very little MP pollution (Allen et al., 2019; Zhang et al., 2019; Brahney et al., 2020). MPs can be entrained into the atmosphere through mechanical processes directly from point sources or from areas with high concentrations of MP such as ocean gyres (Brahney et al., 2021). MPs can also be directly emitted into the atmosphere, either as dust created during agricultural processes or by the turbulence during vehicle tire movement (Brahney et al., 2021). Very little is known about transport pathways once MPs are suspended in the atmosphere. Previous research has shown that atmospheric transport is generally more efficient than oceanic or terrestrial transport as particles in the

atmosphere can travel hemispheric distances in a matter of weeks rather than years (Evangeliou et al., 2020). Due to the physical properties of plastic, MPs can travel much further through the atmosphere than mineral dust particles. That means I unfortunately cannot use research on particles such as Saharan dust as an analog for atmospheric MP transport.

Studies have shown atmospheric MP deposition is prevalent in urban areas and rural areas alike (Dris et al., 2015; Allen et al., 2019). MPs may be deposited from the atmosphere by dry or wet deposition, meaning MP fallout can happen in all weather conditions (Allen et al., 2019). Atmospheric transport and deposition of MPs is influenced by weather events, with evidence of high winds and precipitation increasing the abundance and/or size of MP fallout (Abbasi et al., 2022; Li et al., 2022; Ryan et al., 2023).

2.3 Study area

Newfoundland is an island on Canada's northeast coast (Figure 2.2). It has a relatively low population density (average 1.4 person/km^2; Statistics Canada, 2021), with the majority of the population living in a few urban centres. Most of the island is sparsely populated with a large portion of the land mass having limited or no access by road or river. The commercial fishing industry in and around the province has remained limited since 1992 due to the cod moratorium, meaning there is less fishing-related plastic input than other provinces in Atlantic Canada (Liboiron et al., 2020). Other industries on the island include mining, pulp and paper production, and petroleum (Government of

Newfoundland and Labrador, 2023); however, these are relatively small-scale operations. With most of the population and industry being concentrated in the few cities, most of the island is not exposed to direct sources of MP pollution.

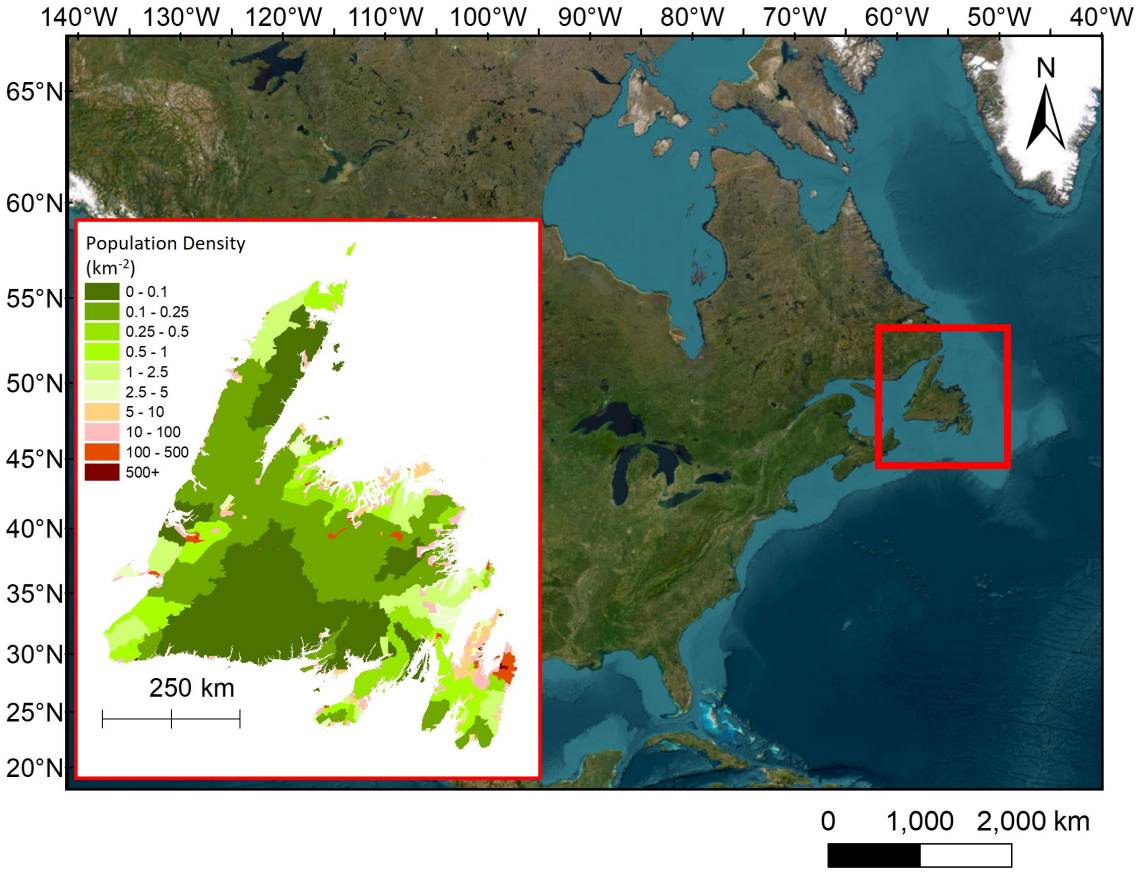


Figure 2.2: Map of North America with location of the island of Newfoundland indicated in the red box and population density (Statistics Canada, 2021) map of the island of Newfoundland (inset).

Newfoundland is a unique study area for atmospheric MPs due to its geographical position and remoteness. Most of the major sources of MP within the province are concentrated along coastlines leaving the interior isolated from local MP contamination (Government of Newfoundland and Labrador, 2023). The dominant winds come from the west over more populated areas in Canada and the United States, potentially picking up MPs from terrestrial sources, while the island is also bordered by opposing ocean currents

in the southward Labrador Current and northward Gulf Stream which both come from areas with polluted surface waters, another potential source for atmospheric MPs (Loder et al., 1998; Jones-Williams et al., 2021). Lastly, Newfoundland is susceptible to extreme weather events (Darack, 2006; Plante et al., 2015) meaning this research is not only extremely relevant to the region but has potential to grow into a long-term study.

Though Newfoundland's coastline is generally very rugged and barren, the interior of the island consists of an elevated, undulating plateau generally dipping northeast (Government of Newfoundland and Labrador, 2024). This interior plateau is characterized by boggy land, forests consisting of small conifers, and thousands of inland lakes of varying size that were formed during the last glaciation (Twenhofel & MacClintock, 1940). Only a small proportion of the lakes are connected to major river systems, with many of the smaller lakes and ponds connected only to small streams or being endorheic. That means that for many of these lakes, with no nearby point sources or riverine transport pathways, the atmosphere may be the key transport mechanism for MP pollution found there.

Newfoundland has a temperate marine climate. The southwestern region of the island receives the most precipitation on average, ranging between 1200 – 1700 mm annually, while the north-central region is the driest averaging between 800 – 1100 mm of precipitation annually (Government of Newfoundland and Labrador, 2024). This in turn means the southwestern region experiences the most runoff (averaging between 1300 mm to 2100 mm annually) while the north-central experiences the least (averaging between

700 mm to 900 mm annually; Government of Newfoundland and Labrador, 2024). Newfoundland also experiences major storms such as hurricanes, nor'easters, and extra-tropical cyclones (Darack, 2006). Therefore, the island of Newfoundland presents a unique location to study atmospheric MP deposition in remote regions.

There has been little MP research done in Newfoundland to date, especially concerning atmospheric transport and deposition. Only one study has looked at MP occurrence in a freshwater environment, and that was an urban lake that had multiple nearby sources (Liboiron et al., 2020). Previous studies have focused on MP occurrence in coastal zones or plastic ingestion by marine species such as fish or seabirds (Liboiron et al., 2020). A report done in 2020 about plastic pollution throughout the province from 1962-2019 found there was an uneven distribution of shoreline plastic pollution, with small-scale hotspots (Liboiron et al., 2020). The main sources of marine and shoreline plastic pollution were commercial and sustenance fishing, wastewater effluent, litter, leakage from landfills, and burning waste (Liboiron et al., 2020). Although the population of Newfoundland and Labrador has not grown for most of the last decade, there is a projected increase in population growth in upcoming years (Government of Newfoundland, 2023). An increase in population often leads to an increase in plastic usage and production, meaning more locally sourced MPs (Ta & Babel, 2023).

CHAPTER 3 TRANSPORT AND DEPOSITION OF OCEAN-SOURCED MICROPLASTIC PARTICLES BY A NORTH ATLANTIC HURRICANE

Chapter 3 is the manuscript "*Ryan, A.C., Allen, D., Allen, S., Maselli, V., LeBlanc, A., Kelleher, L., Krause, S., Walker, T.R., and Cohen, M. Transport and deposition of ocean-sourced microplastic particles by a North Atlantic hurricane.*" The paper was published in Communications Earth & Environment in December 2023. Details on the role of each author and copyright information are given at the end of this chapter.

SUMMARY

The atmosphere can transport large quantities of microplastics and disperse them throughout the globe to locations inaccessible by many other transport mechanisms. Meteorological events have been proven to pick up and transport particulate matter, however, how they influence the transport and deposition of atmospheric microplastics is still poorly understood. Here I present samples of atmospheric fallout collected during Hurricane Larry as it passed over Newfoundland, Canada in September 2021. During the storm peak, 1.13×10^5 particles m^{-2} day^{-1} were deposited, with a decline in deposition after the storm passed. Back-trajectory modelling and polymer type analysis indicate that those microplastics may have been ocean-sourced as the hurricane traversed the garbage patch of the North Atlantic Gyre. This study identifies the influence of North Atlantic hurricanes on the atmospheric transport and deposition of ocean-sourced microplastics and the possible consequences of increased exposure to microplastics in remote areas.

3.1 Introduction

Microplastic (MP) pollution is a growing environmental and public health concern that has been detected in all environments around the globe (Allen et al., 2019; Bergmann et al., 2019; Liu et al., 2019; Zhang et al., 2019) with the risk to cause significant harm to humans, wildlife, and overall ecosystem health, as well as affect climate processes (Cole et al., 2013; Schirinzi et al., 2017; Karbalaei et al., 2019; Marn et al., 2020; Revell et al., 2021; Walker et al., 2022). MPs are generally sourced in continental regions with high levels of anthropogenic activity and then are transported, primarily by rivers, to oceans (Schmidt et al., 2017; Lebreton et al., 2017; Meijer et al., 2021). Therefore, most MP research during the last decade has focused on describing the abundance, composition, suspected sources, and impacts of MPs in such environments (Allen et al., 2022a; Andrady 2011; Browne et al., 2011; Cole et al., 2011; Dubaish & Liebezeit, 2013; Nyberg et al., 2023). Thus, the fate and transport processes of MPs in the atmosphere were not as thoroughly investigated.

Recent studies, however, have determined that the atmosphere may play a crucial role in the transport and deposition of MPs, especially in remote terrestrial regions that have no local source of plastic pollution (Allen et al., 2019; Zhang et al., 2019; Brahney et al., 2020). Furthermore, it has been shown that the ocean, so far considered a long-term sink for MPs, can also act as a source of plastic pollution, with small MPs escaping from the sea surface into the atmosphere via bubble burst ejection or wave action, allowing for a potentially continuous cycle of transport and deposition of MPs (Allen et al., 2020). The idea that MPs could be transported through the air as well as by ocean waters has opened

a new paradigm in plastic research, questioning for instance how major weather events such as tropical or extratropical storms may affect MP transport pathways, deposition patterns, and amounts. Previous studies have found that intense precipitation can increase the abundance of larger atmospheric MP fallout, while high winds and storm systems can broaden the potential transport distances (Abbasi et al., 2022; Li et al., 2022). These observations suggest hurricanes, known for intense precipitation and sustained high winds, could be able to transport MPs across oceans faster than oceanic currents and increase deposition amounts to places that may not receive regular MP deposition from other sources.

Hurricanes in the North Atlantic Ocean usually form off the west coast of Africa, gaining energy as they travel across warm tropical waters and then dissipate over land or colder waters along the east coast of North America. Hurricanes can span hundreds of kilometres in diameter and have winds exceeding 250 km/h (NOAA Atlantic Oceanographic & Meteorological Laboratory, 2021). Such strong winds at ground level have the capability to resuspend large amounts of terrestrial particulate matter, including pollutants, into the atmosphere and transport them, along with particulate matter already in the air, for thousands of kilometres (Harriss et al., 1984; Steinnes & Friedland, 2006). Increased wave activity and strong winds at the sea surface can also resuspend particles entrained in ocean surface waters (Allen et al., 2020; O'Dowd & de Leeuw, 2007; Quinn et al., 1975), whereas heavy rains associated with hurricanes wash particles and pollutants out of the atmosphere, depositing them in locations which could be at far distance from their source (Pan et al., 2017). This has been demonstrated, for example, by the presence

of frozen plankton in ice crystals of cirrus clouds over Oklahoma after Hurricane Nora in 1997 (Sassen et al., 2003), or by the elevated levels of air pollutants, including particulate matter ($PM_{2.5}$), after Hurricane Florence struck North Carolina in 2018 (Bhandari et al., 2022). Consequently, hurricane-driven transport may represent an important mechanism for dispersing MPs to remote locations.

In this study, I investigated the impact of Hurricane Larry on atmospheric MPs deposition in the North Atlantic region by collecting air samples from before, during, and after the passage of the hurricane over Newfoundland, Canada, where it made landfall as a Category 1 hurricane on 11 September 2021 (Brown, 2021; Figure 3.1). The island of Newfoundland has very low population density (1.4 persons km^{-2}; Statistics Canada, 2021), with most of the population living in the provincial capital, St. John's, leaving the rural areas sparsely inhabited. Due to the limited presence of industries beyond the capital city (Government of Newfoundland and Labrador, 2023), most rural areas, particularly in the island's interior, are not expected to possess significant sources of atmospheric MPs. This implies that, apart from coastal and river floodplain regions, the predominant mode for transporting MPs to these remote areas is through the atmosphere. Newfoundland frequently experiences various weather events such as extratropical cyclones, nor'easters, and hurricanes (Darack, 2006; Plante et al., 2015). These weather phenomena can exert a considerable influence on the deposition of atmospheric MPs, making Newfoundland an analog for any remote or sparsely populated mid-latitude terrestrial area that faces extreme atmospheric events. Furthermore, Hurricane Larry followed a track far offshore of the eastern seaboard of the United States and Eastern Canada, maintaining its

trajectory over the ocean and traversing the North Atlantic garbage patch before making landfall in Newfoundland (Law et al., 2010; Figure 3.1, Supplementary Figure 1A). Thus, Hurricane Larry presented a unique opportunity to study MP deposition from a hurricane that had not travelled close to any major urban areas in a sparsely populated location with no obvious local point sources of MPs.

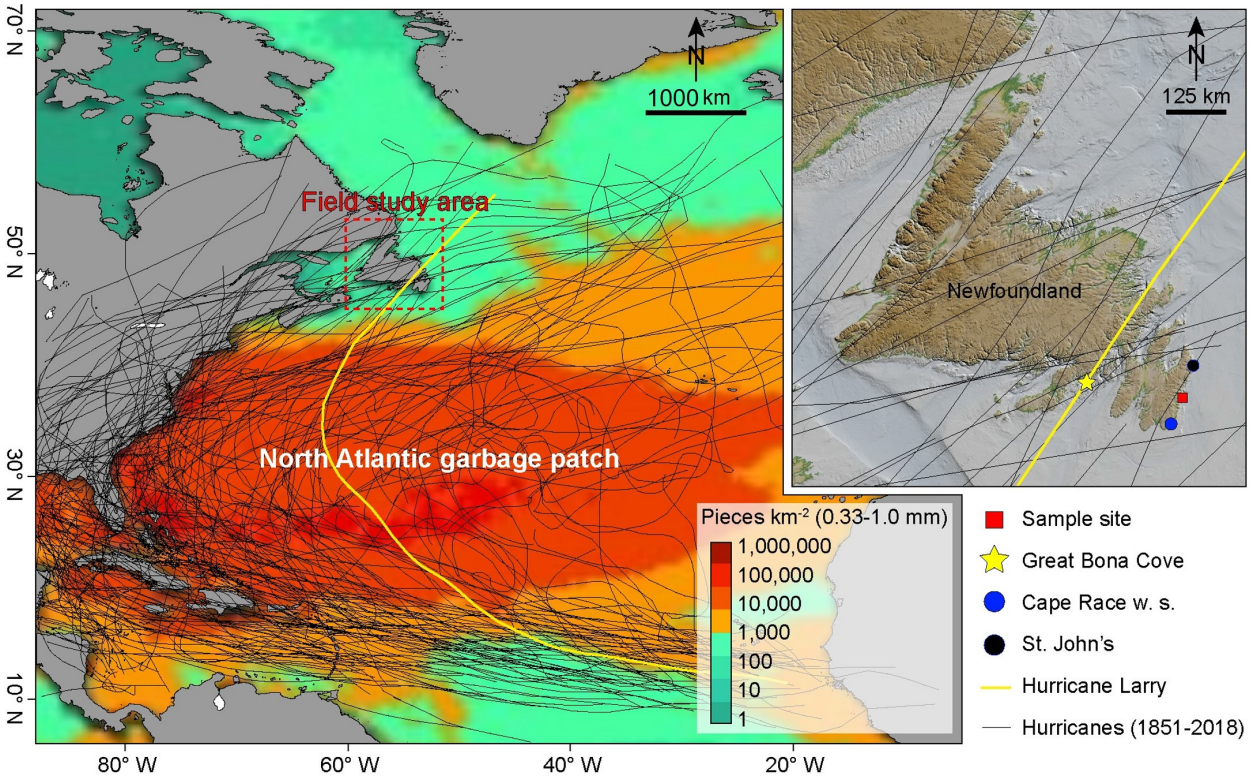


Figure 3.1: Study Area. Past hurricane (>category 1) tracks from 1851 to present (black lines) with Hurricane Larry track (yellow line) and plastic debris concentration (in pieces per square kilometre) in the surface waters of the North Atlantic Ocean, a possible source of microplastics found in these samples (Eriksen et al., 2014). Field study area (dotted red square) is shown (inset) with the locations of the sample site (red square), Cape Race weather station where meteorological conditions were recorded (blue circle), St. John's (nearest urban area, indicated by the black circle), and Great Bona Cove (location of hurricane landfall, yellow star).

3.2 Methods

3.2.1 Sample collection

MP samples were collected in a sterilized 1 m tall glass deposition collector vessel with a 200 mm diameter that was placed on the ground and firmly secured to 3 wooden stakes using black zip ties (Allen et al., 2019). Black MPs are difficult to analyze using μRaman spectroscopy and Nile Red fluorescence and were discounted from sample findings to ensure minimized field contamination of the samples. The vessel was situated at the centre of a clearing, approximately 100 meters from the coast. The clearing presented an area with a higher elevation than the surrounding area, unobstructed by trees, houses, or other structures. Bulk deposition samples were collected every 6 hours commencing September 9 and ending September 12, resulting in a total of 11 samples. The 6 hourly sample duration was selected due to the hazardous nature of collecting deposition samples during the hurricane, acknowledging that hourly samples would have been preferable. The site was selected due to its accessibility, proximity to the coast, and remoteness. It was relatively remote and surrounded on 3 sides by ocean, ensuring there were no direct local point sources of MPs.

At the beginning of each sampling period, the sampling vessel was filled with 100 mL Milli-Q water and the relative wind speed and approximate direction were recorded. Since an anemometer was not available, wind speed was recorded using the Beaufort scale and wind direction was based on visual cues. After 6 hours, the contents of the vessel were decanted into a sterilized glass jar. The sides of the deposition collector vessel were rinsed three times with an additional 100 mL Milli-Q into the jar (Dris et al., 2016). The jar was then firmly shut, labelled, and wrapped in aluminum foil ready for transport back to the laboratory. Field blanks (n=3) were created following sample

protocol for the first, middle, and last samples to account for contamination during sample collection. Precise separation between wet and dry deposition was not possible during the hurricane due to safety concerns so samples were labelled as bulk deposition.

Sample contamination was minimized by taking extra precautions both during sample collection and sample analysis. The sample collection vessel was always approached from downwind and rinsed three times with Milli-Q at the end of each sampling period into the jar with the sample. The jars were wrapped in aluminum foil and paper and stored upright in a cardboard box. They were also transported upright and stored in a 4ºC fridge in these boxes until laboratory analysis could begin. Field blanks were created as full process control blanks that went through all the same steps as the samples. Blanks were also created during laboratory MPs extraction, plus positive controls of spiked samples, to account for potential contamination and sample loss during processing. Glassware was used during both sample collection and analysis. Glassware was rinsed three times with Milli-Q and covered with aluminum foil after every use and between samples.

3.2.2 Laboratory sample preparation and analysis

Samples were transported to a dedicated MP analysis laboratory where they were digested to remove organic material, density separated for sediment and mineral particulate removal and then filtered onto aluminum oxide filters. Surfaces were cleaned with paper towel and Milli-Q then covered in aluminum foil every week, while the entire laboratory underwent a thorough cleaning with Milli-Q each week. During digestion,

sample temperatures were consistently checked to ensure the samples remained under 50ºC so as to not damage any plastic particles. Samples were analyzed in triplicate, with the volume of each sample recorded prior to any sample preparation activities. Samples were filtered onto 1.2 µm Whatman glass fibre filters (GF/C, 47 mm diameter) and the filtered residual material was digested using 30% w/w hydrogen peroxide (H_2O_2) in sterilized, covered borosilicate glass test tubes at 50 °C (dry block) for 48 hours. Samples were then vacuum filtered (1.2 µm pore GF/C 47 mm diameter filters) and the remaining material placed into density separation flasks with 1.6 g cm^{-3} zinc chloride solution ($ZnCl_2$) for a further 48 hours. The suspended particles in $ZnCl_2$ (upper ½ of the density separation flask) were then filtered onto 25 mm diameter aluminum oxide filters in preparation of µRaman analysis. All filters were stored individually in 50 mm diameter sterilized aluminum containers to minimize sample contamination.

All samples were analyzed using µRaman spectroscopy followed by Nile Red fluorescence microscopy. MP particle counts and polymer composition results are derived directly from µRaman analysis, with fluorescence microscopy providing the particle size distribution results. µRaman analysis was undertaken using a Renshaw InVia spectrometer at the University of Birmingham (United Kingdom) with a 780 nm wavelength laser, 1200 gr mm-1 grating and using up to 25% power (filter). Spectra were collected over 200-3000 cm^{-1} using a minimum of 5 acquisitions of 10 s. Filters were analyzed using the sub-sampling method presented in Huppertsberg and Knepper (2018) to ensure a representative 25% of each filter was analyzed (n=2873 particles). µRaman spectra for each particle were then individually analyzed using Spectragryth, the SLOPP,

SLOPPE, Simple (Menges, 2020; Munno et al., 2020; Primpke et al., 2020) and inhouse reference libraries to identify if each particle was plastic and the respective polymer type. Only spectra with an 80% or greater accuracy were considered and counted as plastic polymer to ensure a conservative approach to the analysis was undertaken. Each filter was analyzed three times, with a different section of filter being analyzed each time, to calculate an average count and an error of one standard deviation (1σ). The Limit of Quantification (LoQ) for this analysis (and the subsequent fluorescent analysis) was set to 1.2 µm, acknowledging that the limit of detection (the threshold size for particles to give a signal above background) for µRaman is lower than this (1 µm).

After completion of µRaman analysis, all samples underwent fluorescent microscopy for particle size distribution assessment (necessary due to the limited clarity of µRaman visuals and the absence of particle finder software within the µRaman set up). Filters were dyed using 0.1 µg mL^{-1} Nile Red solution (powder dissolved in 99% pure ethanol (Sigma Aldrich), prefiltered through 1.2 µm GF/C filter), air dried overnight in a 30°C oven (covered in sterilized foil to minimize sample contamination), and then placed in a dark, 4°C fridge prior to analysis. Fluorescence microscopy was completed using an OMAX microscope and imaged using the AmScope MU500 camera and software, with 5% of the sample assessed and the images analyzed using FIJI (ImageJ) (Schindelin et al., 2012). Particles were classified as fragments or fibres, with fibres defined as particles with at least a 3:1 length to width ratio (Löder & Gerdts, 2015).

3.2.3 Back-trajectory modelling

Back-trajectory modeling was carried out with the HYbrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model version 5.2 using Global Data Assimilation System GDAS 1-degree archived global meteorology data (https://www.ready.noaa.gov/gdas1.php). Models were run for 48 hours in backwards mode from a starting elevation of half the planetary boundary layer (~500 m above mean sea level) calculated dynamically by the model at any given starting time above the sampler. The mid-boundary-layer starting height was chosen as it best represents the source of air in the well-mixed planetary boundary layer above the sampler. As a sensitivity test, I ran back-trajectory simulations from a starting elevation of one-quarter and three-quarters of the planetary boundary layer, and the results were not significantly influenced (see Supplementary Figure 9A). The hourly endpoints from each trajectory were summed over a 1-degree grid, and the gridded frequencies were calculated as the number of endpoints in a given grid square divided by the total number of endpoints in all of the trajectories being considered (see Supplementary Note 4A). A new trajectory simulation was run for every hour within the sampling period, resulting in 66 individual trajectories, which were then mapped using Esri (ArcMap 10.4). I judged that any air parcels within 1000 m of the surface as possible entrainment points, as this lower section of the atmosphere experiences sufficient vertical mixing to transport MPs from the surface throughout this section of the air column relatively quickly (Angevine et al., 1994).

3.2.4 Blanks, positive controls, and contamination mitigation

The field blanks were managed and assessed as full process blanks, representing not only the potential field contamination but also any sample contamination due to laboratory sample preparation and analysis. Spiked samples were also prepared and processed as positive controls to quantify analysis efficiency and accuracy (see section 4.2.3 for more information). Sample blank and spike corrections were applied to MP counts from both μRaman spectroscopy and Nile Red fluorescence analyses. Sample analysis was carried out in a clean laboratory outfitted with HEPA filters and restricted access, with all persons in the laboratory required to wear 100% cotton clothing and laboratory coats. All processing steps were done under a laminar flow fume hood using glassware, while all liquid used, including chemicals and Nile red, were prefiltered using a filter of same porosity used in sample filtration (1.2 μm GF/C filter).

3.3 Results and discussion

3.3.1 Hurricane Larry

Hurricane Larry originated offshore the west coast of Africa (11ºN, 20ºW) on August 30, 2021 as a tropical depression and evolved into a category 3 hurricane on September 4 approximately 1600 km east of the British Leeward Islands before making landfall as a category 1 hurricane at 00:30 September 11 near Great Bona Cove (Newfoundland, Canada, Figure 3.1; Brown, 2021), approximately 130 km west of the sampling location of this study (47°11'0.15" N, 52°50'33.50" W; Figure 3.1). Sample collection commenced the evening of September 9, and ended September 12, 2021, with a new sample being collected every 6 hours for a total of 11 composite samples each collected over a 6-hour

period. The sample location experienced fair-weather conditions prior to the evening of September 10 (samples 1 – 3), with wind blowing mainly from the south at average speeds <22 km/h, and gradually increasing leading up to the hurricane (measured at the Cape Race weather station located approximately 60 km south of the sample site; Figure 3.2). The duration of true hurricane anti-cyclonic activity extended from the early evening of September 10 to the morning of September 11 (samples 4, 5, and 6), with a minimum atmospheric pressure of 980 hPa and a maximum sustained wind speed of 109 km/h detected at 00:30 on September 11 (during sample 6) at the Cape Race weather station, marking the passing of the eye of the hurricane. Moderate rainfall occurred from 22:00 September 10 to 01:00 September 11, for a recorded total of 9.4 mm at Cape Race (Figure 3.2). Wind direction at Cape Race shifted from southwest during the height of the storm to gentler westerly winds after the hurricane passed (samples 7 – 11) with speeds between 20 – 50 km/h. No local measurements of wind speed, direction, or amount of precipitation were possible at the sample location due to a lack of equipment; however, weather data collected at Cape Race (Supplementary Figure 2A), located approximately 60 km SSW of the sample location, generally agrees with the observations made at the sampling site.

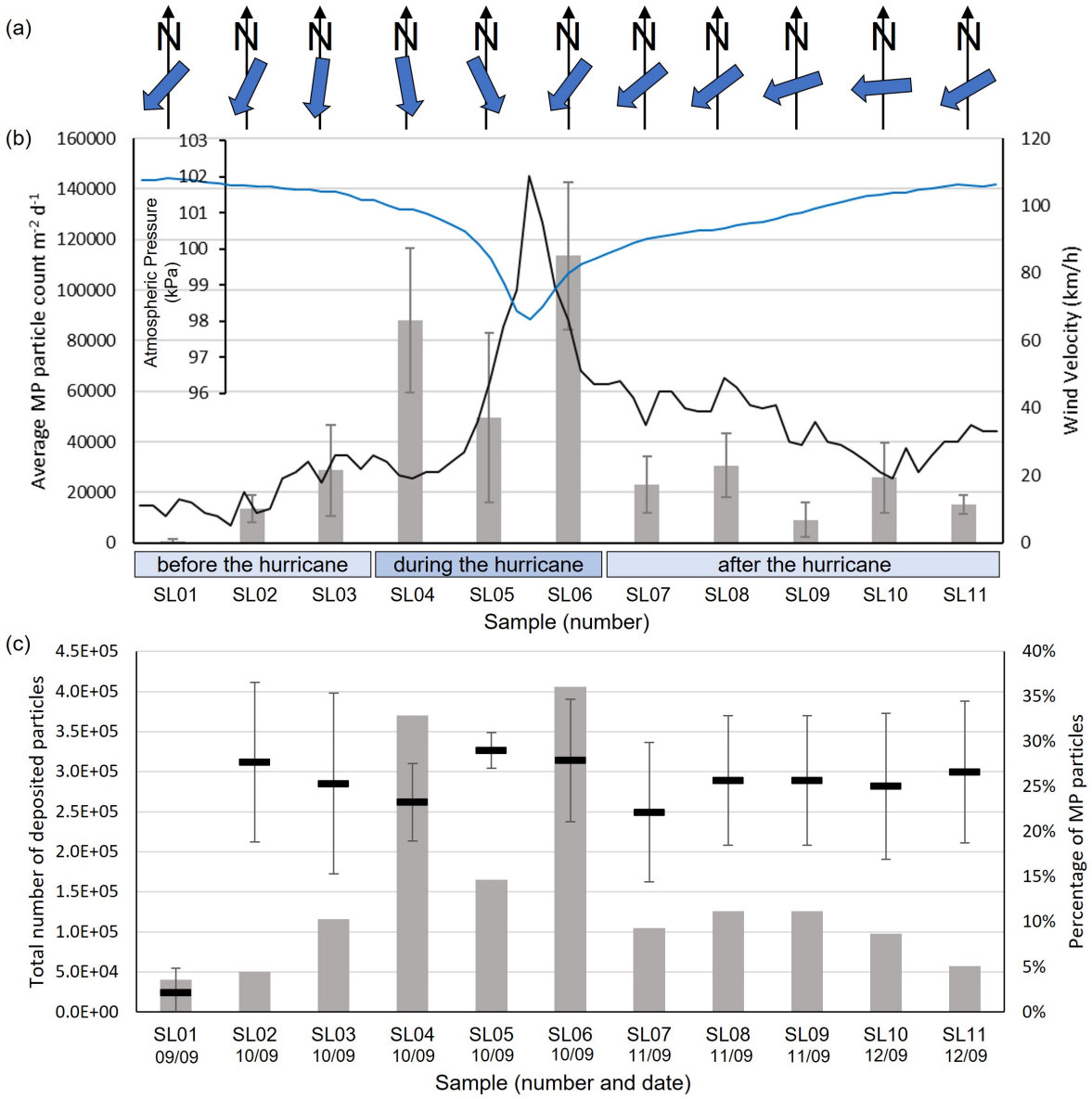


Figure 3.2: MP abundance and weather conditions. (a) dominant wind direction (indicated by blue arrow) during sample period observed at sample site at the beginning of each sample period, (b) average microplastic (MP) particle count (with error bars representing 1σ) for each sample period (in m^{-2} d^{-1}) with wind speed indicated by black line and atmospheric pressure indicated by blue line (collected at Cape Race weather station) showing increase in microplastic deposition in samples collected during the hurricane, and (c) percentage of microplastic particles (indicated by black bars, with error bars representing 1σ) relative to total number of particles deposited (in m^{-2} d^{-1}) during each sample showing there was not preferential deposition of microplastics and increase in deposition of all particles during hurricane conditions, with date of sample commencement indicated below. Sample 1 may be contaminated, indicated by anomalous proportion of microplastics to total particles.

3.3.2 MP deposition

MPs were consistently detected in all samples, including those taken before (samples 1 – 3), during (samples 4 – 6), and after (samples 7 – 11) Hurricane Larry's passage over the sampling location. Samples collected during the passage of the hurricane (samples 4 – 6) exhibited the highest concentration of MP particles (Figure 3.2). The maximum MP count of 113,569 ± 29,215 MPs m^{-2} d^{-1} was detected in sample 6, deposited shortly after the recorded peak in wind velocity and minimum atmospheric pressure (Figure 3.2b). Subsequently, in sample 7, which was collected after the hurricane had passed, there was a marked decline in MP particle counts. During the hurricane, I observed the highest values for total particle counts and numbers of MPs deposited. In addition, while the amount of MP deposited was relatively similar for pre- and post-hurricane samples (excluding sample 1), the total particle count was on average slightly higher after the hurricane in comparison to before it (Figure 3.2). I also found a significant positive correlation between the total particle deposition per day and the average amount of MPs in the samples ($r = 0.97$, $p < 0.05$).

Similar to wind speed, there is an observed upward trend in particle deposition before the hurricane, followed by a decrease immediately after the hurricane, with relatively stable deposition levels afterward. A positive correlation exists between both total particle deposition and MP deposition and the maximum wind speed recorded during the sampling period ($r = 0.61$, 0.68, $p = 0.04$, 0.02, respectively). However, it's important to note that there is no significant correlation between deposition and minimum wind speed or dominant wind direction during the sample period. Additionally, there is a positive

correlation between both total particle deposition and MP deposition and the total amount of rainfall ($r = 0.68, 0.73$, $p = 0.02, 0.01$, respectively). The percentage of particles identified as MP from the total amount of particles deposited remained relatively constant throughout the entire sampling period (Figure 3.2c), staying within the range of 20 – 30%.

The deposited MPs identified in sample 1 were significantly reduced, with only 753 particles m^{-2} d^{-1} (Figure 3.2). Furthermore, the percentage of MP in sample 1 was anomalously low at just 2%. Since weather conditions and total number of particles deposited were relatively similar between sample 1 and sample 2, I decided to treat sample 1 as an anomaly that may be due to human error during collection or analysis, and thus it was excluded from all statistical analyses. The sampling vessel broke due to high winds during sample 6 introducing possible loss of sample, though both MP and total particle deposition was still much higher than other samples.

3.3.3 Comparison to previous deposition

Few studies currently exist on atmospheric MP deposition during a storm event. In the South China Sea, a peak deposition of 2,014 MPs m^{-2} d^{-1} was obtained during the passage of Typhoon Sinlaku (Li et al., 2022), while 22,608 MPs m^{-2} d^{-1} were detected during monsoon rains in Iran (Abbassi, 2021). During the passage of Hurricane Larry over Newfoundland, I recorded a maximum MP count of 113,569 ± 29,215 MPs m^{-2} d^{-1}. This is a significantly higher value, particularly when considering that this sampling site was in a remote location whereas the other studies collected samples close to a densely

populated urban regions (Li et al., 2022) or in an urban centre (Abbassi, 2021). These results may indicate that the North Atlantic Ocean was the likely source of such high concentration of MPs detected in the Hurricane Larry samples.

Counts of atmospheric MP deposition in remote locations during non-storm conditions typically fell within the range of ca. 500 to 6,000 MPs m^{-2} d^{-1} (Allen et al., 2019; Brahney et al., 2020; Hamilton et al., 2021). In this investigation, the samples taken before (samples 1 – 3) and after (samples 7 – 11) Hurricane Larry's passage over the sampling site presented an MP count between 700 and 30,000 MPs m^{-2} d^{-1}. While these values are comparable, they tend to be higher, and this discrepancy may either suggest that these samples could have been influenced by the hurricane's winds, given the extensive reach of the storm, or that this study site has inherently higher background MP levels than other locations, possibly due to its proximity to the ocean.

The percentage of MPs relative to total particle deposition (between 20 – 30%) after digestion and density separation agrees with previous studies (Cai et al., 2017; Dris et al., 2016), evidencing that there is an overall increase in atmospheric deposition during storm events and thus highlights how hurricanes can input substantial amounts of MPs to locations that would otherwise most likely experience very little MP deposition. The sample site chosen for this study had no local (within a 100 km radius) sources for MP pollution as it is located in a rural area and received onshore winds, meaning the main mechanism for MPs deposition to this area is likely to be atmospheric fallout. Hurricanes

affect a much larger area than just where the eye hits so increased deposition would be expected on a regional scale.

So far, Newfoundland has rarely experienced hurricanes greater than Category 1, such as Hurricane Larry. However, the warming of the North Atlantic Ocean due to anthropogenic climate change is expected not only to increase the intensity, duration, and frequency of hurricanes (Knutson et al., 2021) but also to promote a northward shift in their trajectory, thus posing remote areas at higher latitudes at increasing risk of atmospheric plastic pollution. This study does not only evidence this risk for the first time but also provides a baseline to evaluate the relation between hurricane intensity and MP fallout for future events.

3.3.4 Particle size, shape, and polymer composition

The limit of quantification for particle size analyzed by Nile red fluorescence was 1.2 µm, set by the filter porosity. The majority (65%) of particles analyzed were of 2 – 10 µm size (Supplementary Figure 4A). Of the remaining particles analyzed, 34% were <2 µm, with particles between 10 – 20 µm making up only 1% of particles on average. Particles up to 100 µm were found in sample 6; with MPs between 20 – 100 µm accounting for 0.6% of the total particles analyzed in the sample. The largest particles found in all other samples were <40 µm (Supplementary Figure 5A). There was no significant correlation between MP sizes and wind speed or rainfall which was unexpected as previous literature has found larger particles deposited during precipitation (Li et al., 2022); however, there

was very little precipitation (<10 mm) over the course of the sampling period including during the peak of the hurricane (Supplementary Table 1A).

Most of the particles identified were classified as fragments (Supplementary Figure 5A), with fibres accounting for less than 1% of the total particle count in all samples. This may be due to the small size of the particles, as fibres can break down into smaller pieces that are then classified as fragments (Hidalgo-Ruz et al., 2012; Löder & Gerdts, 2015). Additionally, dust deposition samplers have been shown to create a deposition shadow wherein there is a decrease in sampler efficiency with increasing wind speed and particle size, as well as preferential collection of finer particles (Goossens, 2010). The predominant particle sizes during the hurricane were in the same 2 – 10 µm range of those before and after the hurricane, with a skewed distribution towards smaller sizes. There was a slightly larger proportion of particles between 2 – 10 µm deposited during the hurricane compared to before and after, but not by a statistically significant (p-value=0.56>α=0.05) amount.

Overall, the polymer composition of MPs showed little variation between individual samples (Figure 3.3). Polymethyl methacrylate (PMMA) was the most abundant polymer in most samples ranging between 14 – 26% (Figure 3.3), though made up only 6% of MPs in sample 11 (Supplementary Figure 6A). This sample also had a much larger portion of polyethylene terephthalate (PET) and polyethylene vinyl acetate (PVA) than any other sample. The second most abundant polymer types were acrylic and polyester (PES), which both accounted for 14% of MPs analyzed overall. The distribution of PES

MPs was relatively constant throughout the samples, except for samples 2 and 10 which only carried 3% and 7% PES particles, respectively. When comparing conditions before (samples 1 – 3), during (samples 4 – 6), and after (samples 7 – 11) Hurricane Larry, there is little variation in polymer proportions, although the amount of PMMA decreases during the hurricane while PES increases (Figure 3.3). Acrylic is present in all samples in different amounts: MPs in sample 1 consist entirely of acrylic, which may indicate possible contamination (Supplementary Figure 7A), while acrylic particles in samples 2 represent 22% of the total, and 17% in samples 6 and 9. The proportion of nylon/polyamide (PA) was highest in samples collected during the hurricane, accounting for 7% of particles in samples 4 – 6 and only 3% of MPs in pre- and post-hurricane samples (Figure 3). I found there was a positive correlation between the average MP deposition and the proportion of nylon/PA in a sample ($r = 0.71$, $p = 0.01$). There is a noticeably lower proportion of polysulfone (PLS) in samples 1 and 2 (0% and 6%, respectively), while PLS is one of the more dominant polymer types in all the other samples with the proportion of PLS MPs increasing throughout the entire sampling period.

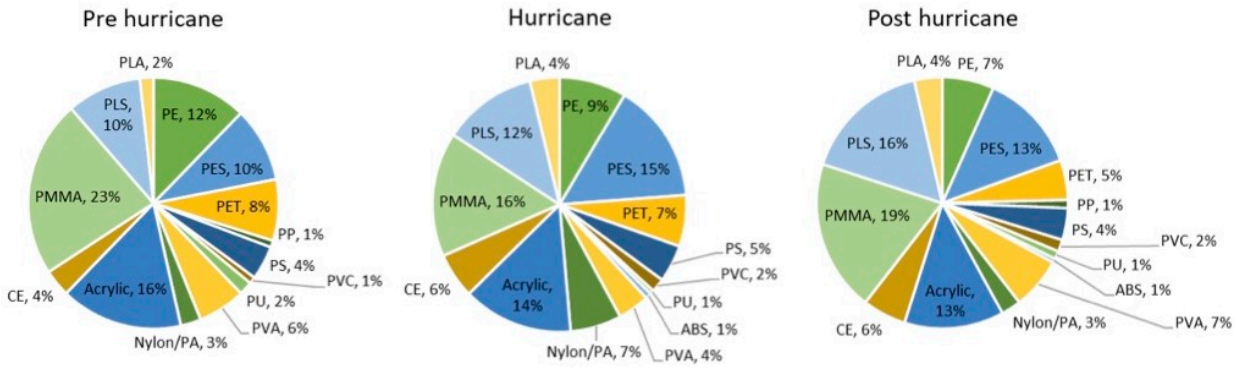


Figure 3.3: Polymer types of deposited particles. Average proportion of each polymer type of microplastic particles collected before (samples 1-3), during (samples 4-6), and after (samples 7-11) Hurricane Larry (see Supplementary Table 2A for a full list of polymer names and abbreviations).

3.3.5 Back-trajectory modelling

Back-trajectory modelling performed with HYSPLIT version 5.2 (Stein et al., 2015; Rolph et al., 2017) was used to determine possible entrainment points and pathways of air parcels that passed over the sampling site during the investigated time interval (Figure 3.4). Any elevation under 1000 m above surface level is deemed to act as an entrainment point for MPs (Allen et al., 2020) as vertical mixing transports MPs lifted off the surface throughout the air column up to 1000 m (Angevine et al., 1994). Once entrained in the atmosphere, MPs in the size range investigated in this study (<100 μm) have an atmospheric lifetime of over 48 hours (Evangeliou et al., 2022), which is consistent with the length of the time window chosen for the back-trajectory analysis. The model shows that within 48 hours of arriving at the sample location, a significant fraction of air parcels passed over the North Atlantic Ocean (Figure 3.4), and thus possible entrainment points are from the marine environment (Figure 3.5). The model also shows that some possible entrainment points are over the continent, particularly for samples 7 to 9 (Figure 3.5). Nonetheless, the absence of a significant difference in polymer type and size distribution in samples 7 to 9 in comparison to the other samples with potential entrainment points over the ocean strongly indicates that the ocean remains the predominant source of microplastics in these particular samples. Sample 1 was not included in the analysis since it was considered contaminated.

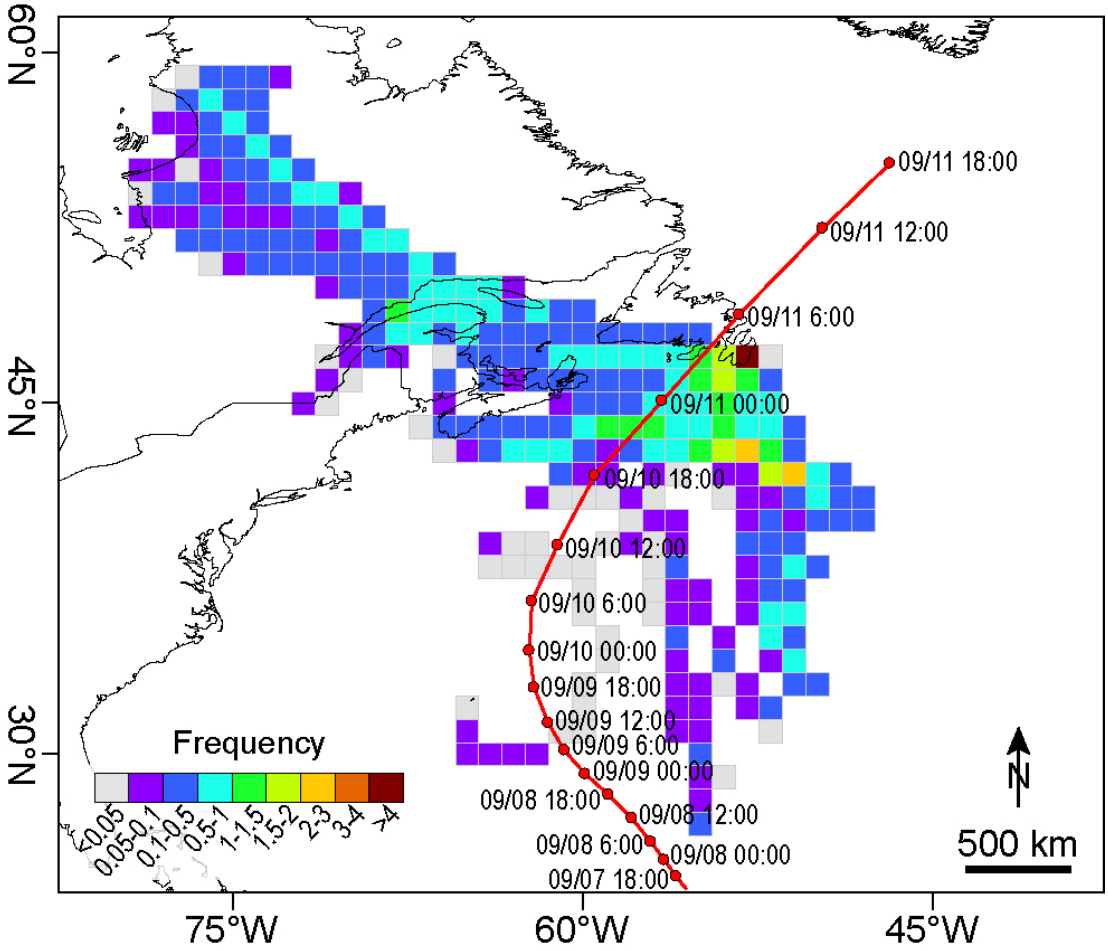


Figure 3.4: Back-trajectory atmospheric modelling. Gridded frequency distribution of hourly endpoints calculated for all 48-hour HYSPLIT back trajectories (n = 66), started each hour within the 66-hour sampling period. All endpoints from 0 – 10000 m above ground level were included. The frequency represents the number of endpoints in each grid square divided by the total number of endpoints (66 x 48 = 3168) for each 1 x 1 degree grid square. Hurricane Larry track is indicated in red with corresponding date and time.

Before and during the hurricane, most of the potential entrainment points were to the southwest and southeast of both the hurricane track and the sampling location, over the North Atlantic Ocean (Figure 3.5), as indicated by the dominant wind directions during these periods (Figure 3.2). After the hurricane passed, the potential entrainment points shifted to the west of the sample location, following the dominant wind directions (Figure 3.2). Within 48 hours of reaching the sample site, the air parcels generally have an

elevation of < 1000 m (Supplementary Figure 8A). Air parcels arriving after the hurricane, however, are generally at higher elevations, reaching over 2000 m (Supplementary Figure 8A). These air parcels also travelled for longer distances (>1000 km) in 48 hours, crossing the provinces of Quebec and New Brunswick (Figure 3.5). Air parcels leading up to the hurricane had the shortest transport distance between possible entrainment points and the sample site (20 – 35 km, see Supplementary Table 1A) and were generally at lower elevations (Supplementary Table 1A). Potential entrainment points for pre-hurricane samples were mostly over the ocean, with only few and low frequency trajectories crossing parts of Nova Scotia and Prince Edward Island (Figure 3.5). Air parcels arriving during the hurricane have potential entrainment points over or close to the North Atlantic garbage patch (see location in Figure 1) within the 48-hour backwards time step, with the sample 6 model showing entrainment points through the middle of the garbage patch (Figure 3.5).

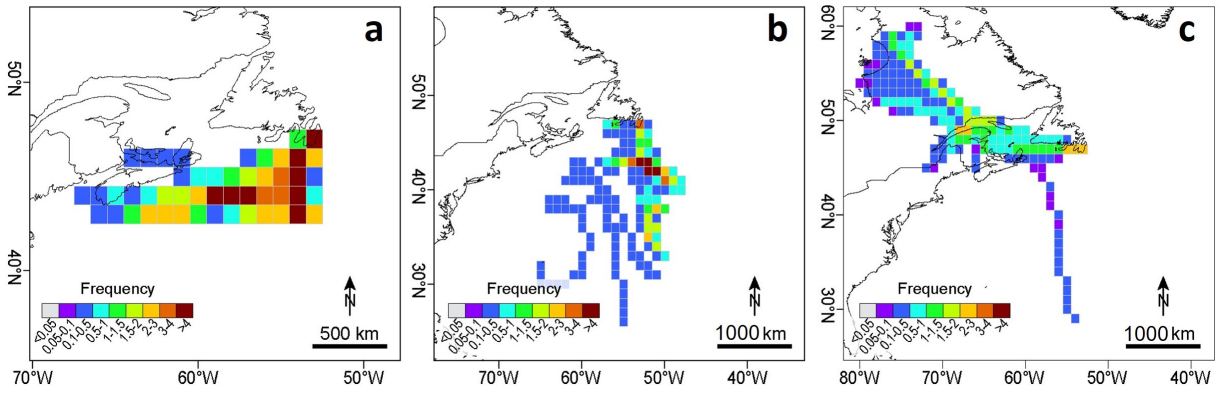


Figure 3.5: Possible microplastic entrainment points. HYSPLIT air parcel 48-hour back-trajectory endpoint frequency distributions indicating locations of possible entrainment points of microplastic for samples collected (a) before (samples 1 – 3), (b) during (samples 4 – 6), and (c) after (samples 7 – 11) the passage of Hurricane Larry over Newfoundland. To represent possible MP entrainment locations, only endpoints with elevation < 1000 m were counted in the frequency summation.

Statistical analysis shows there is no significant correlation between the distance to the closest possible entrainment point and MP abundance ($r = 0.53$, $p = 0.11$; Supplementary Figure 10A). Also, no significant correlation exists between the size distribution of MP particles found in the samples and the distance from possible entrainment points ($r = 0.15$, $p = 0.67$) and between the average distance the air masses travelled and the dominant polymer type in a sample. In pre-hurricane samples, the possible entrainment points are tightly packed together and are mostly within a 500 km radius. Conversely, possible entrainment points during and after the hurricane are spread over a much larger area, with some entrainment points >3000 km away from the sampling location (Figure 3.5). Possible entrainment points have the greatest range in distance for sample 6, which also had the highest numbers of total particles and MP deposition. Air parcel trajectories for post-hurricane samples were all relatively similar, with on average higher frequencies per grid square, while trajectories for samples collected during the hurricane have a greater spread and are more variable (Figure 3.5). For this model, estimated total error ranges between 15-30% of the travel distance (Draxler et al., 2007) and is largely due to inaccuracies in the meteorological fields (wind speed and direction) used to drive the HYSPLIT model. However, the calculated back-trajectories only represent the centerline of a dispersing plume of emitted material, and so, a trajectory does not have to pass directly over an entrainment point for emissions from that point to have impacted the sample. A sensitivity analysis was also performed by starting the model from different elevations, showing there was relatively little variation by changing this parameter (Supplementary Figure 9A).

3.3.6 Source of MP

Hurricane Larry followed a northward trajectory over the northwest Atlantic Ocean, with the eye of the hurricane staying several hundred kilometres offshore of the east coast of North America until making landfall (Figure 3.1). This implies that the hurricane did not pass over any highly polluted urban terrestrial areas, which are considered a source of airborne MPs (Wright et al., 2020), before hitting the sample site, which is also far from urban areas. Hurricane Larry did pass over the North Atlantic garbage patch in the Sargasso Sea (Law et al., 2010), where concentrations up to 580,000 plastic (>335 μm) pieces/km^2 (0.58 pieces m^{-2}) in surface waters have been estimated previously (Law et al., 2010). Additionally, Hurricane Larry passed over the Labrador Current, which brings waters from the Arctic Ocean (Loder et al., 1998), where studies have found concentrations up to 282 MPs (>50 μm) m^{-3} (43 MPs m^{-2}) in sub-surface (7 m depth) waters (Jones-Williams, 2021). Considering the abundance of MPs in surface waters in Hurricane Larry's path (and associated regions of extreme meteorological conditions) and the back-trajectory calculations, it is highly likely that the high amounts of MPs found in all the samples were sourced predominantly from those marine environments (Figure 3.4). Although calculations are uncertain, previous studies have estimated that the ocean is the most dominant source for the atmospheric limb of the global MP cycle (Evangeliou et al., 2022; Brahney et al., 2021).

In samples 2 – 11, the most dominant polymer type is PMMA, which accounts for a maximum of 26% of the total MPs. This polymer type has many uses, including in marine industrial activities because of its resistance to biofouling from algae or marine

bacteria (Dibke et al., 2021). Nylon/PA is commonly used in fishing gear such as nets (Thomas & Hridayanathan, 2006), providing further evidence in addition to the back-trajectory models that the MPs found in the samples were likely sourced from the ocean. PLS is a highly resistant thermoplastic used as a substitute for glass or metal by some industries (Yu et al., 2019), such as offshore petroleum production, which is present in the region affected by Hurricane Larry. The abundance of PLS in these samples can be linked to such activities. A major source of acrylic and PES MPs is synthetic fibres from clothing, with abundances of these polymers generally high in wastewater effluent (Browne et al., 2011), meaning these particles were likely sourced on land and transported to the ocean before being resuspended into the atmosphere. The diversity of polymer types in these samples show that the MPs did not come from one source. Even though the back-trajectory calculations show that the particles may have been entrained over a large area, there is not much change in the proportions of each polymer type across these samples, likely indicating that MPs in the samples were picked up from the ocean where there is a relatively even distribution of polymers (Erni-Cassola et al., 2019), as most MP in the marine environment are sourced on land and are transported throughout the ocean indiscriminately (Geyer et al., 2017; Jambeck et al., 2015).

Strong hurricane winds have the potential to increase quantities of MPs picked up from the ocean by amplifying bubble injection and wave exchange processes (Allen et al., 2020). The MPs entrained in the hurricane are then able to be transported far distances through the atmosphere, and as the hurricane weakens over land or colder waters, the MP suspended by the storm are deposited. There is also potential for hurricanes to alter the

normal trajectory of MP through the atmosphere and transport them further than in fair-weather conditions. The back-trajectory model indicates that there were potential entrainment points of MP within a 100 km radius (Figure 3.5), with the closest potential entrainment point (sample 1) located approximately 20 km away from the sampling location (Supplementary Table 1A). However, the air masses did not pass over areas that could be considered a source of MPs. This study is the first demonstration of a hurricane as a mechanism for large-scale transport and deposition of MPs to a remote area.

3.3.7 Potential effects of atmospheric plastic pollution

The recognition that hurricanes may have the potential to disperse and deposit large amounts of MPs is concerning as there are several potential effects on the local environment. The chemicals added to polymers, such as endocrine disrupting phthalates, can leech directly into ecosystems (Dusaucy et al., 2021), affecting the organisms living there. Additionally, MPs can act as transport vectors for contaminants such as heavy metals and viruses, carrying them between ecosystems or into humans (Wang et al., 2019). Studies have found that ingestion of MPs by biota such as zooplankton in aquatic ecosystems, both marine and freshwater, can lead to significantly decreased algal feeding and therefore decreased function and health of the organism (Cole et al., 2013). This may have adverse effects on higher trophic levels (Eriksson & Burton, 2003; Romeo et al., 2015), though there is a current lack of data on bioaccumulation of MPs. Newfoundland has many lakes with limited riverine input or output, and thus MPs deposited in these lakes are likely to stay for a long time, affecting micro- and potentially macro-organisms (Lusher et al., 2017; Rochman et al., 2019). Potential ingestion of MPs by micro-

organisms in soils and other terrestrial environments poses the same threat as to those in aquatic environments, though there is currently significantly less research conducted on terrestrial biota than marine. MPs in both aquatic and terrestrial environments could negatively impact biodiversity in relatively undisturbed ecosystems not only across Newfoundland, but also in other locations that experience major storms. Increasing deposition of MPs in rural areas may also affect the humans living there as drinking water, sourced from wells fed by groundwater, may become contaminated with MPs. The sizes of MPs found in this study were small enough to be transported through pores of sandy or silty sediment, meaning they are able to be transported by groundwater (Viaroli et al., 2022), and may enter aquifers through deposition into and subsequent leaching from lakes or soils. Due to their small size and because they are already airborne, atmospheric MPs can enter organisms, including humans, also through respiration, the health consequences of which are still largely unknown. Consequently, large-scale deposition of MPs from hurricanes may have severe consequences as it increases the exposure to atmospheric plastic pollution also in remote locations with otherwise good air quality.

While there are many concerns about MP for individual organisms and ecosystems, there is also the potential for large-scale effects of atmospheric MPs on climate. Airborne MPs have the ability to affect radiative forcing, meaning they are able to absorb and scatter radiation in the atmosphere (Revell et al., 2021). The effect of this forcing may impact climate change or the natural structure and transfer of heat through the atmosphere. MPs also have the potential to adjust cloud formation and therefore affect rainfall patterns

(Allen et al., 2022b; Aeschlimann et al., 2022). Being able to quantify the concentration of MPs in the atmosphere will also improve climate models determining the effects of atmospheric MPs, especially during events in which there are an elevated number of airborne MPs, such as Hurricane Larry.

3.3.8 Limitations and future work

This study was done as a preliminary pilot study aimed to quantify MPs deposition from a hurricane event. Due to the nature of a hurricane allowing only limited warning of its occurrence and preparedness for the exact flow path of the event before it occurs, I was only able to set up one sampling vessel in one location. Ideally, we'd recommend deploying at least three sampling vessels and to investigate at least three locations simultaneously in order to improve the robustness of data for rigorously assessing potential sample contamination using statistical methods and to determine spatial distribution patterns in polymer types and deposition amounts throughout a hurricane event. Unfortunately, I did not have the equipment to measure rainfall at the sample site, so I had to rely on data collected at Cape Race weather station, located approximately 60 km from the sample site. Future studies would ideally be able to measure precipitation, wind speed, and wind direction at the sample site. Furthermore, to fully quantify the full potential extent of the impact of future hurricanes on the deposition of MPs, I recommend the continued monitoring of multiple hurricane events, forming at different latitudes with different trajectories and strengths.

The HYSPLIT model – and particularly the meteorological model outputs that are used to drive the HYSPLIT model – used in this study have limitations in reconstructing the complex atmospheric dynamics during a hurricane. This includes the use of air parcel (trajectory) modelling instead of particle modelling due to the overall small size of the particles and since MPs are thought to behave differently to the non-plastic particles for which these models were originally designed. MPs have a larger variety of shapes and are often less spherical and have a much lower mass than non-plastic particles, affecting drag and settling velocity. Since Hurricane Larry was short and intense, a 48-hour-long back-trajectory model was used to capture the event, however it is recognized that a 48-hour window might not allow us to reconstruct all of the most distal source locations for some of the samples. Future studies may sample for a longer period pre- and post-hurricane allowing a longer back-trajectory to develop a more complete picture of MP source locations and how they differ between hurricane and fair-weather conditions. Further, future studies may include forward atmospheric fate and transport modeling from entrainment locations – that can be carried out with HYSPLIT and related models – recognizing the uncertainties in deposition processes noted above.

Author contributions: A.C.R. conceived the study, collected and analyzed field samples, performed the back-trajectory modelling, wrote the manuscript, and drafted the figures. D.A. analyzed the samples, contributed to the back-trajectory modelling and drafting of the manuscript and figures. S.A. analyzed the samples and contributed to the drafting of the manuscript. V.M. conceived the study, contributed to the drafting of the manuscript and figures. A.L.B. contributed to sample collection. L.K. supported data

analysis. S.K. supported data analysis. T.R.W. contributed to the drafting of the manuscript. M.C. supported the back-trajectory modelling. A.C.R., V.M., T.R.W., and N.C. contributed to the final editing of the manuscript.

CHAPTER 4 MICROPLASTIC ACCUMULATION IN REMOTE ENDORHEIC LAKES AND THE ROLE OF ATMOSPHERIC DEPOSITION: A CASE STUDY FROM NEWFOUNDLAND, CANADA

Chapter 4 is a draft version of the manuscript "*Ryan, A.C., Maselli, V., Walker, T.R., Kelly N.E., and Merschrod S., E.F. Atmospheric deposition and contamination of microplastics in remote groundwater-fed lakes: A case study in Newfoundland, Canada.*" The paper will be submitted to the Canadian Journal of Fisheries and Aquatic Sciences in April 2024. Details on the role of each author are given at the end of this chapter.

SUMMARY

Microplastics (MPs) have been identified in virtually all environments around the globe, posing a threat to both humans and nature. Though there have been numerous studies on MPs in aquatic environments, there is still a lack of knowledge on MPs in freshwater lakes, especially small endorheic lakes in remote environments. In this study, I present evidence of MP pollution in sediments from lakes across Newfoundland, Canada. I found between 6,000 – 24,000 MP kg^{-1} w.w. in the sediments, with the majority of particles between 2 – 10 μm in size. Since the lakes were located in sparsely populated areas and were either endorheic or had limited inflow or outflow from rivers, I hypothesized that the primary transport mechanism for MPs to these lakes was the atmosphere. This study identifies the presence of MPs in remote lake sediments that have no local sources and their possible link to storm events while also providing a baseline of MP pollution for future work.

4.1 Introduction

Microplastic (MP) pollution has recently been established as a global threat to natural environments and humans alike (Karbalaei et al., 2018; Horton & Barnes, 2020; Smith et al., 2018; Walker & Fequet, 2023). Studies have shown MPs can negatively affect biota by introducing toxins or leaching hazardous chemicals into an environment or directly into an organism when ingested or inhaled (Cole et al., 2013; Schirinzi et al., 2017; Karbalaei et al., 2019). MPs have now been detected in all types of environments both urban and remote, for example: the middle of the ocean, the tops of mountains, Arctic ice, city air, and even in human placentas (Law et al., 2010; Dris et al., 2015; Allen et al., 2019; Bergmann et al., 2019; Ragusa et al., 2021). Although there has been extensive research in recent years on MPs in marine and urban or riverine freshwater environments (Allen et al., 2022), there have been a paucity of studies focused on rural lakes (Dusaucy et al., 2021), and thus there are still many open questions about the extent of MP pollution in lakes globally and their effects on such freshwater ecosystems.

Previous research has significantly contributed to our comprehension of MP pollution levels in freshwater lakes. However, the majority of these studies have primarily focused on large lakes located near urban centers and connected to major river systems (Felismino et al., 2021; Helm. 2020). These large lakes generally exhibit multiple direct sources of MPs and different processes, such as surface or bottom currents, waves, riverine flow, or human activity, control the import and export of these particles (Helm, 2020). In contrast, there is a noticeable dearth of studies examining MP presence in endorheic lakes, so those with limited inflow and outflow and primarily fed by precipitation and groundwater.

despite the ecological significance of these lakes and their ability to sustain diverse aquatic populations (Wang et al., 2018). These lakes might have prolonged residence times for MP particles, which may impact the aquatic species as they are cut off from other lake systems and the ocean (Wang et al., 2018).

In this study, I investigated MP deposition in small (<10 km^2) remote lakes across the island of Newfoundland, Canada (Figure 4.1). The Province of Newfoundland has a small population (540,000 people; Government of Newfoundland and Labrador, 2023) with only four urban areas and, on average, has a low population density (1.4 person km^{-2}; Statistics Canada, 2021). Additionally, most of the settlements are concentrated around the coastline leaving the interior sparsely populated. The lakes sampled in this study were situated in areas far from major settlements and with no direct sources of MP. Though not all lakes sampled were endorheic, they were fed by very small stream systems meaning there was limited inflow and outflow. This means large amounts of MPs were not able to be transported to these lakes via river or deposited directly from the shores. I therefore hypothesized that most MPs present in these lakes were transported and deposited via the atmosphere and may have been sourced thousands of kilometres away (Ryan et al., 2023). In theory, any atmospheric MPs deposited within the lake's catchment area would also ultimately end up in the lake, transported either by runoff or through groundwater.

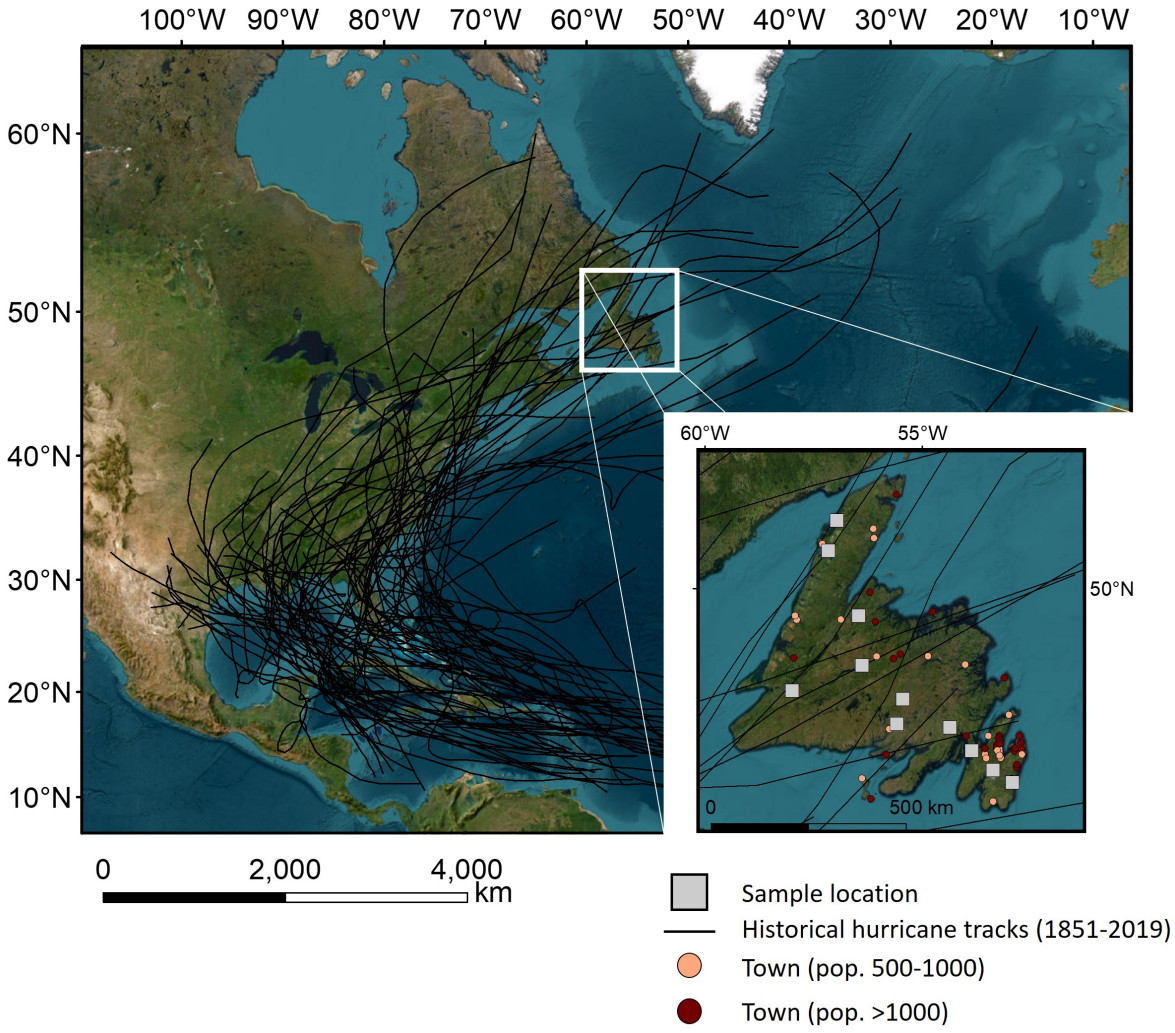


Figure 4.1: Sample locations. Historical hurricane tracks in the North Atlantic from 1851-2019 with location of lakes where sediment samples were collected (grey boxes) and towns with populations greater than 500 (pink circles) or 1000 (red circles) people (inset).

For this study, I collected lake bottom sediment samples from 11 different lakes around the island of Newfoundland. I then analyzed MP amount, size, and polymer type. Chapter 3 of this thesis demonstrated the role of hurricanes in transporting and depositing ocean sourced MP in Newfoundland. If my assumption that MPs in the lakes investigated are primarily sourced by atmospheric fallout, including storm events, I expect to see similarities in the size and polymer types. I will also be determining if there are trends in

the spatial distribution of MP particles that could be attributed to storm systems, such as historical hurricane tracks, or human population patterns. Since there have been no previous studies of this kind in Newfoundland, these results will provide a baseline for MP pollution in remote lakes moving forward.

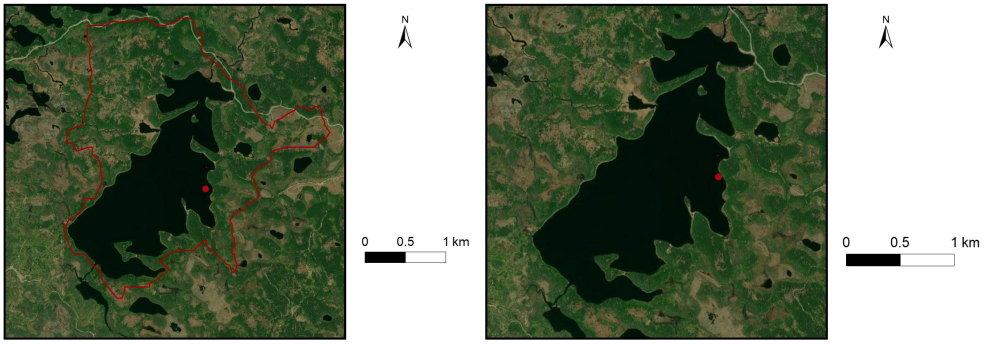
4.2 Methods

4.2.1 Field sampling methods

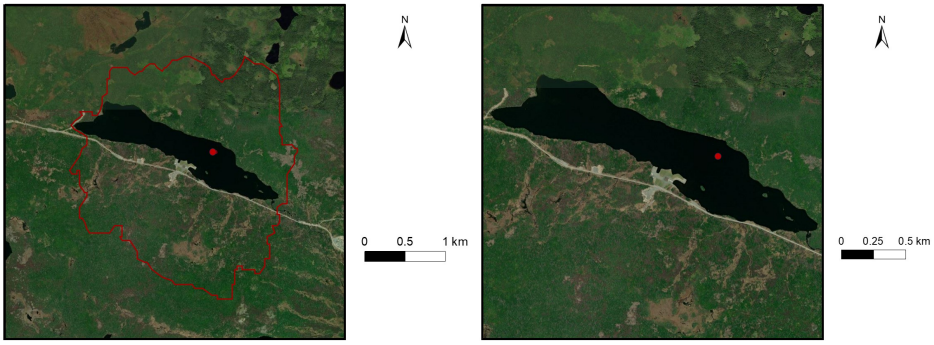
Newfoundland has thousands of freshwater lakes and ponds distributed relatively evenly across the island, many of which are endorheic meaning they are not connected to river systems (Yapiyev et al., 2017) or are only connected with small streams and have very little inflow or outflow, and are primarily fed by precipitation (rain or snow) or groundwater. I collected sediment samples from the bottom of 11 lakes of varying size, depth, and sediment type across Newfoundland (Table 4.1, Figure 4.2), chosen based on remoteness, so preferring lakes with little to no structures or activity surrounding them, but still accessible by road. Samples were collected with a steel Van Veen grab deployed from a dinghy that collected the top 20 cm of sediments from the lakebed. I tried to collect samples in areas with the least amount of vegetation. Most of the lakes had varying sediment type depending on the location within the lake, changing from rocky to sandy to muddy, with organic material clustered in discrete areas of the lake, so sediment type where the samples were taken was not necessarily descriptive of the whole lake. I recorded the coordinates and elevation (metres above sea level) of the spot and approximated depth using a marked rope. Sediment at the top of the grab was scooped into a glass jar with a stainless-steel spoon. Each jar was then sealed, labelled with the

sample number, lake name, and sample coordinates, then wrapped in aluminum foil and paper.

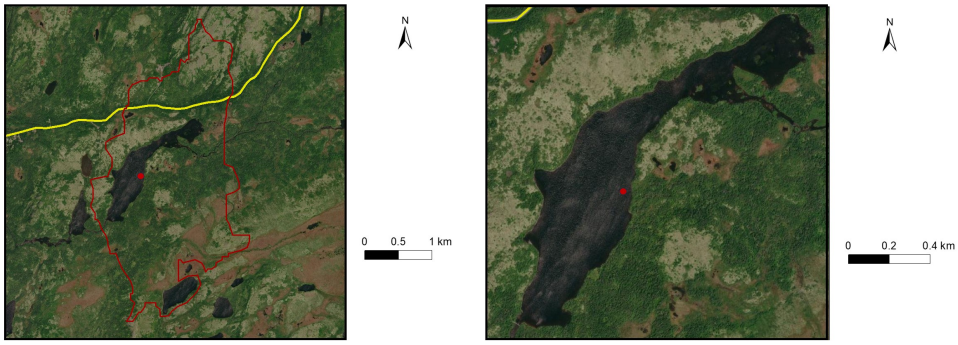
I later determined the catchment area of each lake using the sample coordinates as a drainage point. Catchment areas were delineated in Global Mapper (version 16.0, Blue Marble Geographics) using the Shuttle Radar Topography Mission (SRTM) 1 arc-second digital elevation model available from the United States Geological Survey. Population density for each lake was determined using census subdivision data from the 2021 census (Statistics Canada, 2021). The distance from sample site to the nearest town (for both towns with populations >500 and >1000) was calculated as a straight line from the exact spot each sample was taken to the town centre. Distance to past hurricanes was calculated as a straight line from the sample spot to the closest point of the historical hurricane track. The number of dwellings surrounding the lake was determined by visually inspection of lakeshore and catchment area using satellite imagery. All lakes are assumed to be exposed to similar environmental conditions such as ice cover duration and annual precipitation. Environmental variables were correlated to final MP counts using Pearson's correlation test.



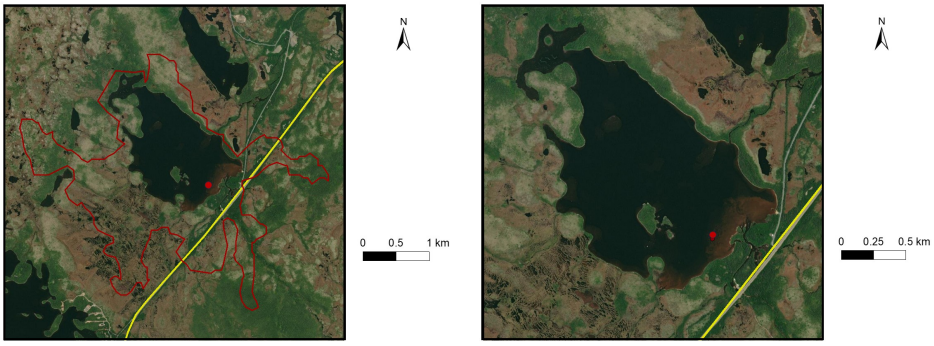
NLS4



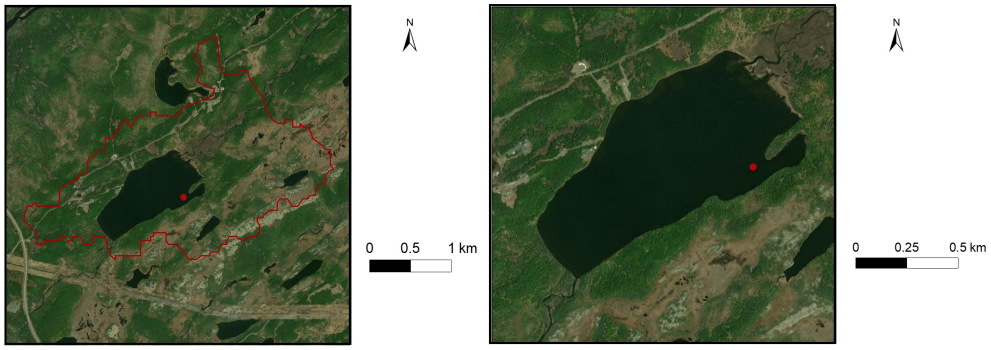
NLS5



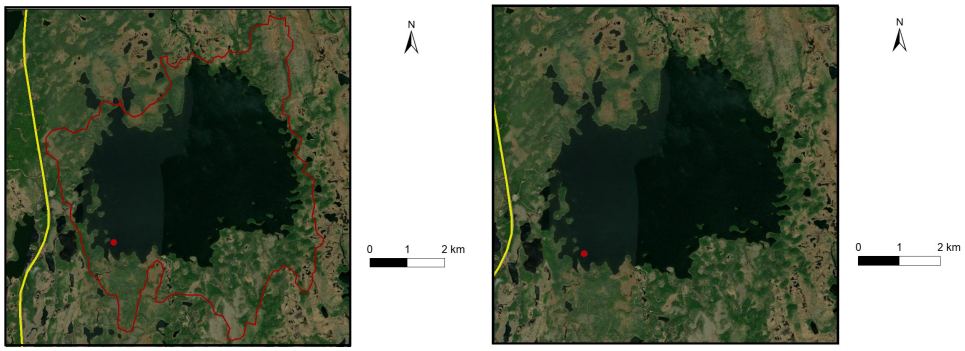
NLS7



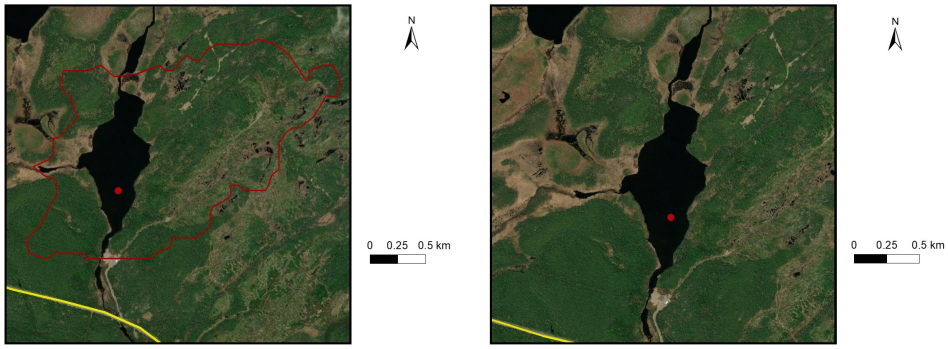
NLS9



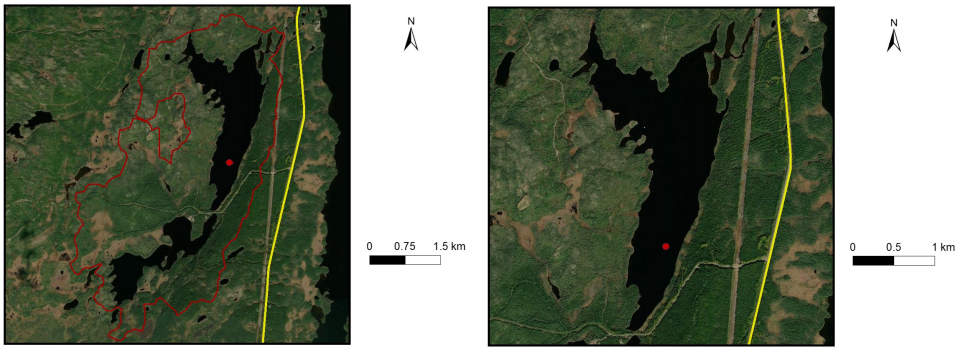
NLS12



NLS14



NLS18



NLS20

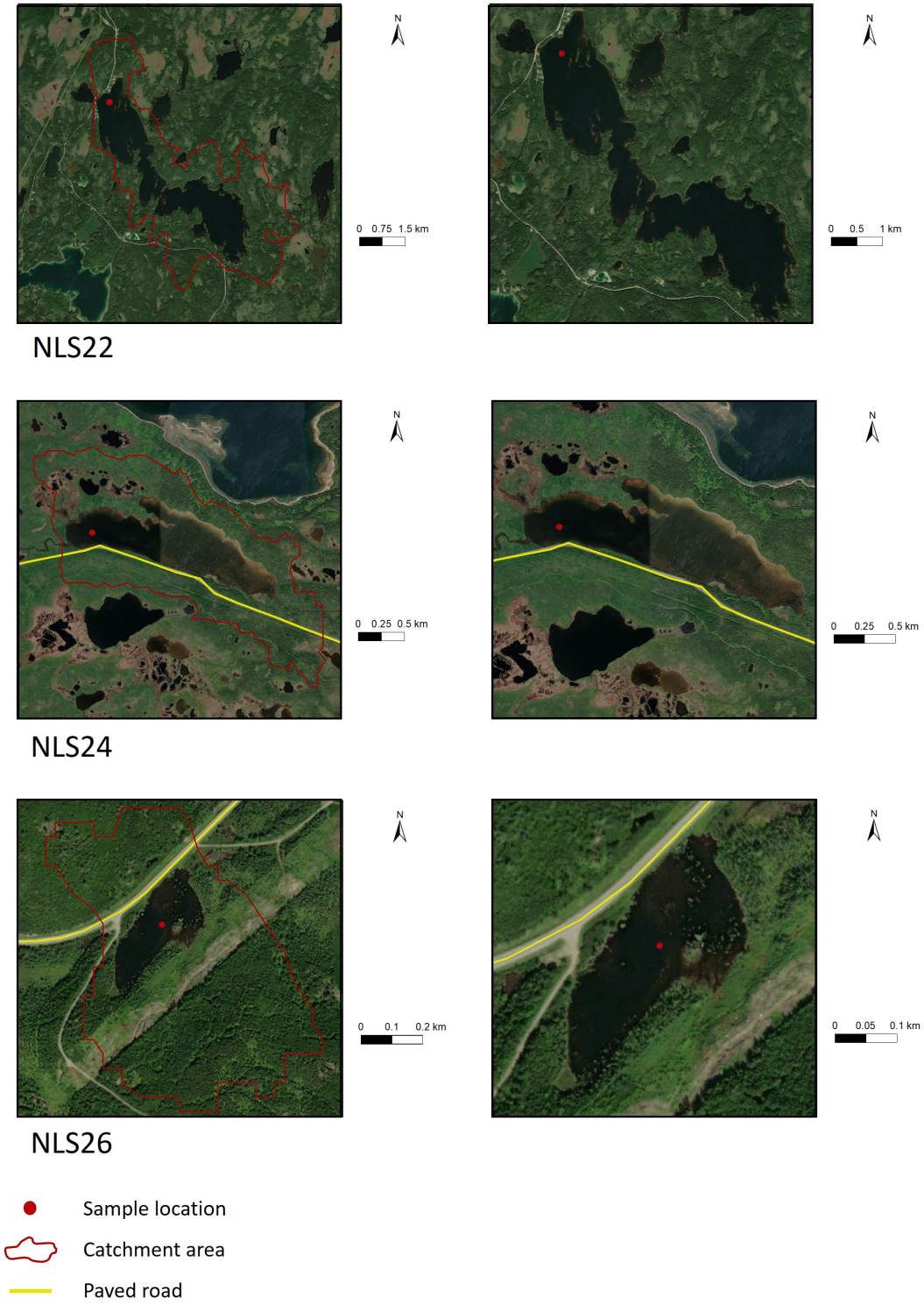


Figure 4.2: Satellite images (Esri, Maxar, Earthstar Geographics, and the GIS User Community) of all lakes sampled with location of sample collection indicated by red dot. The catchment area of each lake is outlined (red line) and paved roads are indicated by yellow lines.

4.2.2 Laboratory sample preparation and analysis

Three subsamples from each sample were analyzed in a dedicated MP analysis laboratory (n = 33). All surfaces were cleaned using Milli-Q and paper towel then covered with aluminum foil before use, while the laboratory underwent thorough cleaning using Milli-Q each week. Samples were kept below 55ºC to avoid damage to plastic particles. Each sample was digested to remove organic material, density separated to remove non-plastic inorganic particles, then filtered on silver membrane filters to be further analyzed. Before extracting MPs from sediment, each jar was thoroughly stirred to homogenize the sediment sample. Approximately 10 g w.w. of sediment was measured into a glass test tube and digested using 40 mL of 30% filtered H_2O_2 for 48 hours at 50ºC (dry block). Samples were then vacuum filtered (1.2 µm pore GF/C 47 mm diameter filters) and density separated using 1.6 g/cm^3 $ZnCl_2$, agitated for a minimum of 48 hours. Particles suspended in the $ZnCl_2$ solution (upper ½ of density separation flask) were filtered onto 25 mm diameter 1.2 µm silver membrane filters and stored in 50 mm diameter sterilized aluminum tins to prevent contamination until further analysis.

One filter for each sample was analyzed using µRaman spectroscopy and all samples were analyzed with Nile red fluorescence (n = 11). µRaman analysis was undertaken using a Renshaw InVia spectrometer at Memorial University of Newfoundland (Canada) with an 830 nm wavelength laser, 122b gr mm^{-1} grating and using up to 25% power (filter). Spectra were collected over 200-3000 cm^{-1} using a minimum of 3 acquisitions of 10 s. Filters were analyzed using the sub-sampling method presented in Huppertsberg and Knepper (2018) to ensure a representative 25% of each filter was analyzed. µRaman spectra for each particle were then individually analyzed using Spectragryth, the SLOPP,

SLOPPE, Simple (Menges, 2020; Munno et al., 2020; Primpke et al., 2020) and inhouse reference libraries to identify if each particle was plastic and the respective polymer type. Only spectra with a 60% or greater accuracy were considered and counted as plastic polymers. A lower accuracy rating was used for these samples compared to the samples collected during Hurricane Larry (Chapter 3 of this thesis) since very few particles had a rating of 80% or higher. This may be due to user error, or because the μRaman used for analyzing these samples had a longer wavelength laser than the instrument used for the Hurricane Larry samples, which may have degraded the match percentage.

After completion of μRaman analysis, all samples underwent fluorescent microscopy for particle size distribution assessment (necessary due to the limited clarity of μRaman visuals and the absence of particle finder software within the μRaman set up) and to determine MP counts (since μRaman results were inconclusive for >50% of samples). Filters were dyed using 0.1 μg mL^{-1} Nile Red solution (powder dissolved in 99% pure ethanol (Sigma Aldrich), prefiltered through 1.2 μm GF/C filter), air dried overnight in a 30°C oven (covered in sterilized foil to minimize sample contamination), and then placed in a dark, 4°C fridge prior to analysis. Fluorescence microscopy was completed using an OMAX microscope and imaged using the AmScope MU500 camera and software, with 5% of the sample assessed and the images analyzed using FIJI (ImageJ) (Schindelin et al., 2012). Particles were classified as fragments or fibres, with fibres defined as particles with at least a 3:1 length to width ratio (Löder & Gerdts, 2015).

4.2.3 Blanks, positive controls, and contamination mitigation

During sample collection, all equipment was cleaned thoroughly with lake water then Milli-Q before use. Plastic equipment during sample collection was avoided, using metal or glass instead including the grab and sample containers. The sample containers were always sealed except when sediment was actively being transferred and were wrapped in aluminum foil to protect from contamination. Sediment transfer was done immediately over the water and down-wind to reduce contamination from the boat. Full process laboratory blanks (n = 3) were created to account for contamination during analysis. Positive controls were created in the form of six spiked blanks with 10 mL of Milli-Q containing ~40 μm polyethylene fragments. Half of the spiked blanks were not processed, while the other half underwent all steps of sample processing to account for sample loss during the extraction process and efficacy of the analysis methods. Sample blank and spike corrections were applied to MP counts. All glassware was cleaned three times with Milli-Q and covered with aluminum foil. The laboratory was outfitted with HEPA filters, had restricted access, and cleaned regularly with Milli-Q. All personnel in the laboratory were required to wear 100% cotton clothing and 100% cotton lab coats during processing. All steps of sample processing post digestion were done under a laminar flow fume hood, while all chemicals were filtered with a 1.2 μm GF/C filter before use.

4.3 Results and discussion

4.3.1 Microplastic abundance in sediments

The largest abundance of MPs was found in sample NLS14, with an average of 24.2 MP g^{-1} wet weight, while the smallest abundance was found in sample NLS24 with an

average of 6.1 MP g^{-1} wet weight (Figure 4.3). The overall average abundance of MPs across all the lakes was approximately 12.4 ± 4.6 MP g^{-1} wet weight (Figure 4.3).

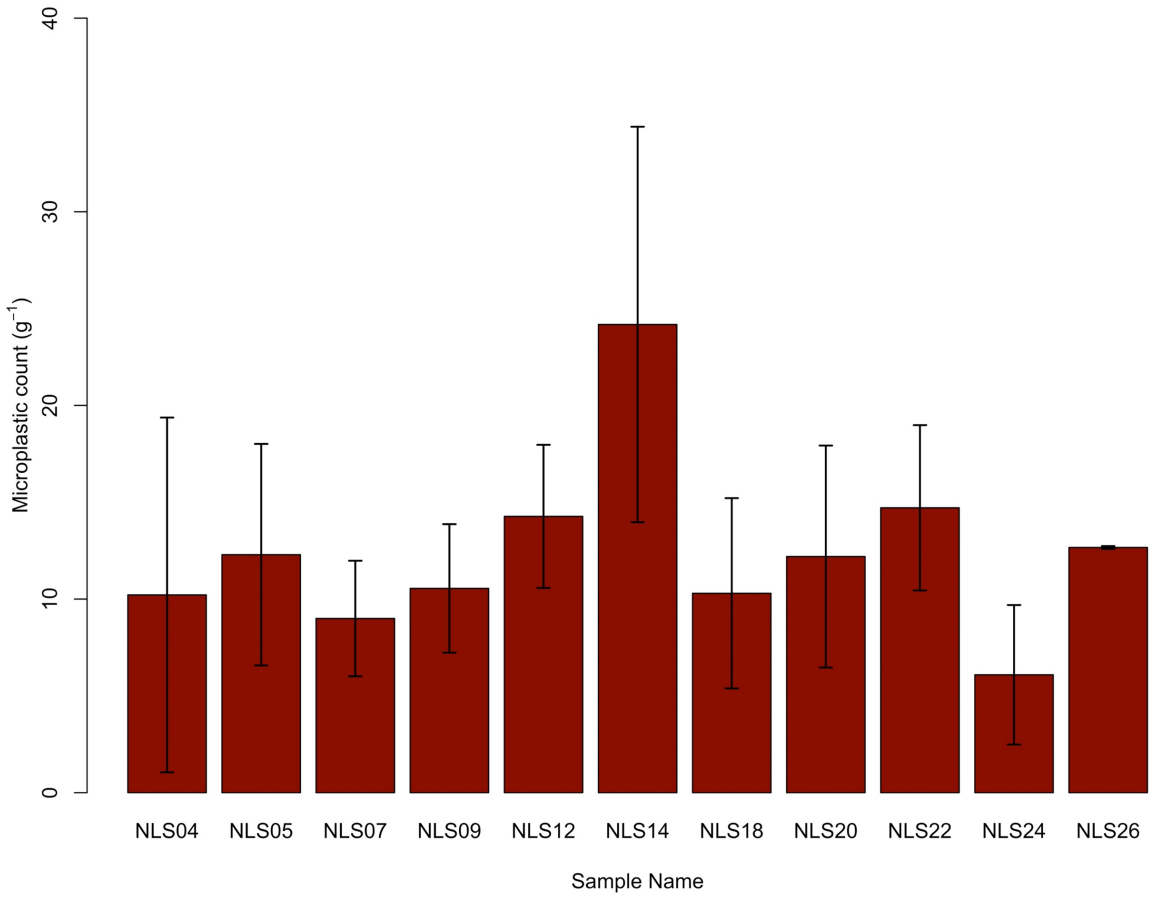


Figure 4.3: MP abundance. Mean microplastic (MP) concentration (with error bars representing 1σ) in each sediment sample (per gram of wet sediment).

The lakes with the highest abundance of MPs (NLS12, NLS14, NLS22) all had a silty sediment type in the area where the sample was collected (Table 4.1). Most of the lakes had a silty or clay sediment type, except NLS7 and NLS18 which are sandy (Table 4.1). Both these samples had approximately 10 MPs g^{-1} w.w., which is close to the overall average. The sample with the highest MP count, NLS14, had silty sediment with some organic material very similar to the sediment found in NLS26. I expected to see more

MPs in lakes with finer sediment (i.e., mud) since clay sticks to MP and the flocculation of clay particles would promote the transport of MPs to the sediments, plus algae or plants growing in the lake could trap MPs and bring them to the sediments as they decomposed. However, there was little variation in MP counts based on sediment size and presence of organic material (Table 4.1), meaning MP deposition was not dependent on sediment type. It is possible that in the muddier samples, some MPs were lost during density separation on account of still being stuck to large clumps of mud. The methodology for processing sediment samples of MP is relatively new and is constantly being refined to be more efficient (Prata et al., 2019).

Since the lake sediment is assumed to be the sink for all atmospheric MPs deposited on the lake surface as well as in the entire drainage basin, I normalized the data by dividing the MP count by lake surface area (Supplementary Figure 1B) as well as drainage basin area (Supplementary Figure 2B). After normalization the greatest MP abundance was 493.9 MP g^{-1} km^{-2} and 32.8 MP g^{-1} km^{-2}, respectively, in sample NLS26. The next greatest MP abundance normalized for area is NLS18 with only 55 MP g^{-1} km^{-2} and 7.1 MP g^{-1} km^{-2}. The anomalously high MP count normalized for area in NLS26 may be due to it being from the smallest lake with the smallest drainage basin, only about a quarter the size of the next smallest drainage basin, or because the lake was located between a paved road and power lines (Figure 4.2), which may have increased the transport of MPs. It is also possible this sample was contaminated, as it had a much lower standard deviation than any other sample.

Table 4.1: Characteristics of each lake sampled and summary of sediment samples with weight of sediment analyzed and average MP count per gram of sediment

Sample name	Latitude (°N)	Longitude (°W)	Elevation (m)	Lake surface area (km^2)	Catchment area (km^2)	Distance from town (pop. >500) (km)	Distance from town (pop. >1000) (km)	Population density (persons km^{-2})	Distance to hurricane track (km)	Number of dwellings around lake	Sediment type	Average wet weight (g)	Average MP count (kg^{-1} w.w.)	Average MP count per lake surface area (kg^{-1} km^{-2} w.w.)	Average MP count per catchment area (kg^{-1} km^{-2} w.w)
NLS04	47°14.113	53°22.276	101	1.0421	5.17	34.4	60.6	6.3	108	0	silty clay	10.67	10,200	9,800	2,000
NLS05	47°02.218	52°55.604	82	0.4286	3.91	10.2	40	28.1	82	1	silty clay	10.33	12,300	28,700	3,100
NLS07	47°32.126	53°52.144	53	0.2531	4.793	35.3	35.3	0.8	46	1	silty sand with organics	10.67	9,000	35,600	1,900
NLS09	47°53.557	54°21.524	175	0.8461	7.965	47.2	47.2	0.5	24	2	silty clay	10.33	10,600	12,500	1,300
NLS12	47°57.076	55°35.347	147	0.3146	4.596	23.2	83.7	0	48	4	silty clay	11	14,300	45,400	3,100
NLS14	48°19.560	55°27.327	223	9.1866	16.314	83.5	106.4	0.1	40	4	silt with organics	10.67	24,200	2,600	1,500
NLS18	48°50.600	56°23.173	182	0.1872	1.453	44.4	83.4	0	0	1	sandy silt	10	10,300	55,000	7,100
NLS20	49°35.378	56°27.993	217	1.1198	12.917	45.6	46.3	0.2	25	0	clay	10.33	12,200	10,900	900
NLS22	50°33.229	57°09.925	50	3.3950	9.436	21.6	152	0.1	0	10	silty clay with organics	10.67	14,700	4,300	1,600
NLS24	50°59.420	56°57.690	7	0.4104	1.499	70.9	166.4	1.2	10	0	clay	11.33	6,100	14,800	4,100
NLS26	48°27.744	57°59.425	139	0.0256	0.3857	82.9	82.9	0.2	13	0	silt with organics	10.5	12,700	493,900	32,800

4.3.3 Size distribution

The particle sizes of MPs investigated had a skewed distribution. Previous studies in remote or urban lakes only measured larger (>50 μm) MP particle sizes (Turner et al., 2019; Dong et al., 2020; Negrete Velasco et al., 2020; Liang et al., 2022), while I analyzed MP particles down to <2 μm. Most MPs identified in these samples were between 2-10 μm (Figure 4.4), with an average of 53% of particles in each sample falling into this size class. The next highest proportion was the 10-20 μm range, with an average of 18% of particles, followed by the 20-30 μm range with an average of 9%. In a previous study investigating MPs in atmospheric samples collected during a hurricane, 65% of particles were between 2-10 μm, 34% were <2 μm, and only 1% were >10 μm in size (Ryan et al., 2023). The size distribution of MPs found in these sediment samples had more particles in the larger size ranges (Figure 4.5). One possible explanation for this is that the collector vessel used for atmospheric samples creates a deposition shadow and preferentially collects smaller particles with increasing wind speed (Goossens, 2010). In lake sediments, there is no deposition shadow and therefore no preference for smaller particle sizes.

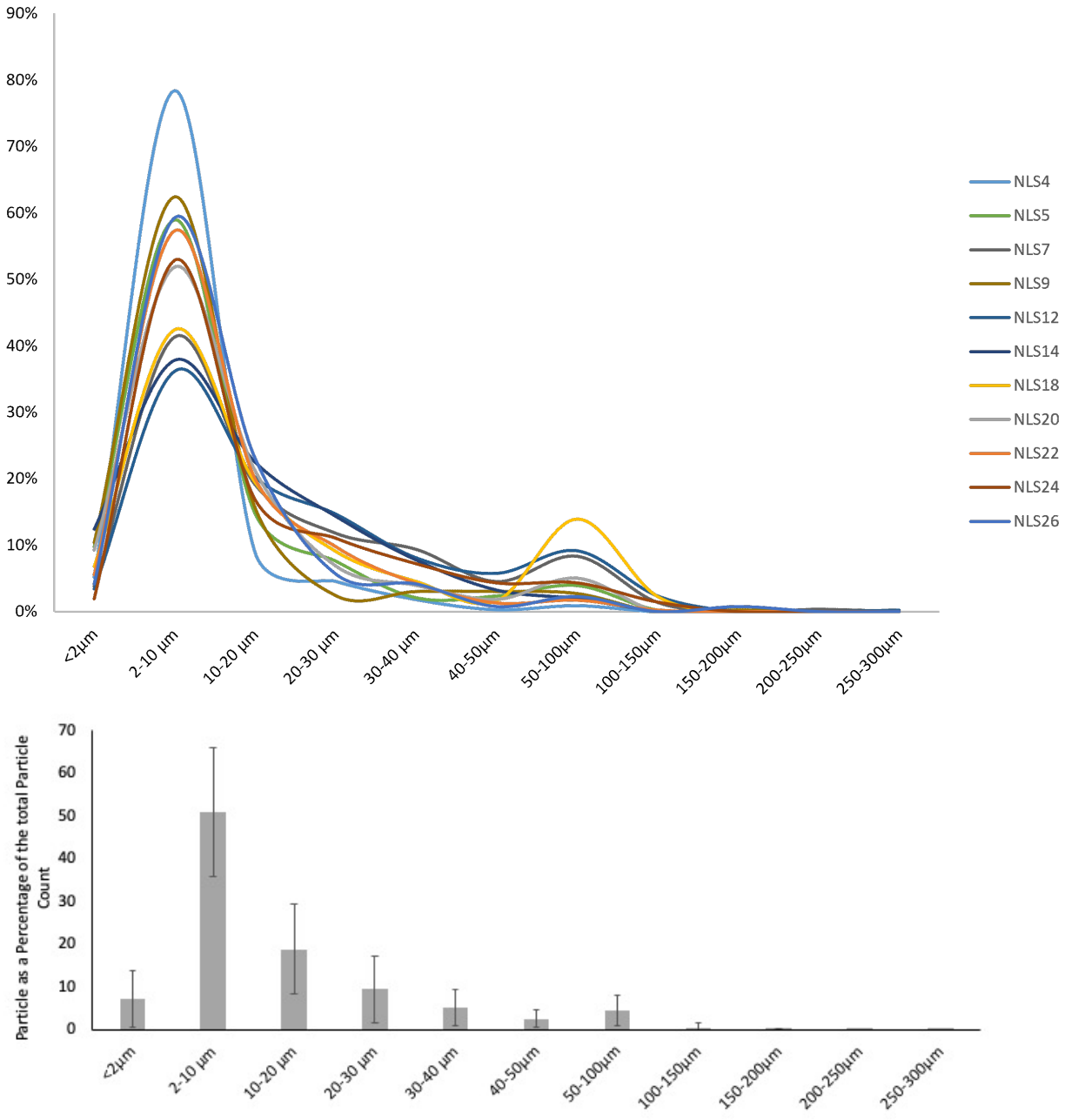


Figure 4.4: MP size. Average size distribution for microplastic (MP) particles found in each sediment sample, by sample (above) and average proportion of MPs by size class for all sediment samples with error bars representing 1σ (below).

The largest MP particle found was between 250-300 µm, in sample NLS12. This sample had the lowest proportion of particles between 2-10 µm at 36%, as well as the second lowest proportion of particles <2 µm at 3.4%. Particles between 50-100 µm were consistently found in all samples (Supplementary Figure 3B), while particles between

100-150 µm were consistently found in over half the samples (with none recorded in NLS4, NLS9, NLS20, and NLS26). The largest particle found in the atmospheric samples collected during Hurricane Larry was between 50-100 µm (Ryan et al., 2023). Particles of the size ranges found in the sediment samples can be transported in the atmosphere for upwards of two days, especially plastic particles which often have longer atmospheric lifetimes than mineral dust due to their low density, shape, and interactions with air molecules (Evangeliou et al., 2022). Though the sizes of MPs found in sediment samples spanned a wider range than those collected during Hurricane Larry, it is likely the smaller sized MPs (i.e. <10 µm) were transported and deposited via the atmosphere (Ryan et al., 2023). Additionally, previous studies investigating MPs in sediments from rivers, the ocean, or urban lakes have mostly looked at larger size classes of >50 µm (; Castañeda et al., 2014; ; Felismino et al., 2021; Turner et al., 2019; Dong et al., 2020; Liboiron et al., 2020) and have found an abundance of MPs. These water bodies have many land inputs of MPs including direct input (e.g. boating or marine activities), wastewater effluent, or runoff. Considering there were very few MPs above 50 µm in size found in this study, it is likely the MPs in the lakes I sampled had a different source.

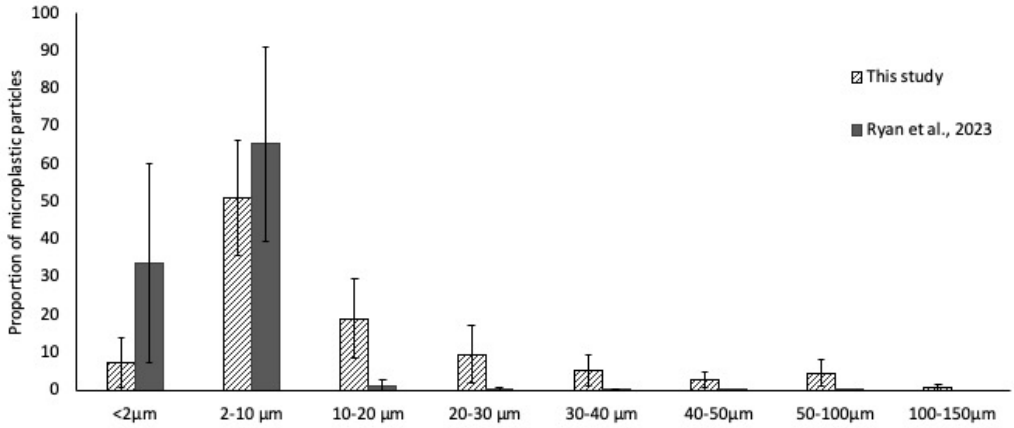


Figure 4.5: Comparison of microplastic particle size distribution for particles detected in lake bottom sediment samples (this study) and particles detected in atmospheric samples during Hurricane Larry (Ryan et al., 2023).

4.3.4 Polymer classification

The most abundant polymers identified were polyvinyl chloride (PVC) and polyurethane (PU). Polystyrene (PS) was detected in three samples and polyethylene (PE) was detected in two samples. Samples for only four lakes out of eleven had over a 60% rating for polymer identification on at least 30% of particles analyzed via μRaman spectroscopy: NLS12, NLS14, NLS20, and NLS26. It is possible there was contamination or user error while analyzing the particles, as plastic was identified in all samples during Nile red fluorescence. There were only two samples with more than three different types of polymers identified, NLS12 and NLS26 (Figure 4.6). In NLS12, the most abundant polymer is PU while in all other samples with identified polymers the most abundant polymer is PVC. The higher occurrence of PVC may be due to its higher density (average density of 1.3 – 1.7 g/cm^3), making it easier for this polymer to sink into the sediments rather than remain floating in the surface waters of the sampled lakes (Andrady, 2011). I was unable to measure lake depth precisely, which may be a factor in the export of MPs from surface waters to the sediments as it is harder and takes longer for MPs to reach the sediments with increasing depth (Liu et al., 2022). Additionally, larger and deeper lakes are likely to have stronger currents which would affect the distribution of MPs within the lake (Bengtsson, 2012). However, polymers, including PS which has an average density of 0.96-1.05 g/cm^3 (Wypych, 2022), were identified in NLS14 which came from the largest and deepest lake (Table 4.1). There were 8 different polymers identified in NLS26 which is the smallest lake by surface area and has the smallest catchment basin area, as well as the greatest MP count per area (Table 4.1). It is possible that due to its small size, the MPs were more concentrated in this lake, plus it experiences less wave action which

restricts the resuspension of particles into the atmosphere. Also, it was a shallow and relatively uniform lake so particles may have been more easily exported to the sediments.

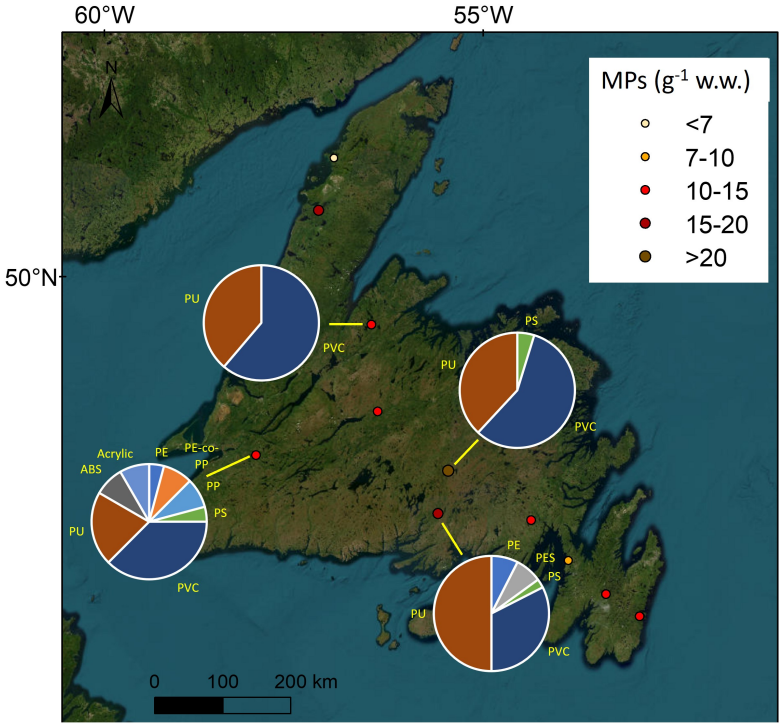


Figure 4.6: Polymer type of deposited particles. Average proportion of each polymer type of microplastic particles found in samples NLS12, NLS14, NLS20, and NLS26 (the only samples in which polymer type could be determined via µRaman analysis). Sample locations for all lakes are indicated, with the size and colour of the dots representing the abundance of MPs identified in each sample using Nile red fluorescence.

While PVC and PU were the most commonly identified polymers, they only accounted for 2% and 1% of total MPs analyzed in atmospheric samples during Hurricane Larry, respectively (Ryan et al., 2023; Figure 4.7). The most abundant polymer identified in samples from Hurricane Larry, polymethyl methacrylate (PMMA), was not definitively detected in any of the sediment samples from this study. Both PVC and PU have higher densities on average than PMMA (Wypych, 2022), meaning they may be more likely to settle to the sediments than lower density polymers such as PMMA, polyester (PES), or acrylic. Both PVC and PU were present in samples collected during Hurricane Larry,

meaning they are able to be transported and deposited via the atmosphere (Ryan et al., 2023).

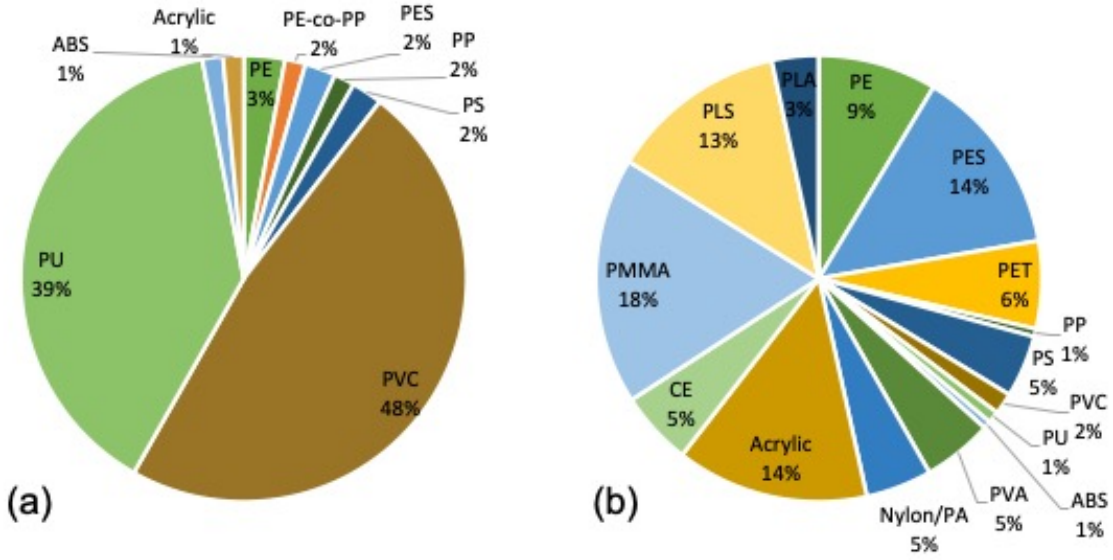


Figure 4.7: Comparison of overall average polymer type distribution for (a) MPs detected in sediment from samples NLS12, NLS14, NLS20, and NLS26 (this study), and (b) MPs detected in atmospheric samples during Hurricane Larry (Ryan et al., 2023).

4.3.5 Spatial distribution of MPs

There is no spatial trend in both the abundance of MPs (Figure 4.8) and the MP count normalized for lake surface area (Supplementary Figure 4B) or catchment area (Supplementary Figure 5B). There was a positive correlation between the amount of MP deposited and the surface area of the corresponding lake ($r = 0.88$, $p < 0.05$), and a positive correlation between catchment basin area and MP abundance ($r = 0.71$, $p = 0.01$; Figure 4.9). This supports the hypothesis that MPs deposited within the catchment area can accumulate in the lake and eventually the lake bottom sediments. There was no correlation, however, between the amount of MP and population density, between MP and distance to the closest town (both population >1000 or >500), or between MP and the number of dwellings surrounding the lake (Figure 4.9), suggesting the amount of MPs in

the sediment is not related to the surrounding human population and human activity in the surrounding area of the lakes was not an important source of MPs. There was also no correlation between MP amounts and distance to the closest hurricane track (Figure 4.9); however, hurricanes affect a much broader area than the track of the eye, plus Newfoundland experiences many other storms in addition to hurricanes, so this is not an accurate measurement of how much each lake is affected by extreme meteorological events.

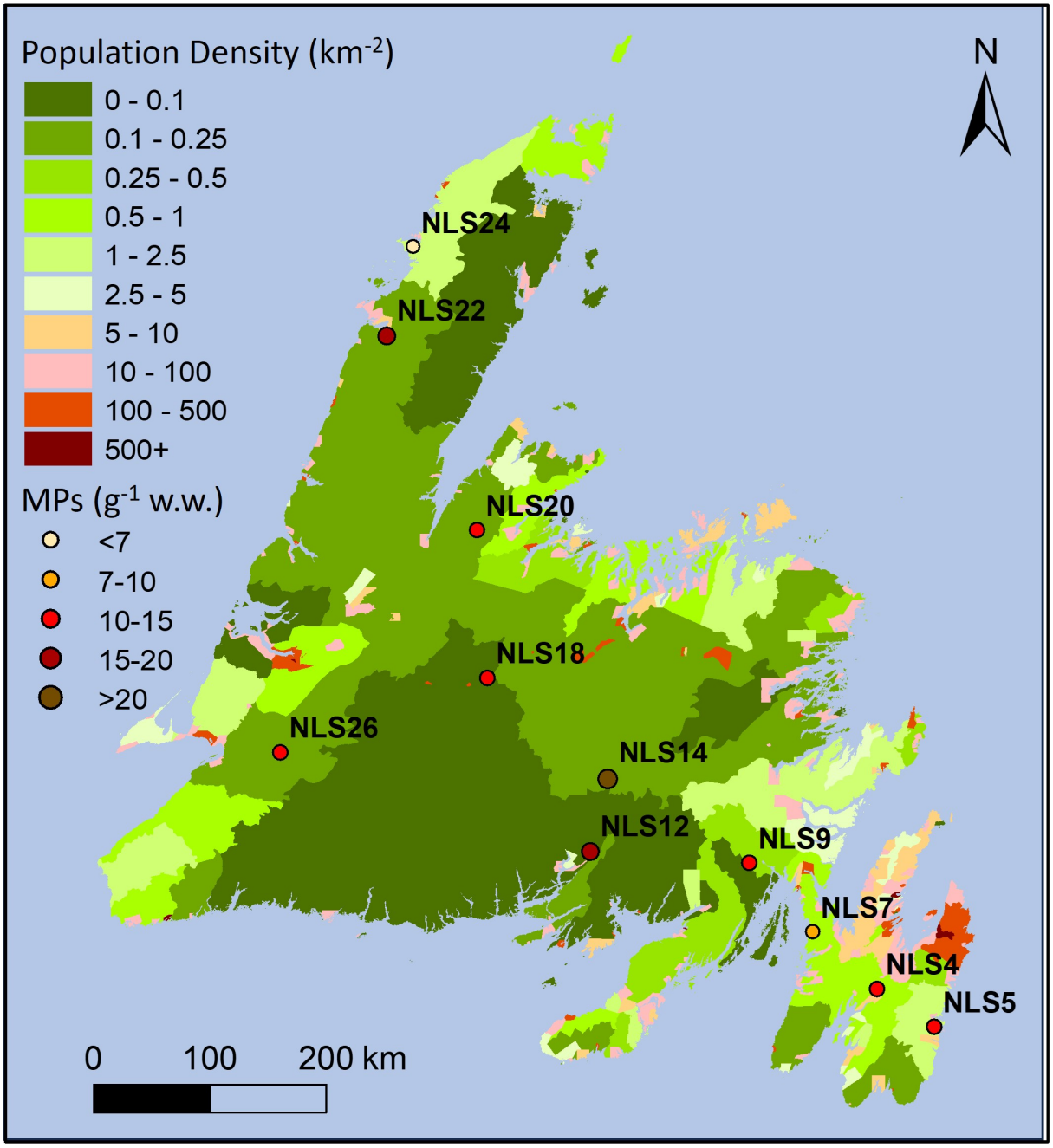


Figure 4.8: Population density. Map of population density for the island of Newfoundland by dissemination area (Statistics Canada, 2021) with microplastic abundances for each sample location indicated by size and colour of dot at the sample location.

There were also no spatial trends in particle size distribution. MP size had no correlation to population density, distance to the nearest town, number of dwellings surrounding the

lake, or distance to the nearest hurricane track (Supplementary Figure 6B). Since particle polymer types were able to be identified in only four samples, I was not able to determine any spatial trends in polymer type nor were there any correlations between polymer type distributions and other variables.

Lakes where samples NLS7, NLS9, NLS14, NLS18, NLS20, NLS24, and NLS26 were collected were all either adjacent to paved roads or had paved roads running through the catchment area (Figure 4.2). NLS14 had the highest MP count while NLS24 had the lowest (Figure 4.3), with the other samples showing comparable MP counts to the lakes with no paved roads in the catchment area. Since I was unable to measure tire wear particles (TWPs), I used road surface material as a proxy for volume of vehicles that use it assuming paved roads are used more often than unpaved roads. There is no evidence therefore to say that higher traffic roads contribute to MP abundance in lake sediments.

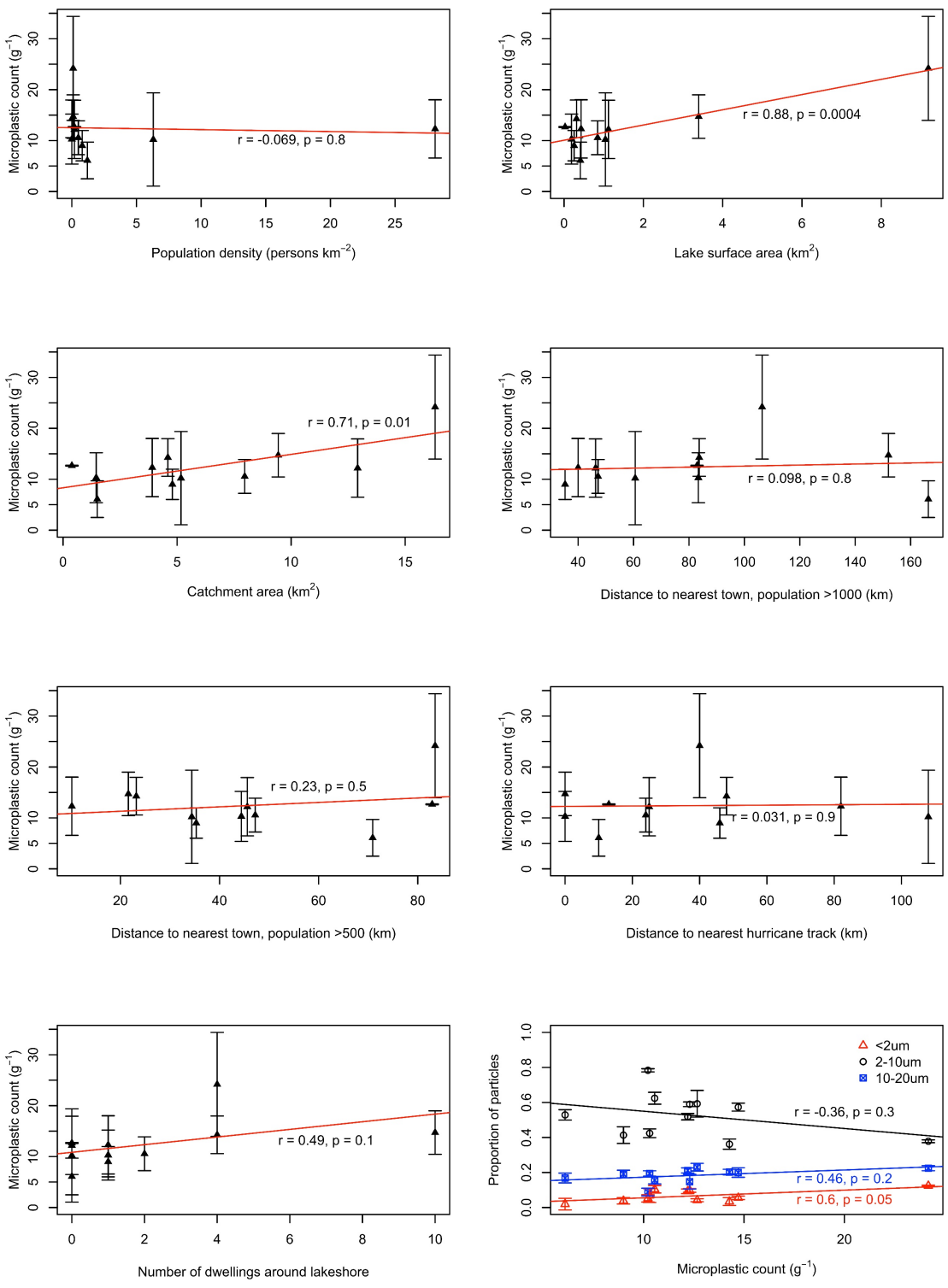


Figure 4.9: Correlation plots. Scatter plots for different lake characteristics and characteristics of deposited particles versus average microplastic count showing possible correlations between variables (with error bars representing 1σ).

4.3.6 Source of MPs

I was able to identify polymer types in four of the samples and therefore determine possible sources for the MPs in these samples. The most abundant polymer identified, PVC, has many different uses including building materials, packaging, and recreational equipment (British Plastics Federation, 2024). Many of the lakes sampled, including all four samples with identified polymers via μRaman spectroscopy, had dwellings or ATV trails located on the lakeshore or within the catchment basin (Figure 4.2), meaning a portion of the MPs found in these samples may have been locally sourced from the building materials used to make these dwellings or from the ATVs and other recreational equipment. PU also has a wide variety of uses including athletic footwear, though is more commonly used for insulation and protective coatings or adhesives (American Chemistry Council, 2024; Polyurethanes, 2024). These possible sources, however, do not account for the amount of MPs found in these samples as there is not an increase in either polymer in areas with higher population densities and dwelling counts (Figures 4.6 and 4.9). For example, there is a higher proportion of PVC in NLS14 than NLS12 despite both lakes having the same number of dwellings and population densities of <0.2 persons km^{-1} (Table 4.1). There were also two additional polymer types identified in NLS12 (PE and PES). If all MPs found in the samples were due to local sources, it is likely these two samples would differ less in polymer type distribution. Additionally, NLS26 had the smallest catchment basin and no dwellings, but had the highest number of polymer types identified. There may be some contamination and local input from the nearby ATV trails, however, there are not enough activities or sources within the catchment area to explain all the polymers identified within the sample. Though I cannot directly link the MPs

identified to a specific source, it is likely that they came from outside the catchment basins of the lakes sampled. There may also be input from active or historic mining activities, though the lakes sampled had no visible signs of industry activity within the catchment areas.

4.3.7 Transport mechanisms for MPs

I infer that atmospheric transport and deposition is the most likely mechanism for delivering MPs to lake bottom sediments, either via direct deposition onto the lake surface or via runoff and groundwater flow from deposition within the catchment area, since there is very little flow into the lakes investigated via rivers and there are few direct sources that could be inputting MPs directly into the lakes. Additionally, the size distribution of the MPs found in the lake bottom sediments is consistent with those found in other studies investigating atmospheric deposition (Allen et al., 2019; Ambrosini et al., 2019; Brahney et al., 2020; Abbasi et al., 2022; Ryan et al., 2023). The polymer types found in the study differed from those found in previous studies of atmospheric MP deposition and from the most abundant polymer types in the marine environment (Santana-Viera et al., 2021), which is a source for atmospheric MPs (Brahney et al., 2021). This could be because this study investigated MPs stored in sediments rather than MPs at the lake surface. Since most polymers are less dense than freshwater, they do not readily sink to the lake bottom leaving them in the surface waters and making them available to be potentially resuspended into the atmosphere via hydrodynamic processes such as bursting bubbles, breaking waves, or falling raindrops (Allan et al., 2020; Lehmann et al., 2021; Ferrero et al., 2022). However, there is less wave action and bubble

injection in lakes than the ocean, especially the smaller lakes sampled that were relatively sheltered, which would decrease the amount of MPs being resuspended into the atmosphere from the lake surface (Allan et al., 2020).

4.3.8 Limitations and future work

This study was done as the first of its kind in Newfoundland, presenting evidence of MPs in remote lake bottom sediments that were not locally sourced and were likely transported and deposited via the atmosphere. Due to budget and personnel restrictions, as data collection took place during the Covid-19 pandemic, I was limited in my sampling. I was only able to sample lakes accessible by road so many of them did have structures on the shores or were used regularly for recreational activities. Ideally sediment would be collected from lakes with no human activity in the drainage basin. I would also like to sample several more lakes from around Newfoundland as well as Labrador, which is even more remote, to create a more robust data set and be able to do more in-depth statistical analyses. I was also limited in the μRaman analysis and therefore was not able to give definitive conclusions on polymer type and sources. In future studies, I recommend performing a polymer type analysis as well as investigating tire wear particles to determine if there is local contamination from nearby roads and recreational vehicles.

Future work may also include investigating MP pollution in groundwater, as many remote communities rely on groundwater wells as their primary drinking source. I would also like to look at sediment cores taken from lakes around Newfoundland to see if there are trends in MP deposition over time, as well as collect sediment cores from a lake with

varves, pairs of thin silt and clay layers that represent a single year of deposition in a lake, to accurately date MP deposition. MPs deposited in known years can then be connected to major storms passing over that area to see if there is a relationship between deposition amounts and storm events. I recommend long-term monitoring of MPs in both surface waters and lake sediments to investigate MP deposition with future storm events and compare deposition amounts or possible sources with historical data.

Author contributions: A.C.R. conceived the study, collected and analyzed samples, wrote the manuscript, and drafted the figures. V.M. conceived the study, contributed to the drafting of the manuscript and figures. T.R.W. contributed to the editing of the manuscript. N.E.K. contributed to the editing of the manuscript. E.F.M.S. supported data analysis.

CHAPTER 5 CONCLUSION

Microplastic pollution is a global issue that is predicted to increase in severity in the upcoming years, as more plastic is produced and released into the environment every year. The study of atmospheric transport and deposition of MPs to remote areas without local sources highlights the pervasive nature of MP pollution and the complex processes that influence global MP distribution. I have identified, for the first time, the presence of MPs in a North Atlantic hurricane and the importance of major atmospheric events as a transport mechanism for MPs. Not only was the abundance of MP fallout from the atmosphere during this hurricane an unprecedented amount compared to previous studies, potentially reaching a maximum of over 100,000 particles m^{-2} d^{-1}, but polymer type analysis and back-trajectory modelling evidenced the possibility these MPs were mainly sourced from the ocean. This study provided further evidence that the ocean can be a source for MPs, as particles in surface waters can be resuspended into the atmosphere and transported far distances in a relatively short amount of time. High winds during a hurricane only increase the distance these particles can travel and broaden the area of potential entrainment points, increasing contamination to ecosystems that would otherwise be considered pristine and isolated from anthropogenic pollution.

MPs were also identified for the first time in lake-bottom sediments from relatively small, remote lakes around the island of Newfoundland. A maximum of 24 MPs g^{-1} w.w. was identified. The MPs in the sediment samples had a similar size distribution to MPs in the atmospheric samples collected during Hurricane Larry, suggesting the MPs found in the sediments were likely deposited from the atmosphere. Although some of the lakes were

used for recreational activities or had dwellings within the catchment area, local input of MPs can be considered negligible. This study provided evidence that atmospheric MPs can be deposited and accumulate in remote freshwater environments with limited local sources of contamination, posing a potential risk to the ecosystem. The detection, quantification, and characterization of plastics in the environment is one of the priorities in Canada's Plastics Science Agenda (CaPSA), which outlines how Canada can move towards a circular economy in regard to plastic (Environment and Climate Change Canada, 2019).

The results of this project are intended to act as a baseline for future studies, with more robust research needed to make definitive conclusions about the effect of extreme weather events on atmospheric MPs in this region. The island of Newfoundland presents an opportune location to monitor atmospheric MP deposition for the North Atlantic region to create a long-term record. The more information we have on this topic, the better we understand how MPs move through the atmosphere and how that movement is influenced by different factors. This in turn helps policy makers improve plastic reduction policies and gives as indication of where to focus mitigation and management efforts.

It is difficult, if not impossible, to remove MPs once they are in the environment so the best way to manage this problem is to limit the amount of plastic pollution being released. Recent developments include a microfibre filter attachment for washing machines that catches MP fibres in washing machine water and prevents them from being released into effluent (Erdle et al., 2021). However, the most effective form of plastic

waste management is to reduce the amount of plastic produced in the first place. Several countries have already banned manufacturing microbeads, and many countries, including Canada, have proposed or passed regulations banning single-use plastic (Environment and Climate Change Canada, 2023). The G7 Ocean Plastics Charter aims to bring industry and government together towards increasing recycling, reuse, and recovery of plastics (Environment and Climate Change Canada, 2021), while the UN's proposed Global Plastics Treaty aims to end plastic pollution through a circular economy (Plastics Europe, 2023). Consumers can contribute by opting for products made with alternative materials, buying items second-hand, and reusing existing plastic items instead of throwing them out. Though the main goal of this project was to better understand atmospheric microplastic transport and deposition, these results will hopefully spread awareness about the sheer abundance of microplastics and the direness of this situation.

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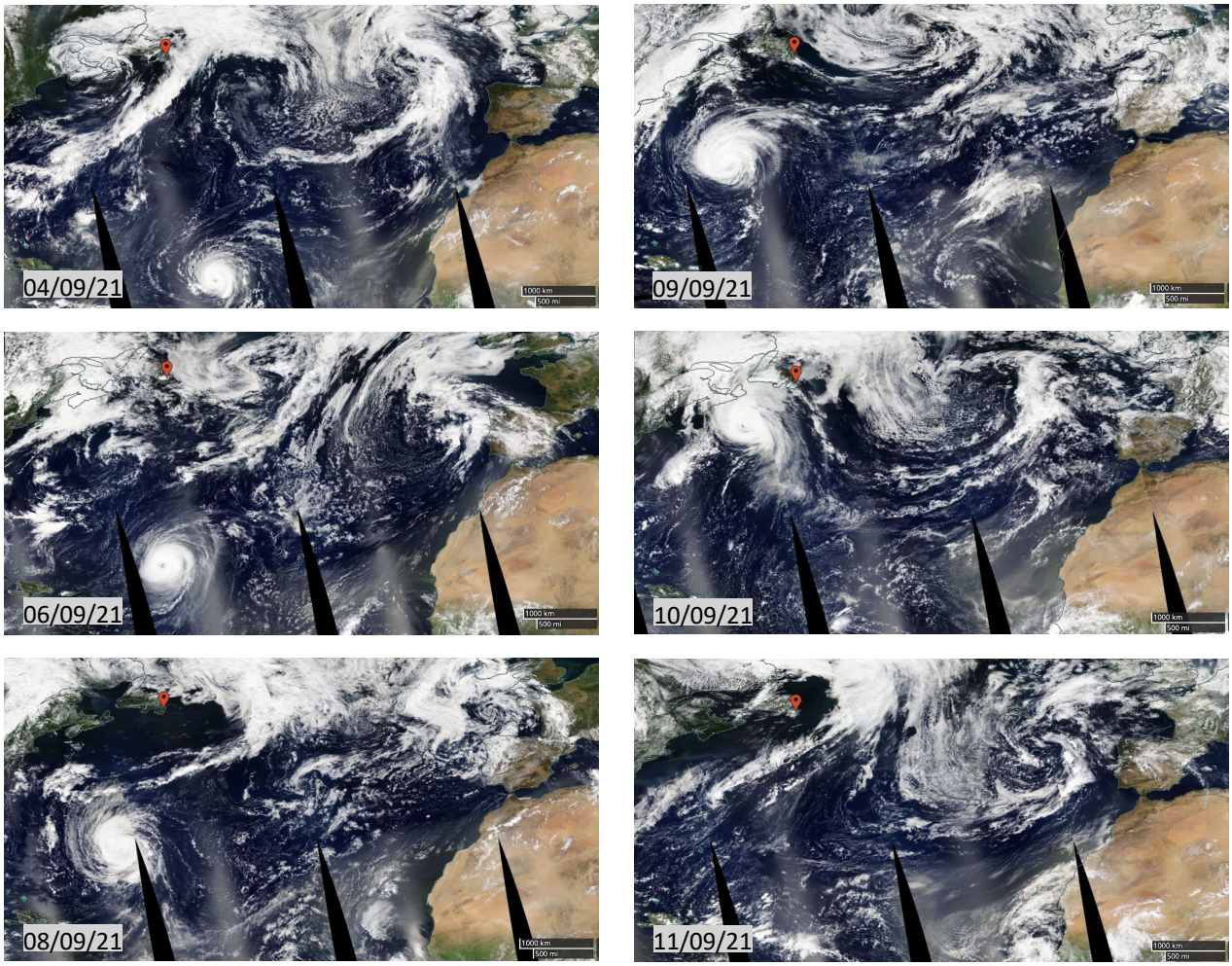
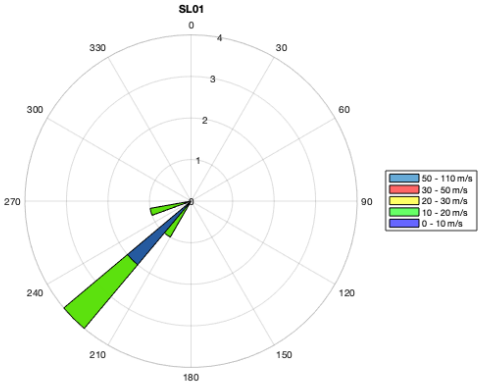
APPENDICES

APPENDIX A: Supplementary material for "Transport and deposition of ocean-sourced microplastic particles by a North Atlantic hurricane"

Supplementary Note 1A – Meteorological Conditions



Supplementary Figure 1A: Satellite view of Hurricane Larry track during sample period taken from EOSDIS Worldview (NOAA-20/VIIRS, https://worldview.earthdata.nasa.gov/) with sample site indicated by red marker.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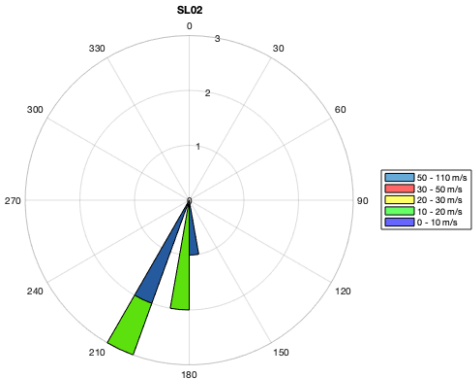
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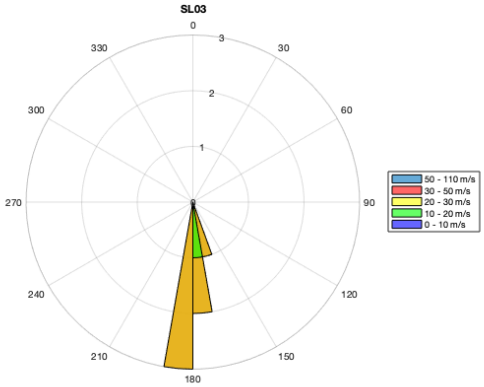
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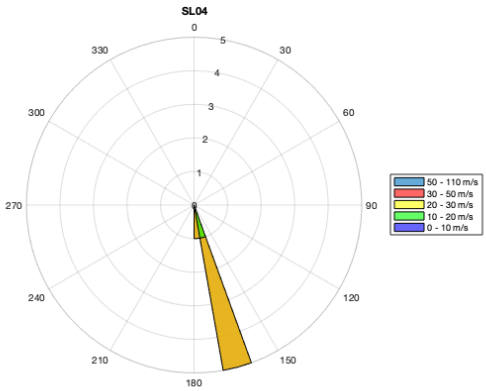
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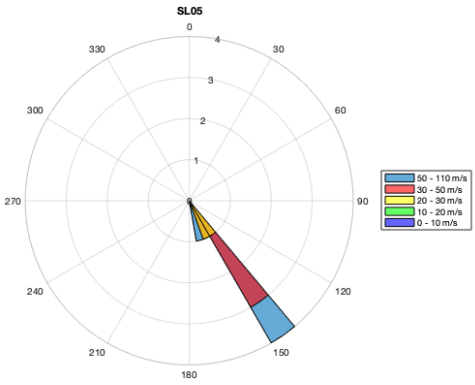
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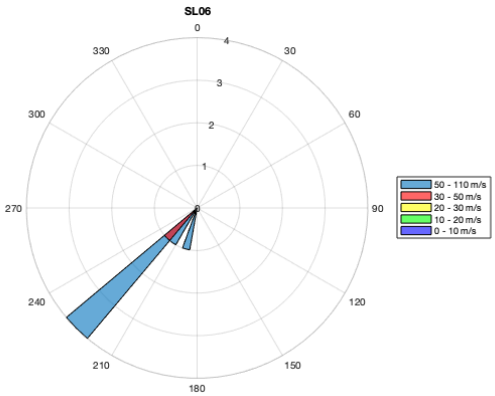
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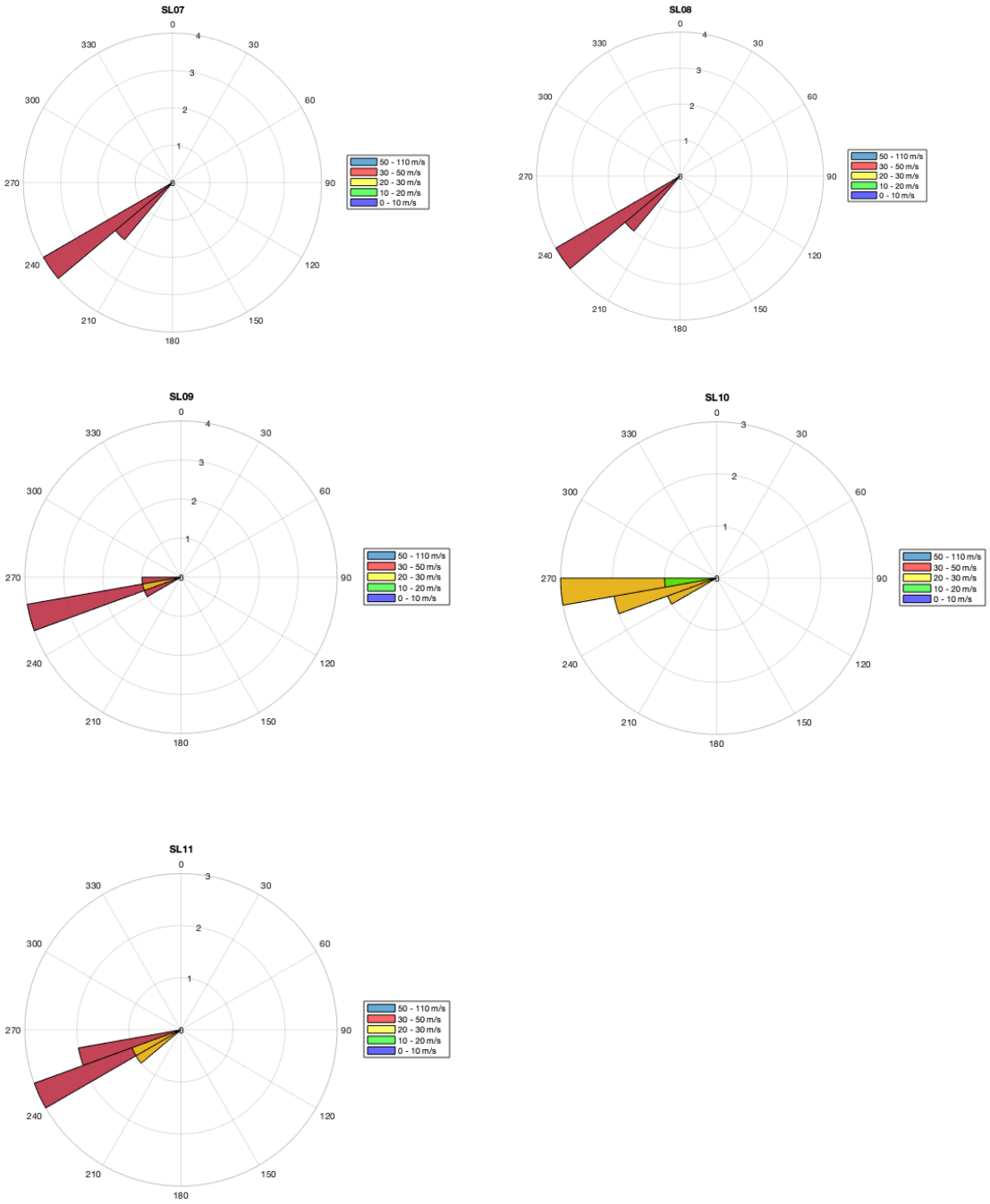
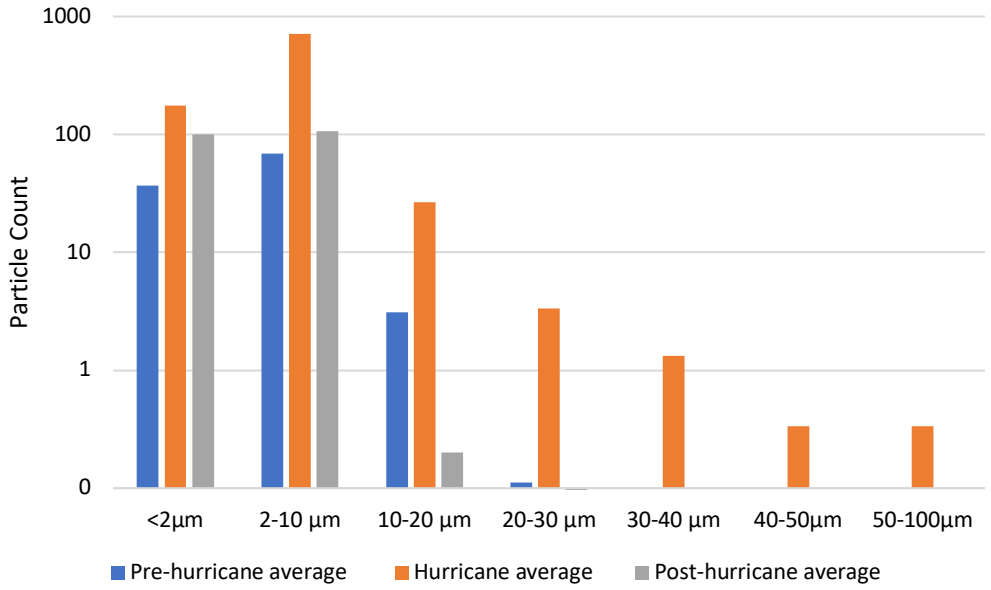
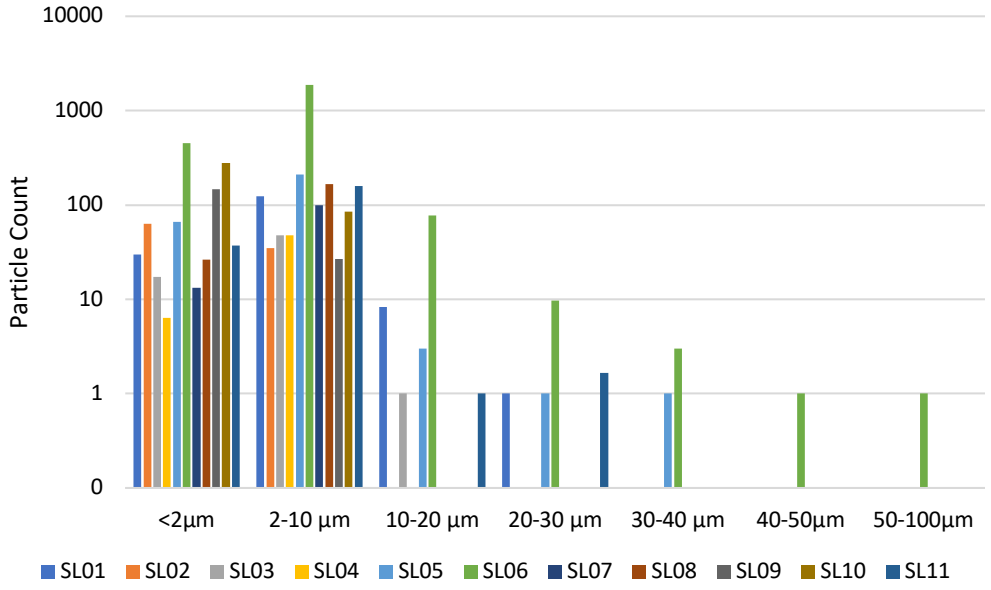


Supplementary Figure 2A: Wind rose diagrams showing recorded wind speed and direction at the Cape Race weather station (Newfoundland) during the collection of each sample.

Supplementary Note 2A – Size Distribution

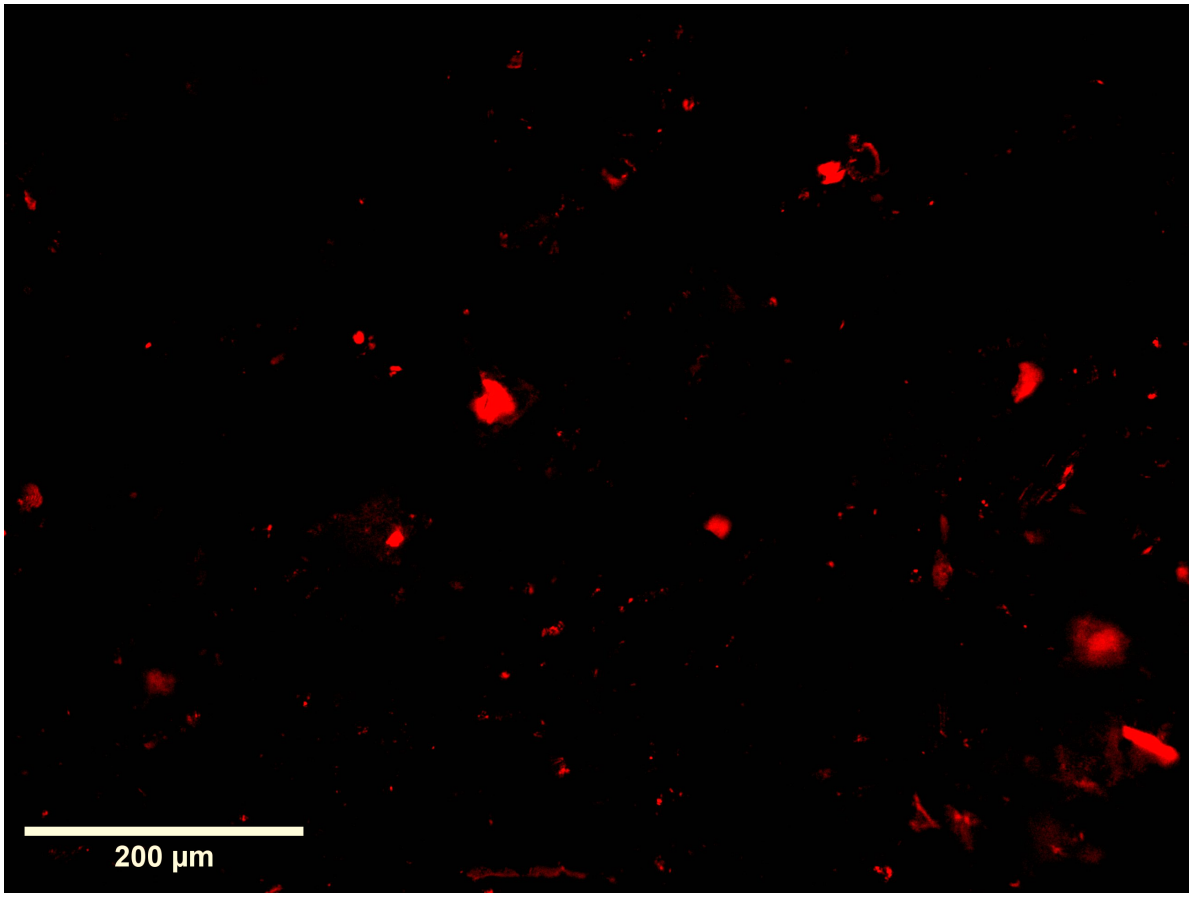


Supplementary Figure 3A: Number of microplastic (MP) particles (n MP per 5% of sample filter) for each size class calculated by averaging values for samples collected before (SL01 – SL03), during (SL04 – SL06), and after (SL07 – SL11) the passage of Hurricane Larry.



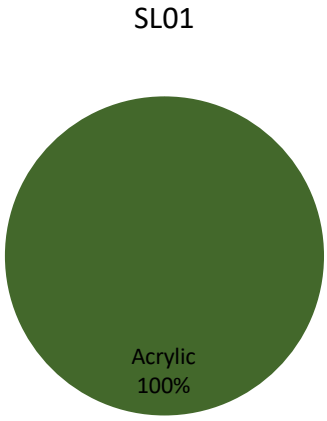
Supplementary Figure 4A: Size distribution of MP particles (n MP per 5% of sample filter) for each sample.

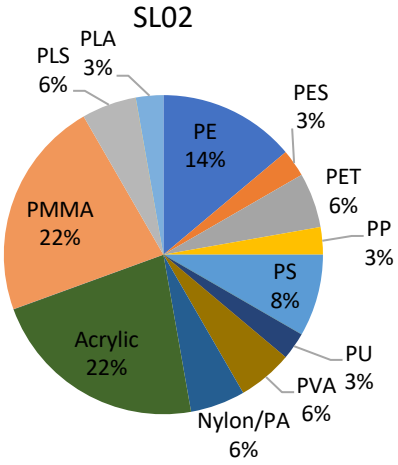
Microplastic particle size classification was performed using FIJI/ImageJ. This method is subject to user bias as it is a visual identification method.

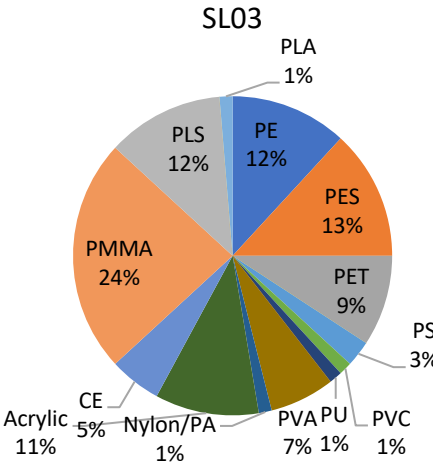


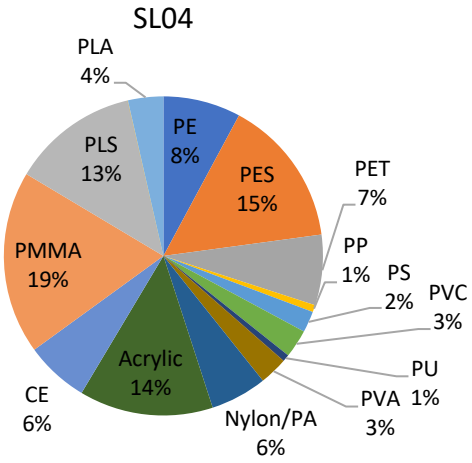
Supplementary Figure 5A: Microscopic image of sample 6 filter (collected during the hurricane) stained with Nile Red under fluorescent light. Note that the particles analyzed were mostly fragments.

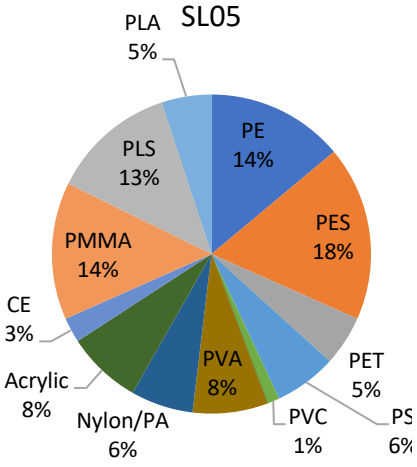
Supplementary Note 3A – Polymer Types

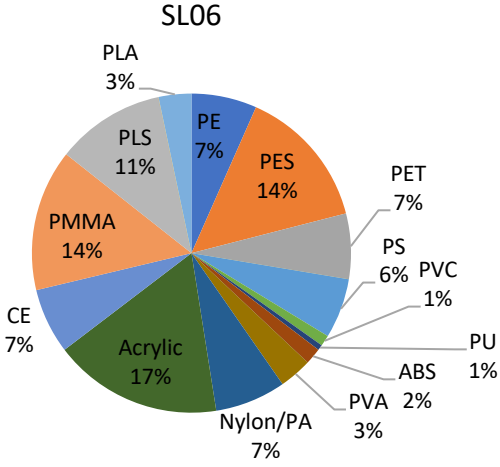


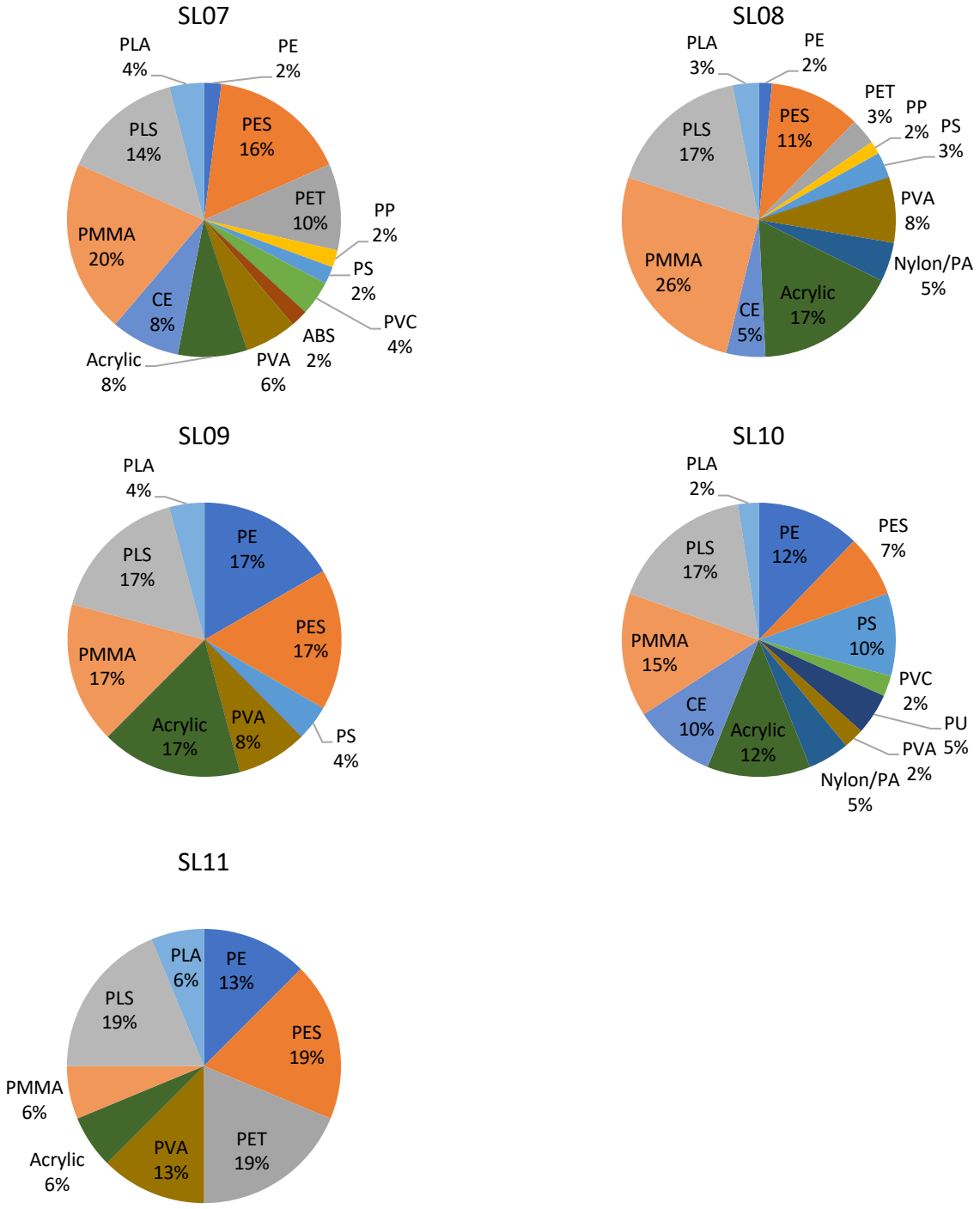




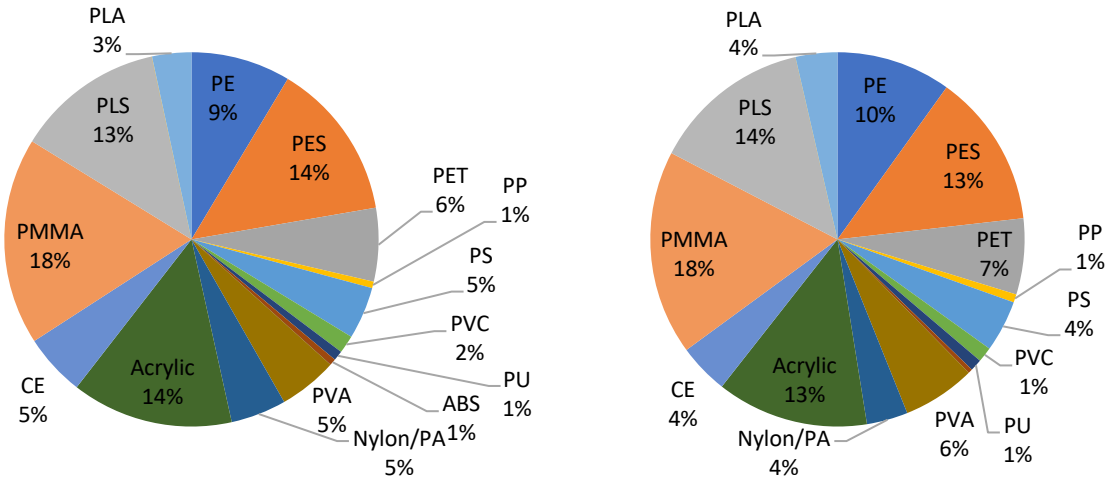






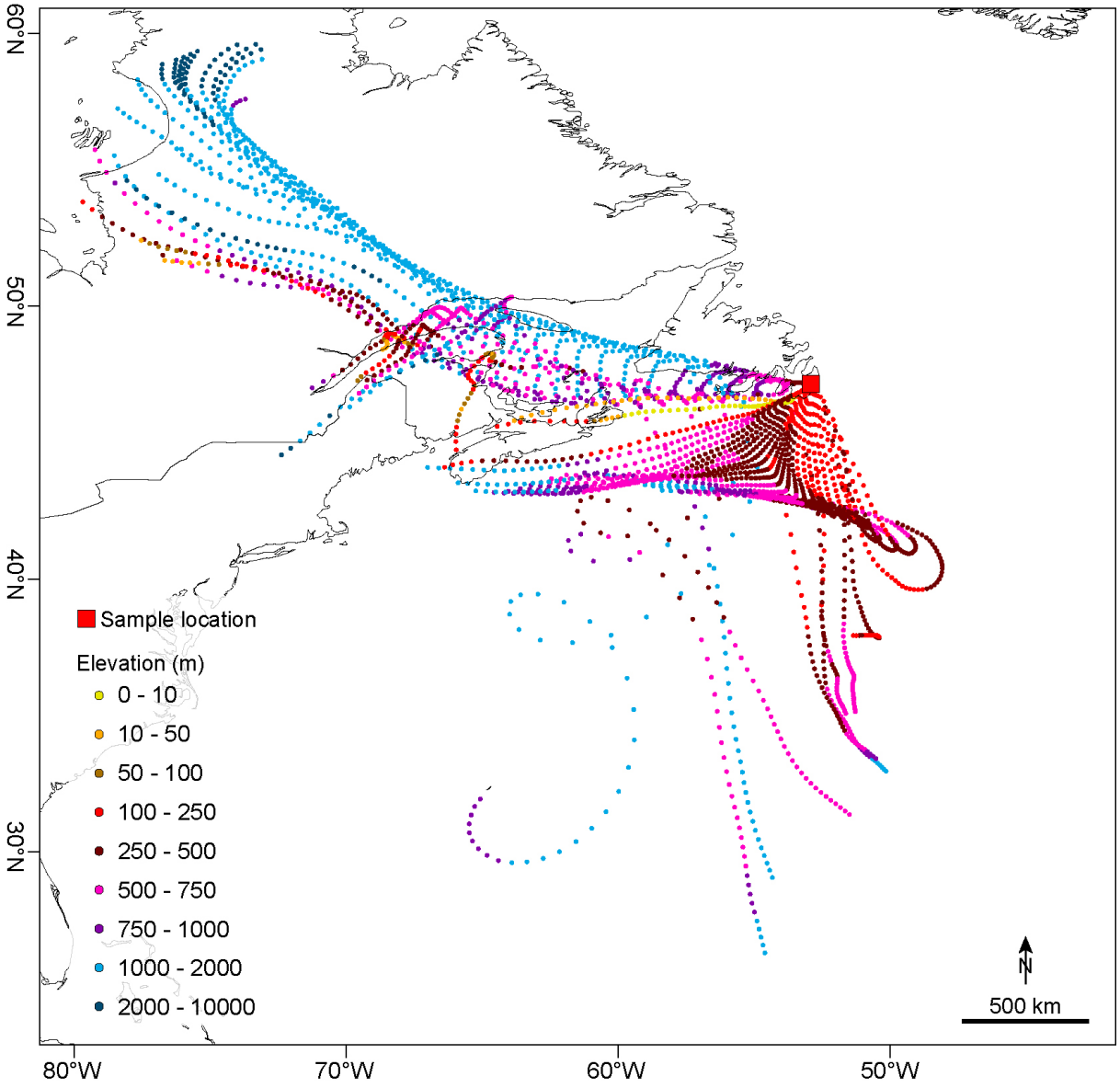


Supplementary Figure 6A: Polymer type distribution for each sample from µRaman spectrographic analysis.



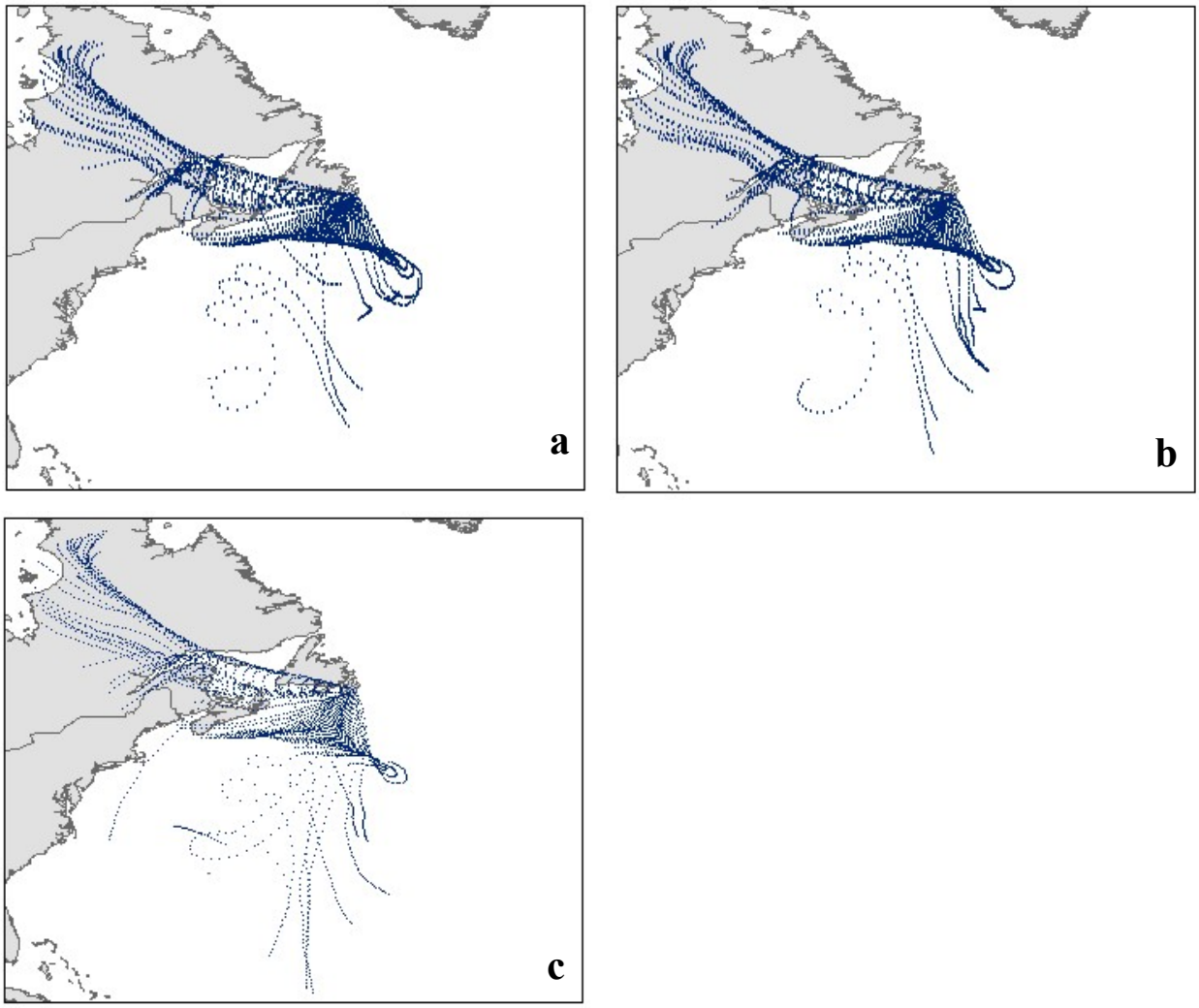
Supplementary Figure 7A: Overall polymer composition of microplastic particles for all samples (left) and overall polymer composition for all samples excluding SL01 (right), which was considered a contaminated sample and was excluded from the statistical analysis.

Supplementary Note 4A – Back-trajectory modelling and sensitivity analysis



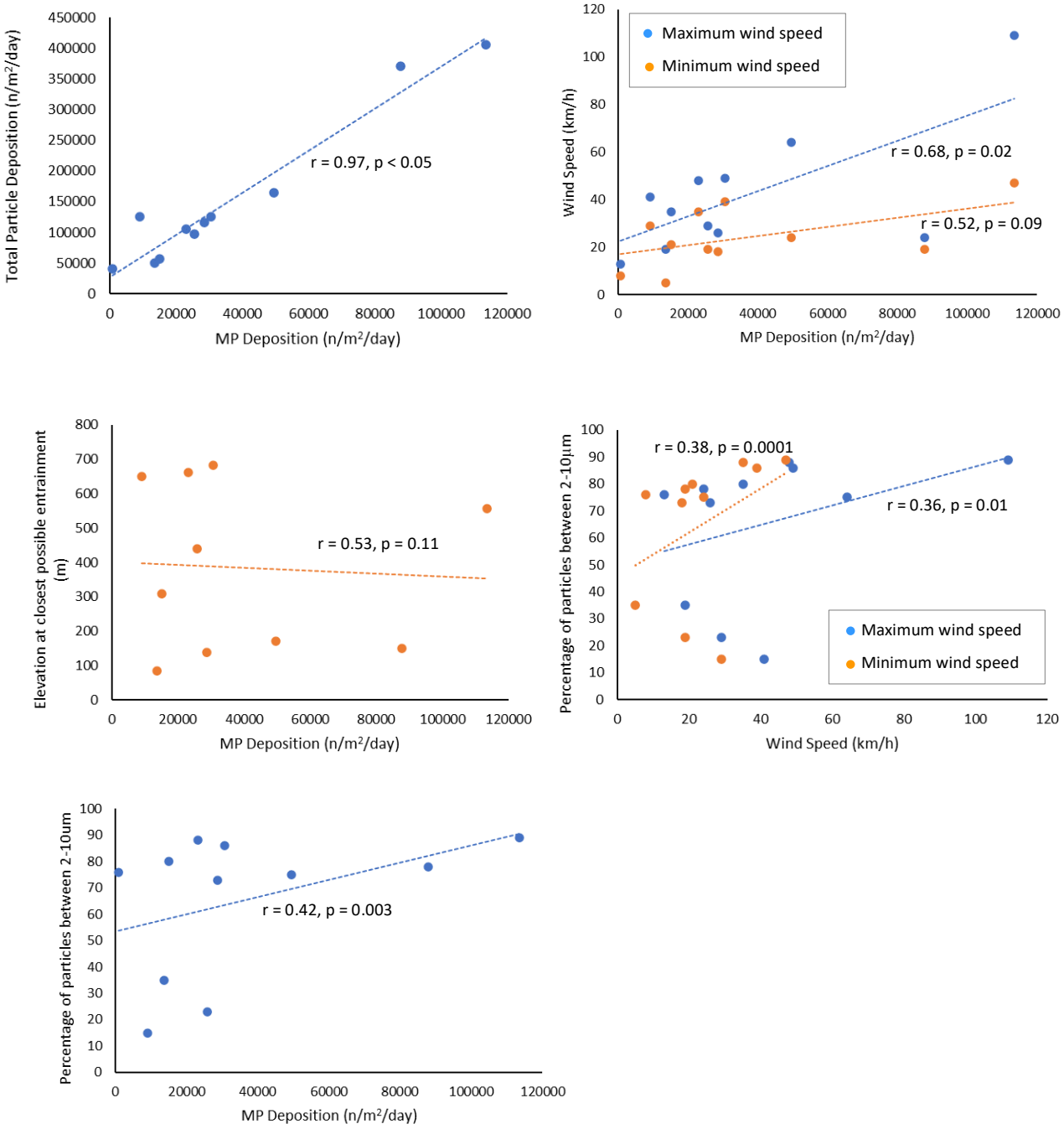
Supplementary Figure 8A: Air parcel 48 hour back-trajectories for every hour within the sampling period (n = 66) modelled using HYSPLIT (version 5) from a starting elevation of 0.5 the planetary boundary layer (~500 m above ground level) using archived GDAS 1 degree meteorological data. Each dot represents a trajectory endpoint, with endpoints for every hour during each trajectory. Air parcel elevation above ground level is represented by colour, with sample site location also indicated (red square).

I ran the HYSPLIT model from three starting elevations (0.25, 0.5, and 0.75 the planetary boundary layer). Endpoints were created for every hour of the 48 hour back-trajectory, with a new trajectory for every hour of the sampling period resulting in 66 total trajectories. I used residence time two, meaning that the frequency sum displayed in the figure is the number of endpoints in a grid square divided by the total number of endpoints (n = 3101). The air parcel trajectories show relatively little variation depending on model starting elevation meaning the HYSPLIT has low sensitivity to this parameter.



Supplementary Figure 9A: HYSPLIT back-trajectory models (48 h backwards timestep) for all sample trajectories (n = 66) run with GDAS 1 degree meteorological data from starting elevation of (a) 0.25 the planetary boundary layer, (b) 0.5 the planetary boundary layer, and (c) 0.75 the planetary boundary layer.

Supplementary Note 5A – Statistical Analysis



Supplementary Figure 10A: Correlation plots with reported Pearson correlation values and significance values for different variables measured or analyzed during Hurricane Larry as discussed in the text.

Supplementary Table 1A: Summary of particle deposition, model results, and weather conditions at Cape Race weather station for each sample.

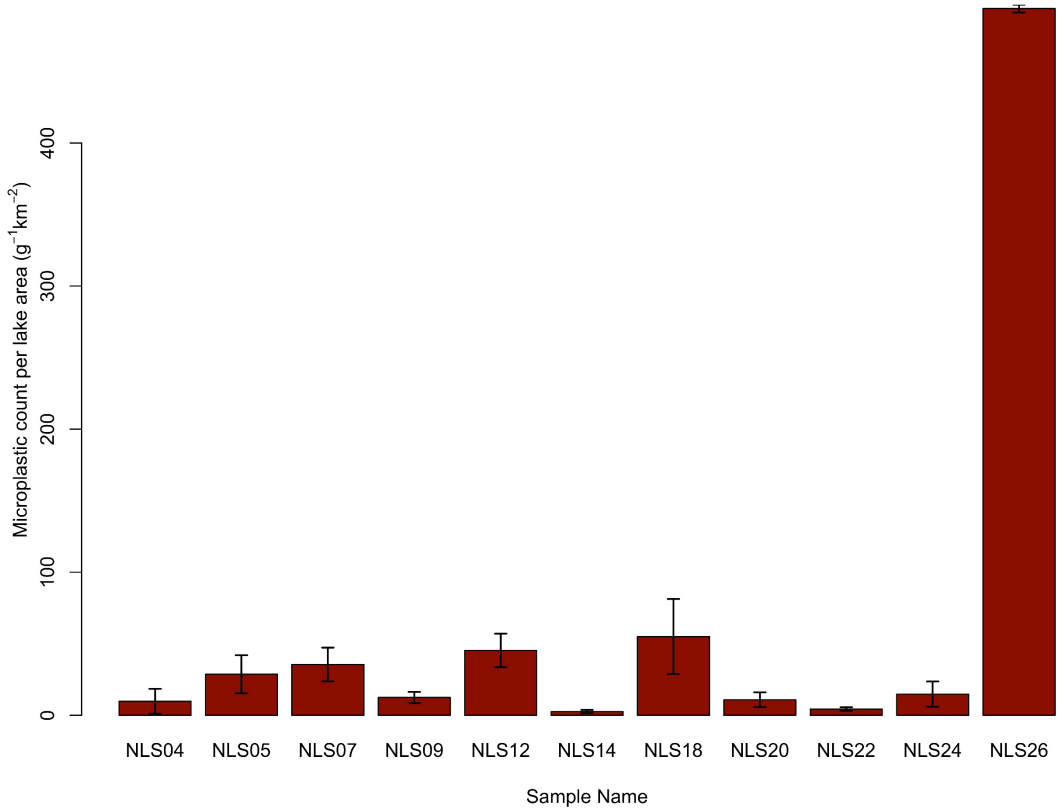
sample	Total particles ($n/m^2/d$)	MPs ($n/m^2/d$)	Distance to closest possible entrainment point (km, average, under 1000 m)	Elevation at closest possible entrainment point (m, average, above ground level)	Total rainfall (mm)	Max wind speed (km/h)	Min wind speed (km/h)	Dominant wind direction	Dominant particle size (µm)	Dominant polymer type
1	40282	753	19	81	0.2	13	8	SW	2-10	acrylic
2	50447	13553	23	85	0	19	5	SW	<2	Acrylic/PMMA
3	115953	28612	34	138	0	26	18	SSW	2-10	PMMA
4	370196	87843	46	149	0	24	19	SSE	2-10	PMMA
5	165020	49569	64	172	5.7	64	24	SE	2-10	PES
6	405961	113569	123	558	3.7	109	47	SW	2-10	Acrylic
7	104941	23059	85	661	0	48	35	SW	2-10	PMMA
8	126118	30588	78	683	0	49	39	SW	2-10	PMMA
9	126118	9035	75	650	0	41	29	WSW	<2	PE/PES/acrylic/PMMA/PLS
10	97882	25725	54	439	0	29	19	W	<2	PLS
11	57412	15059	31	310	0	35	21	WSW	2-10	PES/PET/PLS

Supplementary Table 2A: Polymer Abbreviations

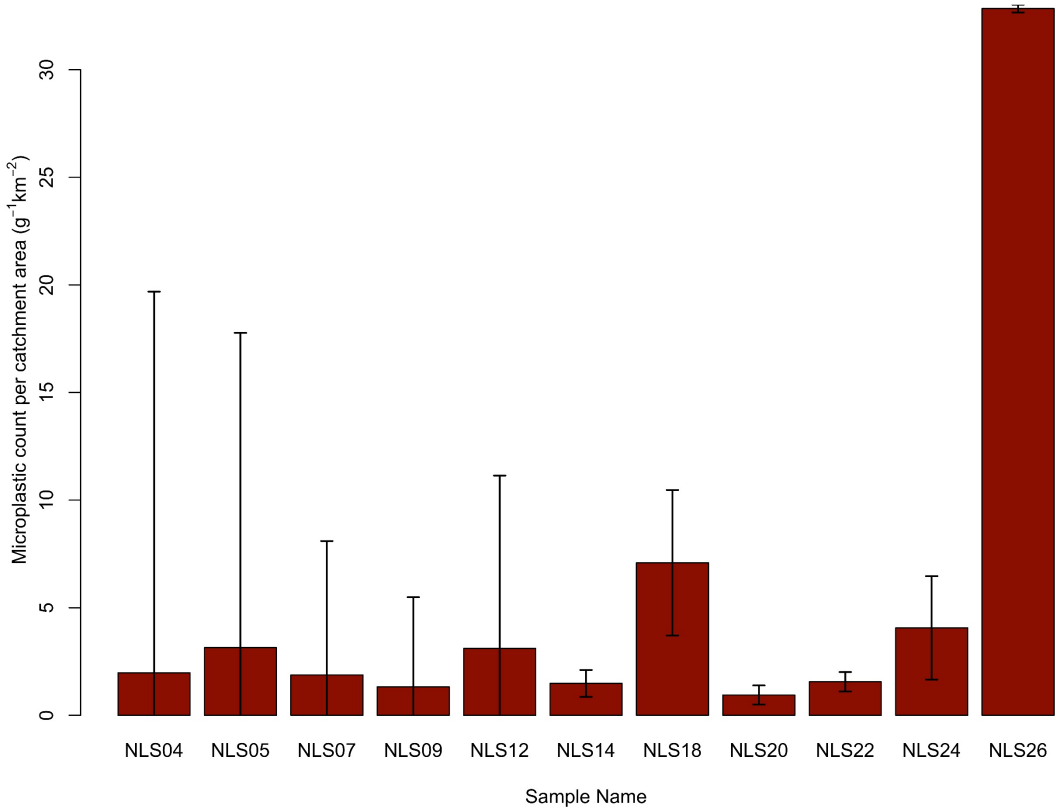
ABS	Acrylonitrile Butadiene Styrene
Acrylic	Acrylic
CE	Cellulose acetate
Nylon/PA	Nylon/Polyamide
PE	Polyethylene
PES	Polyester
PET	Polyethylene terephthalate
PLA	Polylactic Acid
PLS	Polysulfone
PMMA	Poly(methyl methacrylate)
PP	Polypropylene
PS	Polystyrene
PU	Polyurethane
PVA	Polyvinyl alcohol
PVC	Polyvinyl chloride

APPENDIX B: Supplementary material for "Microplastic accumulation in remote endorheic lakes and the role of atmospheric deposition: A case study from Newfoundland, Canada"

Supplementary Note 1B – Normalized Microplastic Abundance

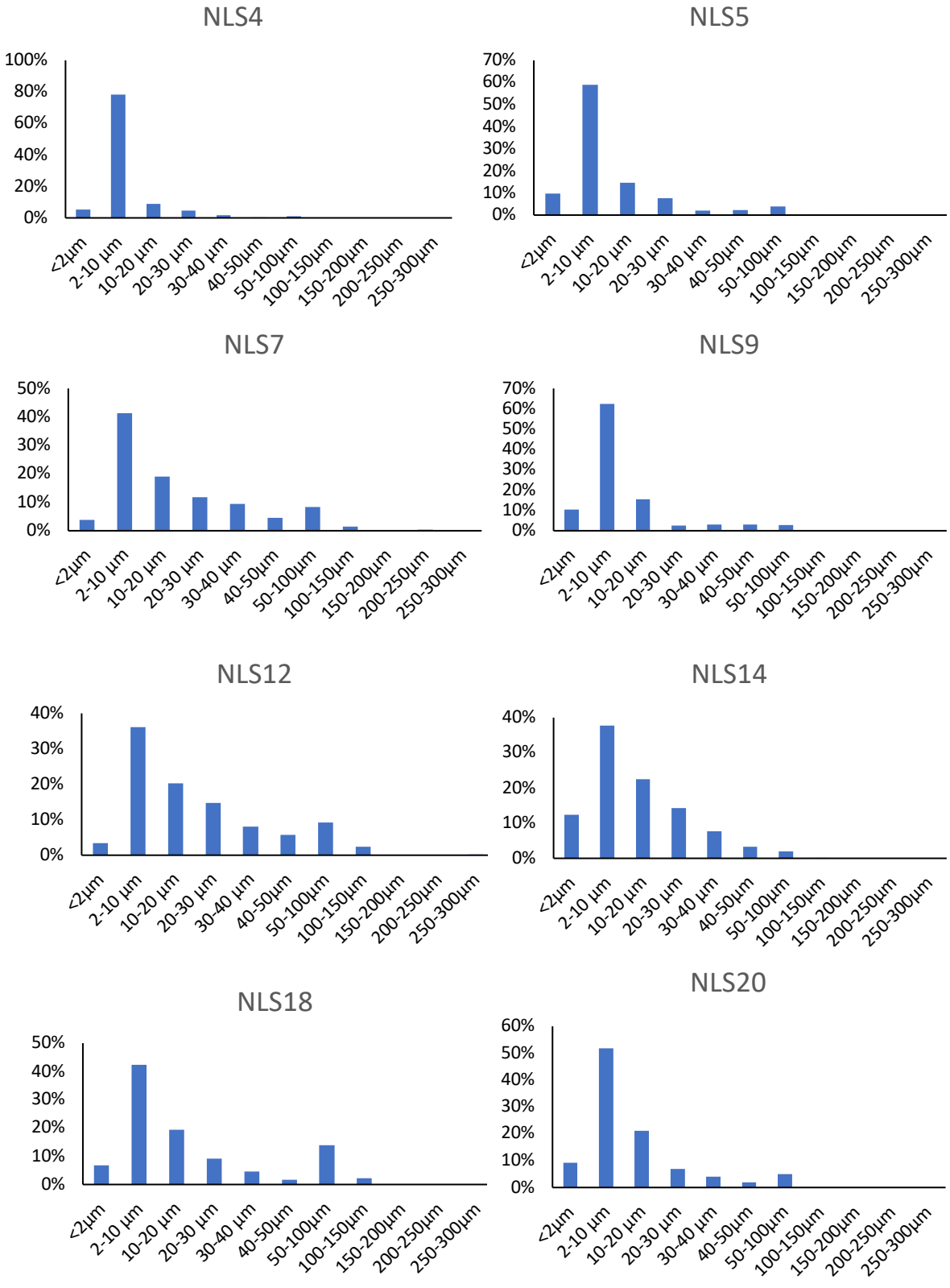


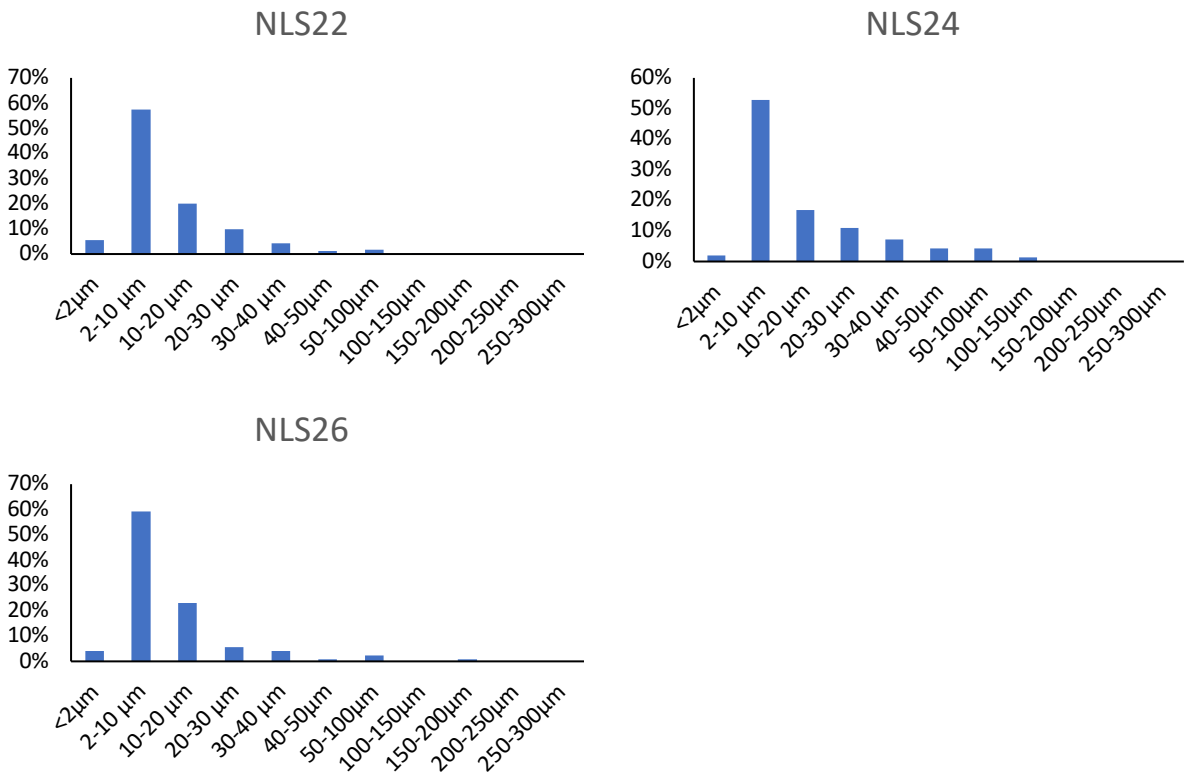
Supplementary Figure 1B: Microplastic particle count per lake surface area for each lake sampled (with error bars representing 1σ).



Supplementary Figure 2B: Microplastic particle count per lake catchment area (with error bars representing 1σ), delineated using Global Mapper and outlined in Figure 4.2, for each lake sampled.

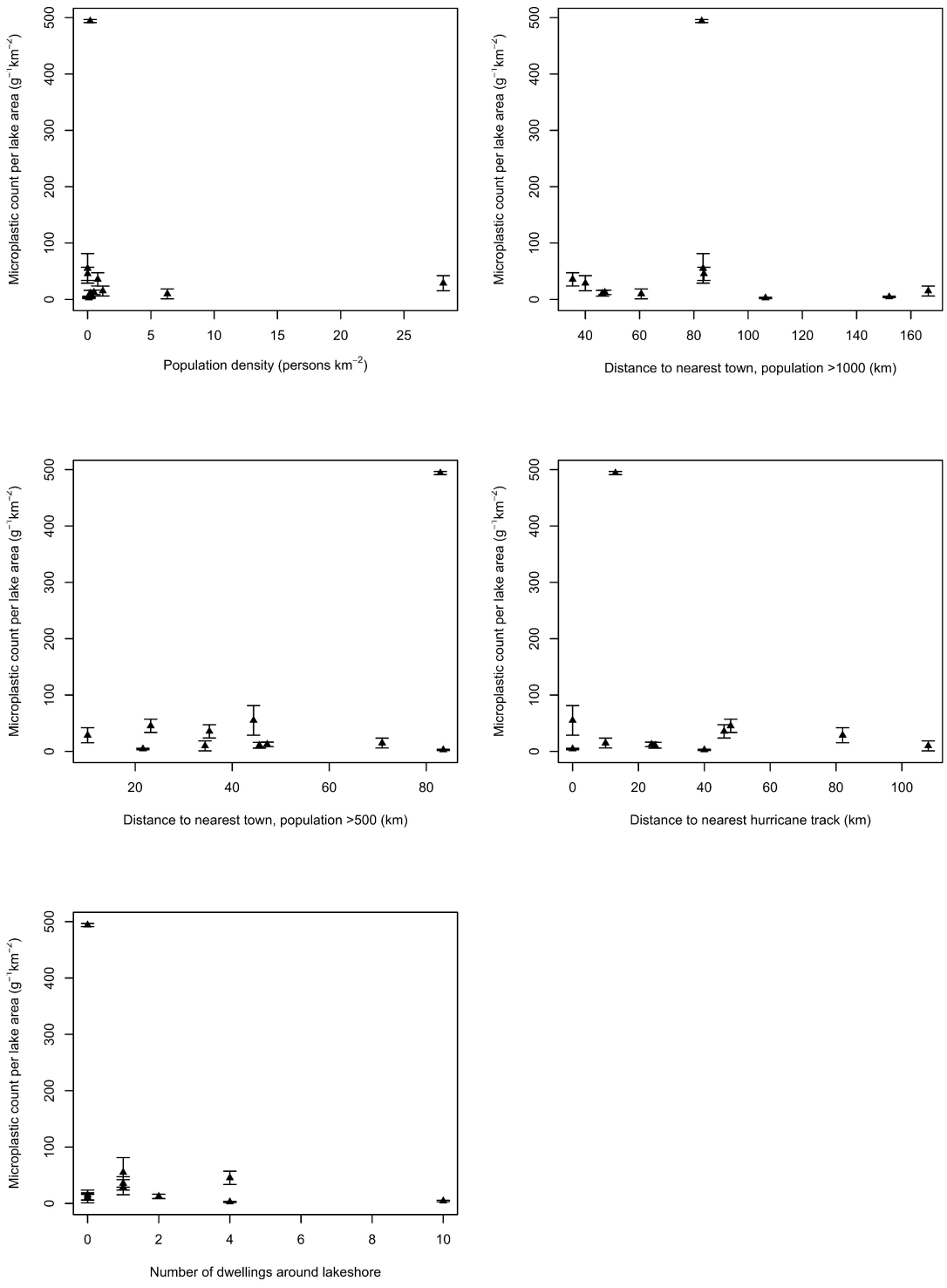
Supplementary Note 2B – Size Distribution



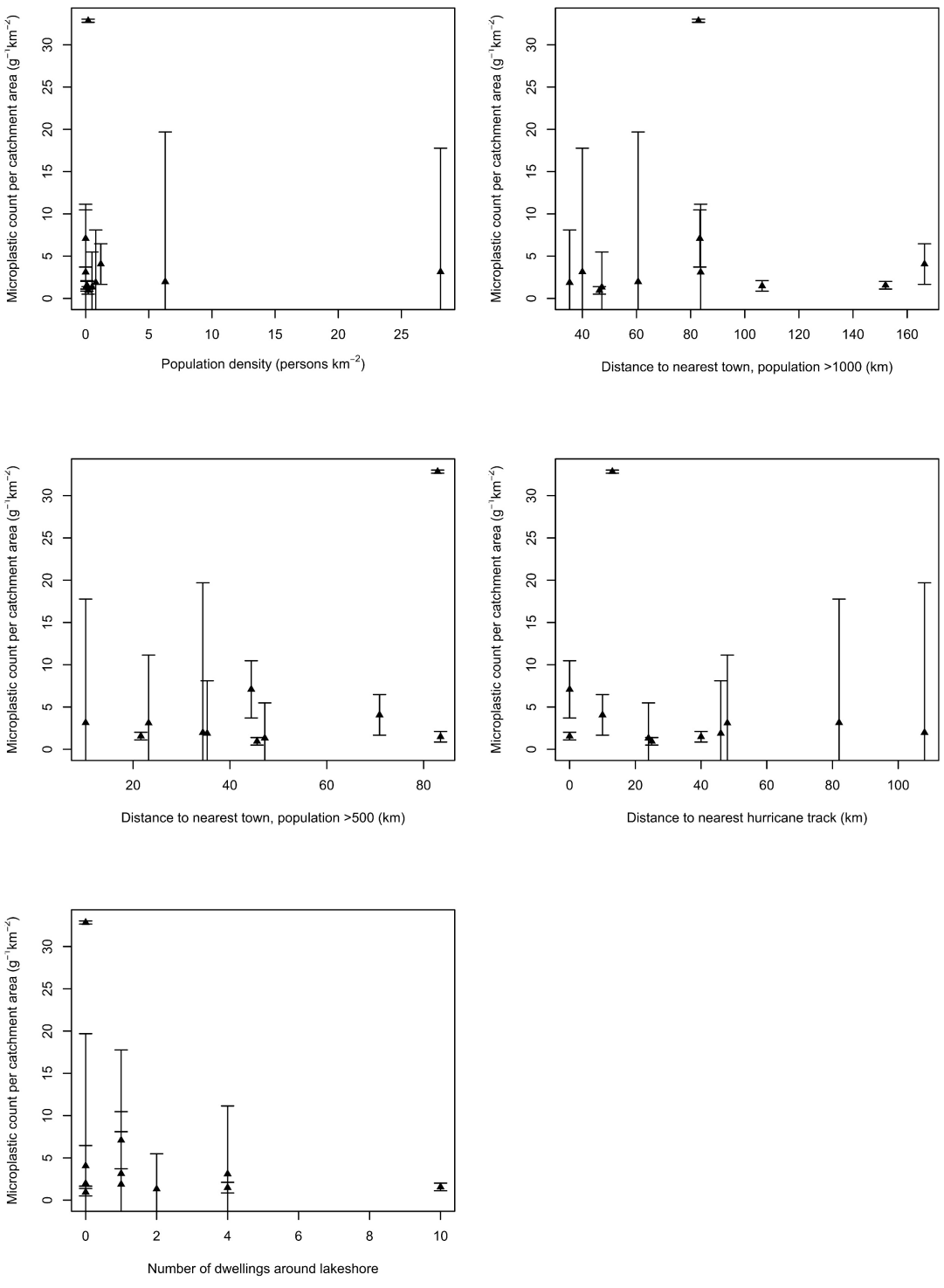


Supplementary Figure 3B: Microplastic particle size distribution for each sample with percentage of total particle count on y-axis and size class on x-axis.

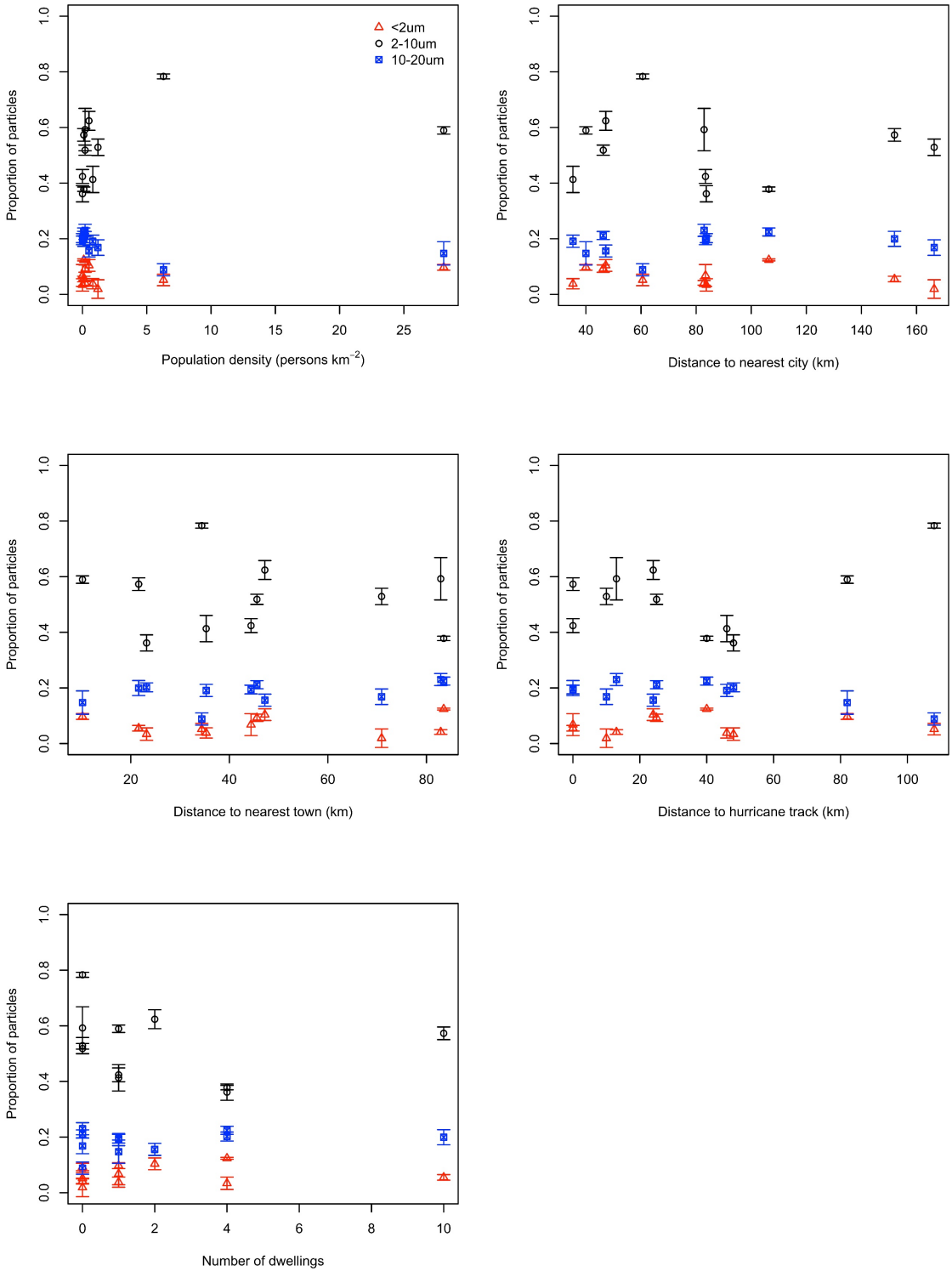
Supplementary Note 3B – Statistical Analysis



Supplementary Figure 4B: Correlation plots for different characteristics of lakes with average MP count normalized for lake surface area showing no correlation exists.



Supplementary Figure 5B: Correlation plots for different characteristics of lakes with average MP count normalized for lake catchment area showing no correlation exists.



Supplementary Figure 6B: Correlation plots for average proportion of microplastic particles in the three most common size classes (<2 µm, 2-10 µm, 10-20 µm) with different characteristics of lakes showing no correlation exists.