Time trends in abundance and composition of microplastic particles deposited in profundal sediment of two headwater reservoirs within the Grand River Watershed (Ontario, Canada)

by

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Author’s Declaration

This thesis consists of material all of which I authored or co-authored: see Statement of Contributions included in the thesis. This is a true copy of the thesis, including any required final revisions, as accepted by my examiners.

I understand that my thesis may be made electronically available to the public.

Statement of Contributions

I was the sole author for Chapters 1 and 3 which were written under the supervision of Dr. Roland Hall and were not written for publication.

This thesis consists in part of one manuscript written for publication (Chapter 2). I have contributed to this paper by writing the original draft, actively participating through the editing process, and leading all data acquisition and analysis. Co-authors of this manuscript to be submitted for publication include Dr. Fereidoun Reza Nezhad, Dr. Philippe Van Cappellen, and Dr. Roland Hall.

Abstract

Plastic pollution has become pervasive in the environment, raising concern for degradation of aquatic ecosystems by microplastics (MPs). Studies on the supply and abundance of MPs in Canadian freshwaters are rapidly emerging, however temporal trends spanning several decades remain sparse. In this study, we report multidecadal records of MP abundance and composition in dated sediment cores from two headwater reservoirs (Belwood Lake and Conestogo Lake) located within an agricultural region of the rapidly urbanizing Grand River Watershed (GRW; southern Ontario, Canada), a major tributary of Lake Erie. Extracted MPs from contiguous 1-cm thick intervals of sediment cores were enumerated and categorized by shape (fragment, fiber). A subset of samples at approximately decadal intervals were chemically characterized using Laser Direct Infrared (LDIR) Spectroscopy. Results reveal that MP concentrations in both reservoirs varied within a similar range (~50-550 particles g dw^{-1}) with no observed increasing trend since the start of each record (1957 for Belwood Lake, 1985 for Conestogo Lake). MP flux in Conestogo Lake increased from ~50 particles cm^{-2} $year^{-1}$ in the mid-1980s to ~100 particles cm^{-2} $year^{-1}$ in recently deposited sediment, whereas MP flux varied without a trend between ~5-60 particles cm^{-2} $year^{-1}$ in Belwood Lake since 1957, apart from a peak in the uppermost sample (~150 particles cm^{-2} $year^{-1}$). More rapid sediment deposition at Conestogo Lake accounts for the difference in MP flux between the reservoirs, suggesting reduction of sediment transport could reduce the supply of MPs to aquatic ecosystems in the GRW. Analysis by LDIR revealed that polyamide, rubber, and polyethylene were the most abundant polymer types in both reservoirs. The relative abundance of rubber particles has increased since the 1990s, indicating an increase of paved road surface and vehicle traffic as a potential source. The findings suggest MP accumulation in these upstream rural reservoirs of the GRW may be primarily driven by local, regional, and hydrologic factors instead of the rise in global plastic production.

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Chapter 1:
Introduction

Plastic is mass-produced and widely used by humankind, but most of the plastic produced accumulates in landfills, or in the environment, where it may cause irreversible detrimental ecological effects (Geyer et al., 2017; Macleod et al., 2021). This problem will worsen, as an additional 33 billion tonnes of plastic are expected by 2050 if consumption trajectories continue at current rates (Rochman et al., 2013b). Microplastics (MPs), small plastic particles less than 5 mm in size, are mostly generated through in-situ breakage of larger plastic waste (Andrady, 2011). These tiny particles are ubiquitous in the environment, which raises concern for potential toxic effects and implications this may have for biota, ecosystems, and human health. The consequences of MP pollution are not fully known and are debated (Kramm et al., 2018), but studies have demonstrated toxic effects to biota via disruptions in reproduction, feeding performance, and energy metabolism (Anbumani & Kakkar, 2018; Ogonowski et al., 2018). Long term ecological effects are yet to be fully characterized; however, alterations of ecosystem productivity and biodiversity have been suggested for aquatic ecosystems (Ma et al., 2020). Evolving research suggests increasing risks towards human health as the abundance of MPs continue to increase in the environment (Prata et al., 2020; Lim, 2021). Even if plastic production was halted immediately, MP pollution would persist for many decades due to the slow natural removal rate, infeasibility of cleanup methodology, and continuation generation of MPs via the breakdown of macroplastics in legacy waste (Lau et al., 2020; Macleod et al., 2021).

1.1 Microplastics in Canadian Freshwaters

To date, most of the research on MPs in aquatic ecosystems has prioritized marine environments, but there is increasing need to characterize the extent of MP pollution in freshwater bodies (Wagner et al., 2014; Horton et al., 2017; D'Avignon et al., 2022). Freshwater ecosystems are high in biodiversity and are relied on heavily as a resource for human populations, and, concerningly, MPs have been reported in all environmental compartments in freshwaters worldwide (including the water column, littoral and profundal sediments and biota) (Dudgeon et al., 2006; Eerkes-Medrano et al., 2015; D'Avignon et al., 2022). One of Canada's most important freshwater resources is the Laurentian Great Lakes basin, which is depended on for drinking water, recreation, transportation, power generation, and economic opportunities. MPs have been found in abundance in the Laurentian Great Lakes and surrounding area, with several review papers summarizing the rapidly emerging studies (Driedger et al., 2015; Earn et al., 2021; Fuschi et al., 2022; Hataley et al., 2024; McIlwraith et al., 2024). However, most of these studies provide a one-time snapshot of the abundance and composition of MPs in an environmental compartment at the study sites. Improved knowledge of the complexity of processes that affect MP transport, storage and degradation, and past plastic loadings is essential for the development of effective strategies for mitigation of problems caused by MP pollution.

Recently, the Canadian Government has taken steps to prioritize the mitigation of plastic waste entering the environment. This was done through development of the Canada-Wide Strategy on Zero Plastic Waste, a framework maintaining clear actions and timelines to reduce future and current plastic waste in the environment (Canada Council of Ministers of the Environment, 2018). Additionally, Canada's Plastic Science Agenda (CaPSA) outlines key themes to be addressed in Canadian plastics science research and identifies knowledge gaps and areas of priority (Environment and Climate Change Canada, 2019). As a result of these initiatives, a special call for research was made (NSERC Alliance grants on *Plastics Science for a Cleaner Future*) with the goal to fund multidisciplinary, collaborative research to improve our knowledge of the MP problem in freshwater ecosystems of Canada and our capacity to solve it.

1.2 The Microplastics Fingerprinting Project

A team of researchers at University of Waterloo received funding from the NSERC Alliance *Plastics Science for a Cleaner Future* competition for the 'Microplastics Fingerprinting Project', which attempts to explore the complexity of processes affecting MP behaviour at a watershed scale. This project aims to better understand the sources, transport, fate, and exposure risks of MPs through a watershed scale analysis focusing on streams, wastewater, and stormwater management systems in southern Ontario, where Canada's population is relatively dense. The main study areas for this project are the watersheds of the Grand River and Don River in southern Ontario. These are two rapidly urbanizing regions that provide opportunity to assess MP pollution in landscapes adjacent to the Lower Laurentian Great Lakes. Researchers in the Microplastics Fingerprinting Project work closely with stakeholders in these watersheds who have a broad knowledge base on local water resource management and watershed dynamics, allowing for a comprehensive understanding on regional factors that may influence MP loading. Knowledge gained on MP sources, transport, fate, and exposure risk from these regions may be applied to other watersheds across Canada and globally.

The scope of the research in the Microplastics Fingerprinting Project includes refining methods for the detection, quantification, and characterization of MPs in the environment, and developing approaches based on measurements and modeling to understand the sources and fate of different types of MPs. The data generated by this project will be further used to inform science-based risk assessments, governance approaches and watershed management, as well as test the real-world applicability of new fingerprinting methods, watershed-scale modeling, and risk mitigation approaches. The Microplastics Fingerprinting Project attempts to specifically fulfil Theme 1 of CaPSA, which considers the detection, quantification, and characterization of plastics in the environment (Environment and Climate Change Canada, 2019).

To do this, the Microplastics Fingerprinting Project aims to address several objectives:

1: Develop detection methods for structural and chemical fingerprinting of MPs,

2: Use MP fingerprinting to identify the factors influencing MP sources and entry pathways along the river to lake continuum,

3: Quantitatively link the sources, transport, and fate of MPs at a watershed scale,

4: Determine the contribution of extreme hydrological events to MP emissions from land to water bodies,

5: Assess the preservation of MPs in aquatic sediments and undertake analyses of sediment cores from depositional basins to reconstruct past changes in MP pollution,

6: Quantify the loading of MPs from urbanizing watersheds to the lower Laurentian Great Lakes,

7: Perform an economic analysis of plastic pollution in the Great Lakes, and

8: Analyze MPs in drinking water sources and assess exposure risks.

1.3 Research Objectives

Long-term data on MP pollution in freshwater ecosystems are scant, however the amount and type of MPs released to the environment have likely varied through time due to rapid advancement of the plastics production industry since the 1950s (Geyer et al., 2017). Long temporal data can be used to identify when MP loading to waterbodies began to increase, quantify the scale of the problem, and inform mitigative actions and policy. As plastic remains persistent in the environment, substantial plastic waste may be buried in lakebed sediments. Analysis of sediment cores from lakes and reservoirs, thus, can be used to generate these temporal data by effectively assessing past changes in MP deposition in the bottom sediment (Smol, 2008). Recent studies have shown that analysis of dated sediment cores can be used to successfully characterize past trends in MP pollution (Turner et al, 2019; Dong et al, 2020; Almas et al, 2022; Belontz et al., 2024).

My MSc research contributes to the above Objective 5 of the Microplastics Fingerprinting Project. The goal is to analyze MPs in dated sediment cores collected from two reservoirs within the GRW (Belwood Lake and Conestogo Lake) to determine past variation in MP abundance, flux and composition and identify the possible processes that could account for any changes that are detected. Belwood Lake lies directly on the Grand River and nearby Conestogo Lake lies on the Conestogo River, which is a major tributary of the Grand River. They are the only reservoirs that lie directly on major waterways in the GRW, and they are well-established as upstream sinks for sediment and contaminants that would otherwise be transported downstream (Loomer & Cooke, 2011). The results will be used to identify potential MP sources and pathways of MPs delivered to the GRW and to improve modelling of MP storage and transport through the GRW.

The following chapter (Chapter 2) presents a scientific article intended for submission to a peer-reviewed journal, entitled "*Time trends in abundance and composition of microplastic particles deposited in profundal sediment of two headwater reservoirs within the Grand River Watershed, (Ontario, Canada)*", by Meredith Watson, Fereidoun Reza Nezhad, Philippe Van Cappellen, and Roland Hall. As first author, I have contributed to this paper by writing the original draft, actively participating through the editing process, and leading all data acquisition and analysis. The third and final chapter of this thesis presents synthesis and recommendations for this research project.

Chapter 2:

1. Introduction

Plastic pollution has become a major environmental problem, with an estimated 79% of plastic produced ending up in landfills or the environment (Geyer et al., 2017). In Canada, plastic production has increased approximately 230-fold since the 1950s and remains a growing industrial sector (Environment and Climate Change Canada, 2019). Environmental dispersal of microplastics (MPs), small plastic particles <5 mm in size, is an emerging concern due to potential negative and toxic effects to biota and humans (Rochman et al., 2013a; Ogonowski et al., 2018; Smith et al., 2018; Prata et al., 2020). Despite the concern, the production of plastic waste worldwide is expected to continue to rise, increasing risk of negative effects of MPs on aquatic environments (Geyer et al., 2017; Borelle, 2020).

In the last decade, studies have rapidly emerged that document MPs as a pervasive form of contamination in freshwater environments worldwide, where MP particles have been identified in water, sediments, and biota of profundal and shoreline habitats (Eerkes-Medrano et al., 2015; D'Avignon et al., 2021). MPs are transported to aquatic ecosystems via runoff and air, and a portion of them settle to bottom sediment via behavior that is similar to fine-grained sediment (Corcoran et al., 2019; Vincent & Hollein, 2021). This means that MPs accumulate in depositional environments such as profundal sediments in lakes and reservoirs, where they typically are well preserved (Tibbetts et al., 2018; Wilson et al., 2021; Dhivert et al., 2022; Simon-Sanchez et al., 2022). Plastic degrades very slowly even when exposed to UV radiation and warm temperatures, so low UV radiation and colder temperatures in profundal locations promotes preservation of MPs (Bancone et al., 2020; Chamas et al., 2020). Analyses of MPs in dated sediment cores provides one of the only ways to generate long-term data on the supply of MPs to freshwater ecosystems, because MPs have only recently emerged as a contaminant of concern and, consequently, long-term monitoring records are largely unavailable (Turner et al., 2019; Dong et al., 2020; Almas et al., 2022; Belontz et al., 2024). Knowledge of temporal trends in abundance, flux, and composition of MP particles in freshwater ecosystems, however, is needed to inform policies and practices that safeguard our freshwaters environments from plastics pollution.

In this study, we focus on the GRW which is the largest inland river system in southern Ontario, Canada, and an important tributary discharging into the eastern basin of Lake Erie. The GRW is dominated by agricultural land (70%) and currently supports the most rapid urban growth in Ontario, the most populous province in Canada (GRCA, 2022; Pickel, 2024). Knowledge of MP dispersal to freshwaters of the GRW, however, remains minimal, despite reported high concentrations of MPs in Great Lakes tributaries (McIlwraith et al., 2023). To our knowledge, there are only two relevant published studies on MPs in the GRW. One undertook a spatial assessment of MPs in surficial sediment at nearshore locations within Lake Erie, which revealed the highest concentrations occur at the mouths of the Detroit River and Grand River (Dean et al., 2018). The other, a contemporary study of freshwater mussels in the GRW, determined that concentrations of MP particles are highest at downstream locations in the watershed (Wardlaw & Prosser, 2020). While these studies provide important information on spatial distributions of MP abundance in the GRW and Lake Erie, there is lack of longer temporal data capable of improving our understanding of how MP abundance, flux and composition have varied in the GRW during the decades since the "plastic age" began in the 1950s. To address this, we undertook analyses of sediment cores from the two largest reservoirs in the GRW (Belwood Lake, Conestogo Lake) to generate records of MP abundance, flux, and composition since the late 1950s. In doing so, we aimed to characterize temporal trends in MP deposition to aquatic sediments in the rural, headwater region of the GRW and identify possible processes influencing these trends. The results from this study may assist with development of a highly recommended MPs monitoring program for the Great Lakes basin (Hataley et al., 2023; McIlwraith et al., 2023).

2. Methods

2.1. Study site

The study sites are located within an agricultural region of the GRW and upstream of the main urban centers (Kitchener, Waterloo, Cambridge, Guelph, Brantford). Sediment cores were collected from two flood control reservoirs, Belwood Lake (593-ha) and Conestogo Lake (735-ha), located on the Grand and Conestogo rivers, respectively (Figure 2.1). These reservoirs were selected because they accumulate sediment representative of the upstream portions of the largest sub-watersheds of the GRW. The reservoirs were constructed with establishment of the Shand Dam in 1942 (Belwood Lake) and Conestogo Dam in 1958 (Conestogo Lake). Their watersheds have a low population density (≤ 80 people km^2), and the land use is primarily agricultural (Loomer & Cooke, 2011). The dams have created profundal depositional areas for sedimentation that act as a sink for contaminants from upstream regions and a potential source to downstream locations (Loomer & Cooke, 2011).

Figure 2.1: Map of Conestogo Lake, Belwood Lake, and sediment core collection sites. The map displays the boundaries of the Grand River Watershed (blue line) with locations of the two reservoirs (red), nearby urban centers (white), and coring sites (purple) identified. Map created using free and open source QGIS.

2.2. Field methods

2.2.1. Sediment core collection

Sediment cores were collected from Belwood Lake on February 14th, 2018, and from Conestogo Lake on June 13th, 2023, using a hammer driver gravity corer. Coring was performed in the central deep-water basin of both reservoirs in depositional zones behind the dams. Cores were collected at a depth of ~9.5 m in Belwood Lake and ~225 m upstream from the Shand dam, and at ~16 m water depth in Conestogo Lake and ~430 m upstream from the Conestogo dam. All sediment cores were sectioned into 1-cm intervals into Whirl-Pak bags. Two cores were obtained from each reservoir, with the longer core selected for analysis of MPs at both Belwood Lake (48 cm) and Conestogo Lake (67 cm).

2.3. Laboratory analyses

2.3.1. Sediment composition

Loss-on-ignition analysis was performed on contiguous 1-cm intervals of the cores to determine variation in sediment composition (Heiri et al., 2001). For each sample, approximately 0.5-1 g of wet sediment was placed into a pre-weighed ceramic crucible and sequentially heated at 90°C for 24 hours, 550°C for 2 hours, and 950°C for 2 hours. After heating to each temperature, samples were allowed to cool to 60-90°C and were placed in a desiccator for approximately 2 hours before weighing. Water content was determined from the mass loss after heating to 90°C, organic matter content determined from the mass loss after heating to 550°C, and carbonate content was determined by multiplying the mass loss after heating to 950°C by 1.36. The carbonate-free mineral matter content was determined as the residual dry sediment mass. Data generated from these analyses are reported in Tables A1-A3 of the Appendix.

2.3.2. Radiometric dating

For radiometric dating of a core from each reservoir, subsamples of sediment were prepared from the top three 1-cm intervals and from alternating 1-cm intervals below 3-cm depth. For each sediment interval, a measured mass of well mixed freeze-dried sediment was packed into pre-weighed polycarbonate tubes to an approximate height of 3.5 cm, sealed with a septum and 1 mL of epoxy, and stored for at least 21 days before analysis of radioisotope activity to allow for ^{222}Rn and its decay products to equilibrate with ^{226}Ra (Binford, 1990; Appleby, 2001). Radioisotope activities (^{210}Pb, ^{214}Bi, ^{214}Pb, and ^{137}Cs) were measured with an Ortec HPGe Digital Gamma Ray Spectrometer using Maestro 32 software at the University of Waterloo, Canada. Unsupported ^{210}Pb activity was determined as the difference between total ^{210}Pb activity and supported ^{210}Pb activity, where supported ^{210}Pb activity was estimated using ^{226}Ra (determined as the average of ^{214}Pb and ^{214}Bi activity). Unsupported ^{210}Pb was used in conjunction with the Constant Rate of Supply (CRS) model to determine the age-depth relations and sedimentation rates (Appleby and Oldfield, 1978; Sanchez-Cabeza and Ruis-Fernandez, 2012). To evaluate the accuracy of the ^{210}Pb-based chronology, ^{137}Cs activity profiles were assessed for presence of a peak associated with above-ground nuclear weapon testing in 1963 (Appleby, 2001). Data generated from these analyses are reported in Tables A4 and A5 of the Appendix.

2.3.3. Microplastics extraction and enumeration

MP particles were extracted from well mixed subsamples of wet sediment at selected stratigraphic depth intervals using methods described by Yu et al. (2024). Briefly, ~2 g of wet sediment was dried at 65°C and spiked with a known number of MP standards that ranged in shape, size, polymer, and colour, allowing for generation of recovery rates that accommodate for the diversity of MP behavior during separation from the sediment matrix (Polyethylene (PE) Blue Bead– 38-45 µm, PE Orange Bead – 53-63 µm, PE Pink Bead– 63-75 µm, PE Green Bead– 180-212 µm, PE White Bead – 300-355 µm, PE Yellow Bead– 355-425 µm, polyamide (PA) Red fiber – 20-500 µm, polyethylene terephthalate (PET) Green Fragment – 200 µm). The dried sediment samples were wet-sieved through a 500-µm sieve using a 60% $ZnCl_2$ solution (density = 1.8 g mL^{-1}). The <500-µm fraction was filtered through a 20-µm sieve to capture particles (MPs, sediment, organic) in the 20-500 µm size range. These particles were transferred into a glass pitcher placed in a glass tray for density separation in $ZnCl_2$. This solution was well-mixed via aeration and left overnight to allow the MPs to float and the sediment particles to settle. The upper portion of the solution containing MPs was poured out of the glass pitcher into the glass tray and the density separation was repeated a second time. The fraction of $ZnCl_2$ in the glass tray containing MPs was filtered through a 20-µm sieve to collect MPs. MP particles were rinsed off the sieve using 70% ethanol and evaporated at 50°C prior to removal of organic residues via Fenton Oxidation. Finally, extracted MPs were collected on a wet nylon mesh (28-µm) and placed on a glass petri dish.

Using a stereomicroscope, spiked MP standards were identified by size and colour and enumerated to generate an average recovery rate for fragments and fibers. Fragments and fibers were identified and enumerated at 25x magnification. These raw manual count data are reported in Tables A6 and A7 of the Appendix. Manually counted MPs were then stored in 70% ethanol for further analysis by laser direct infrared (LDIR) spectroscopy. To minimize uncertainties between samples, every fifth (Belwood) or tenth (Conestogo) 1-cm interval was processed in duplicate. This procedure was performed in a laminar flow hood while wearing cotton lab coats, and blank samples were run continuously to monitor for laboratory contamination.

2.3.4. Chemical characterization of microplastic composition

Chemical characterization of the composition of MP particles by LDIR spectroscopy was conducted on a subset of the stratigraphic intervals to achieve approximately decadal resolution. Prior to LDIR sample preparation, the extracted MPs that had been stored in ethanol were allowed to fully evaporate at 50°C to remove the ethanol. For each sample analyzed, 5 mL (50 x 100 µl) of reagent grade isopropyl alcohol was added to each glass vial containing the extracted and dried MPs, and the vials were vortexed to homogenize the sample. Subsamples of 50-100 µl (2 x 25 or 2 x 50 µl) of this solution were pipetted onto a clean Kevley slide and scanned using an Agilent 8700 LDIR spectrometer. Composition of MPs was assessed using Agilent Clarity software by comparing selected particles to a built-in spectral library that was enhanced with spectra of common lab and field contaminants, MP standards, and degraded particles.

2.4. Data analysis

2.4.1. Determining microplastic concentration and flux

Microplastic concentrations and fluxes (totals, fragments, fibers) were generated using manual counts. Counts for each category of MP (fragment, fiber) were adjusted for contamination during processing in the laboratory by subtracting average contaminant count as identified in processed laboratory blank samples (fragments: 2.6 particles, fibers: 6.9 particles). Counts were then adjusted for loss of MPs during processing by using averaged recovery rates of microplastic standards. Recovery-corrected counts for the fragments and fibers were then summed to get total MP counts. All recovery-corrected counts were divided by the estimated dry mass of the processed wet sediment to generate MP concentrations in the sediment (particles g dw^{-1}). MP flux (particles cm^{-2} $year^{-1}$) was determined by multiplying the MP concentration with the dry mass sedimentation rate generated from the CRS model. Each flux value was corrected for sediment focusing by dividing by the ^{210}Pb-determined focus factor (FF) for each lake (Belwood: 3.497, Conestogo: 5.454) (Muir et al., 2009).

2.4.2. *Determining microplastic composition*

Prior to analysis of samples from the sediment cores, spectra for MPs common in laboratory and field samples were loaded into the LDIR spectral library, in addition to the built-in spectral library designed for MP analysis. The compositional data generated for each sediment core sample by LDIR spectroscopy were filtered in Microsoft Excel. MP particles were filtered out of the dataset if the match quality was ≤0.7 (following Bao et al., 2022), and if they were identified as a lab contaminant, field contaminant or naturally occurring polymer. To capture the data for particles that are representative of MPs determined by manual counting, we removed particles from the LDIR results with an average dimension <28 μm. Remaining MP particles (synthetic origin, non-contaminant, >28 μm) were counted by identified polymer type to determine relative abundance of each polymer by count within samples. The mass of the MP particles was estimated using an average density for each polymer type (using values reported in Dreidger et al., 2015) multiplied by the calculated volume based on average particle dimensions, as determined by the LDIR spectrometer. We used these data to estimate the relative mass of each polymer type in each sample analyzed.

3. Results and Interpretation

3.1. *Radiometric dating*

Total ^{210}Pb activity declined with depth in the sediment cores from Belwood Lake and Conestogo Lake, with the exception that the activity increased between the uppermost two samples at Belwood Lake (Figure 2.2A). Total ^{210}Pb activity was higher in the basal sediments of the core from Conestogo Lake (~75-100 bq kg^{-1}) than from Belwood Lake (~60 bq kg^{-1}). Activity profiles for ^{226}Ra, which are used to estimate supported ^{210}Pb activity, remained relatively constant throughout the cores (Belwood Lake: ~23-37 bq kg^{-1}; Conestogo Lake: ~36-44 bq kg^{-1}). Total ^{210}Pb activity was higher than ^{226}Ra activity at the bottom of the cores from both reservoirs, indicating supported ^{210}Pb activities were not reached, likely due to rapid sedimentation. An interpretation of rapid sedimentation is supported by evidence from ^{7}Be activity, a short-lived radioisotope (half-life = 53 days), which was detected at decreasing levels in only the uppermost 2 cm of the core from Conestogo Lake (0-1 cm = 133.5 bq kg^{-1}; 1-2 cm = 121.2 bq kg^{-1}).

Application of the Constant Rate of Supply (CRS) model to stratigraphic profiles of unsupported ^{210}Pb activity provided estimates of the relation between core depth and age of sediment and sedimentation rates. Basal dates were estimated to be ~1957 for the core from Belwood Lake and ~1985 for the core from Conestogo Lake (Figure 2.2B). The activity profile for ^{137}Cs could not be used for independent verification of the ^{210}Pb-based chronology due to absence of a well-defined activity peak associated with peak above-ground nuclear bomb testing in 1963. The absence of a ^{137}Cs peak at Conestogo Lake, however, is consistent with a basal date of ~1985 because it post-dates 1963 by two decades. At Belwood Lake, ^{137}Cs activity rises below 38 cm, which was deposited during ~1955 - 1965 based on ^{210}Pb dating and may capture the tail of the ^{137}Cs peak associated with above-ground nuclear bomb testing. Dry mass sedimentation rate was estimated to be approximately twice as high at Conestogo Lake (~0.75-1.18 g cm^{-2} year^{-1}) compared to Belwood Lake (~0.35-0.62 g cm^{-2} year^{-1}) (Figure 2.2C). At Belwood Lake, sedimentation rate was slightly lower during ~1975-1990 and ~1995-2005 than at other times. The core from Conestogo Lake also recorded an interval of lower sedimentation during ~1995-2005.

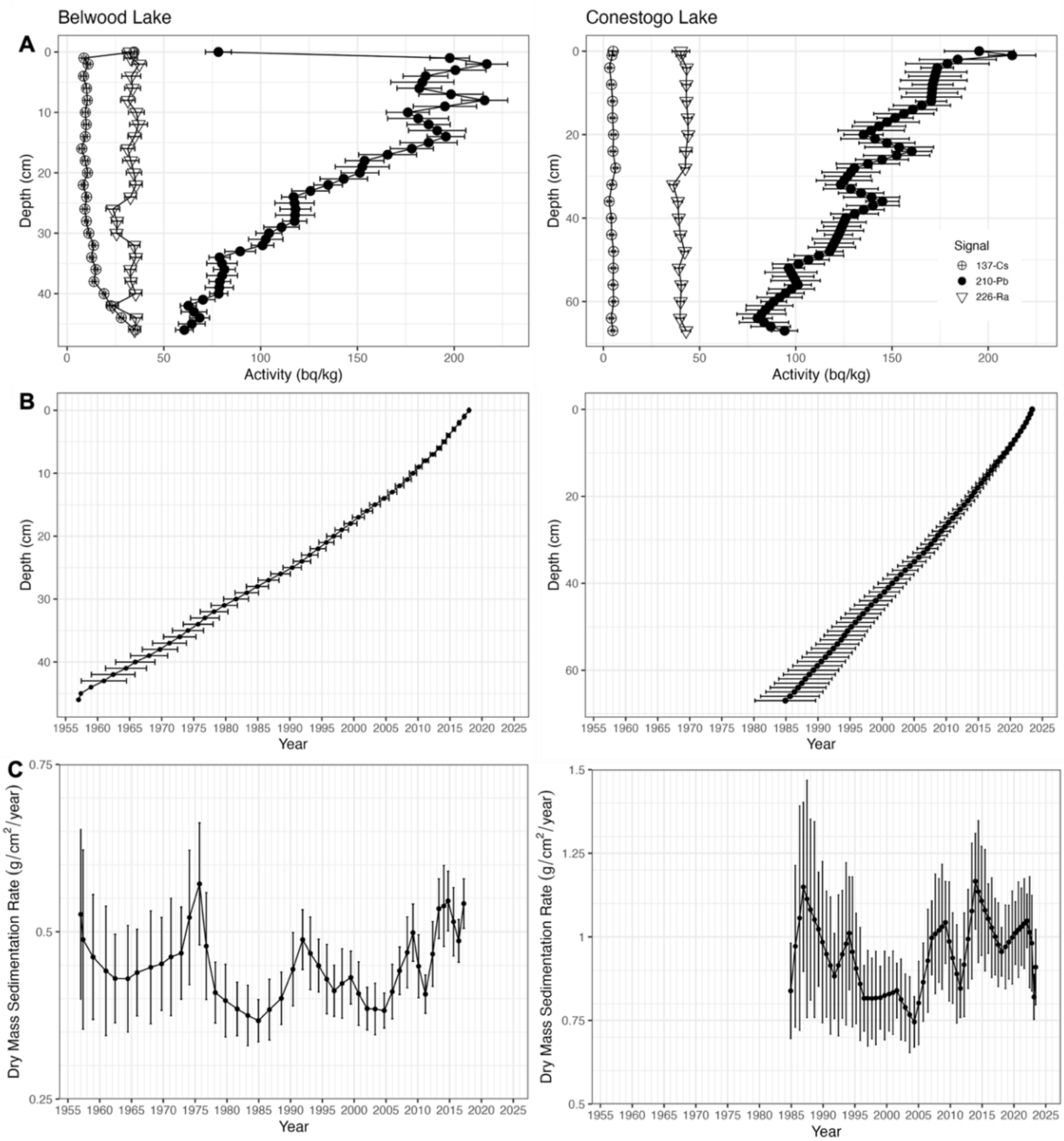

Figure 2.2: Radiometric dating of cores from Belwood Lake (left column) and Conestogo Lake (right column). Row A presents stratigraphic profiles for ^{210}Pb, ^{226}Ra, and ^{137}Cs activity (error bars display uncertainty as ±1 standard deviation unit). Row B presents the age vs depth relations based on ^{210}Pb dating (error bars display uncertainty as ±2 standard deviation units). Row C presents temporal variation in the estimated dry mass sedimentation rate (error bars display uncertainty as ±1 standard deviation unit).

3.2. Sediment composition

Stratigraphic variation of sediment composition was subtle in both reservoirs. Organic matter content was low and relatively constant throughout the cores from both reservoirs (Belwood Lake: ~8-13%; Conestogo Lake: ~7-10%), and the sediment was dominated by mineral matter (Belwood Lake: 63-75%; Conestogo Lake: 75-80%; Figure 2.3). Carbonates contributed 15-25% of the sediment at Belwood Lake and 10-15% at Conestogo Lake. Mineral matter content declined gradually through the core from Belwood Lake, and carbonate content increased (Figure 2.3).

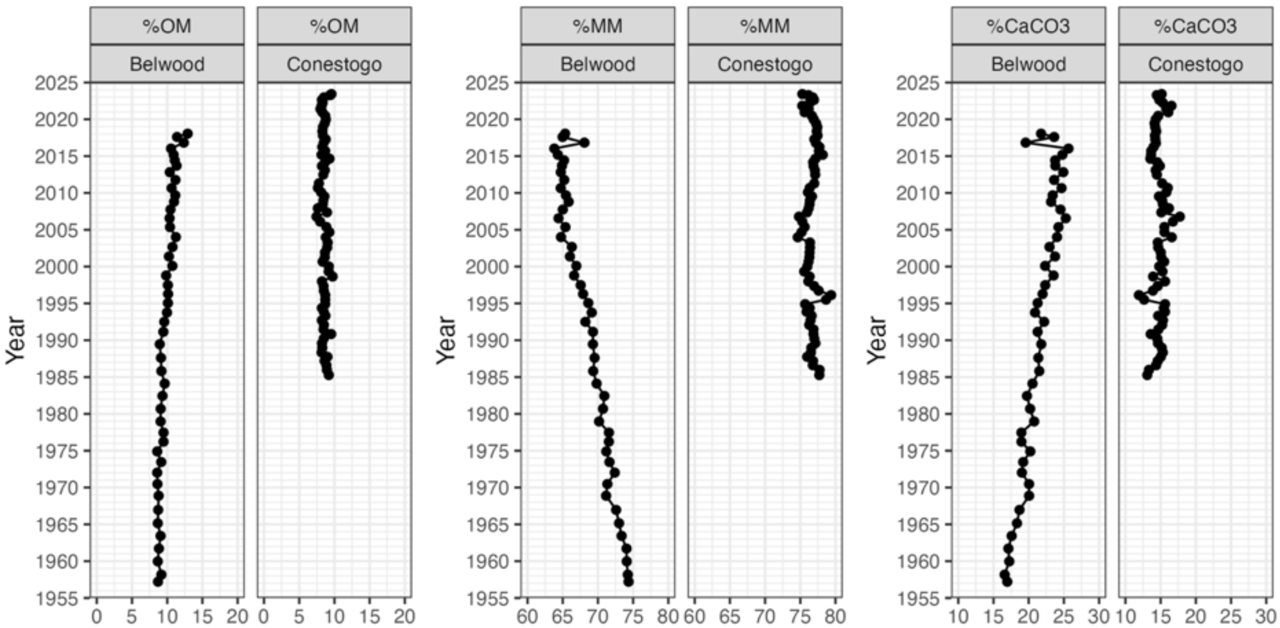

Figure 2.3: Stratigraphic variation in sediment composition in cores analyzed from Belwood Lake and Conestogo Lake. From left to right, graphs show organic matter content (%OM), carbonate-free mineral matter content (%MM), and carbonate content (%$CaCO_3$) by estimated year of deposition in each sediment core. Values are expressed as a percentage of dry sediment mass.

3.3. Microplastic concentration and flux

Sedimentary concentrations of MP particles were comparable in the cores from both reservoirs (Belwood Lake: ~64.2-507.8 particles g dw^{-1}; Conestogo Lake: ~187.9-563.9 particles g dw^{-1}), and values varied between adjacent samples without obvious trends (Figure 2.4). Fragments were typically more abundant than fibers in both reservoirs. The concentration of fragments declined after ~2005 at Belwood Lake, whereas they remained high at Conestogo Lake.

A single distinct peak of fiber abundance was observed in the sediments of both reservoirs (~2013 at Belwood Lake; ~2009 at Conestogo Lake).

Total flux of MPs fluctuated between ~ 6.7-68.0 particles cm^{-2} $year^{-1}$ in the core from Belwood Lake with no obvious trend over time, although a peak value of 156.5 particles cm^{-2} $year^{-1}$ occurred in the uppermost sample (Figure 2.4). Total MP flux in the core from Conestogo Lake varied ~ 30.4-98.4 particles cm^{-2} $year^{-1}$, which is roughly two-fold higher than total MP flux at Belwood Lake. The higher flux at Conestogo Lake is attributable to its higher sedimentation rate because total concentrations of MPs are comparable (Figures 2.2 and 2.4). A rising trend of total MP flux is apparent at Conestogo Lake since ~1985 associated with peaks that exceed 80 particles cm^{-2} $year^{-1}$ in ~2009, ~2011, ~2020, which consist mainly of fragments (Figure 2.4).

3.4. Composition of MP particles

Analysis of sediment core samples at approximately decadal time steps from both reservoirs by LDIR resulted in identification of particles consisting of 18 polymers. These included acryl butyl styrene (ABS), cellulose acetate, low density polyethylene (LDPE), polycarbonate (PC), polyethylene (PE), polyethylene terephthalate (PET), polylactic acid (PLA), polymethyl methacrylate (PMMA), polyamide (PA), polyoxymethylene (POM), polypropylene (PP), polystyrene (PS), polytetrafluoroethylene (PTFE), polyurethane (PU), polyvinylchloride (PVC), rubber (RB), styrene-acrylonitrile resin (SAN), and silica. Six of the polymers (LDPE, PMMA, silica, PC, PS, cellulose acetate) contributed less than 1% to the abundance or mass of a sample and were excluded from further analysis.

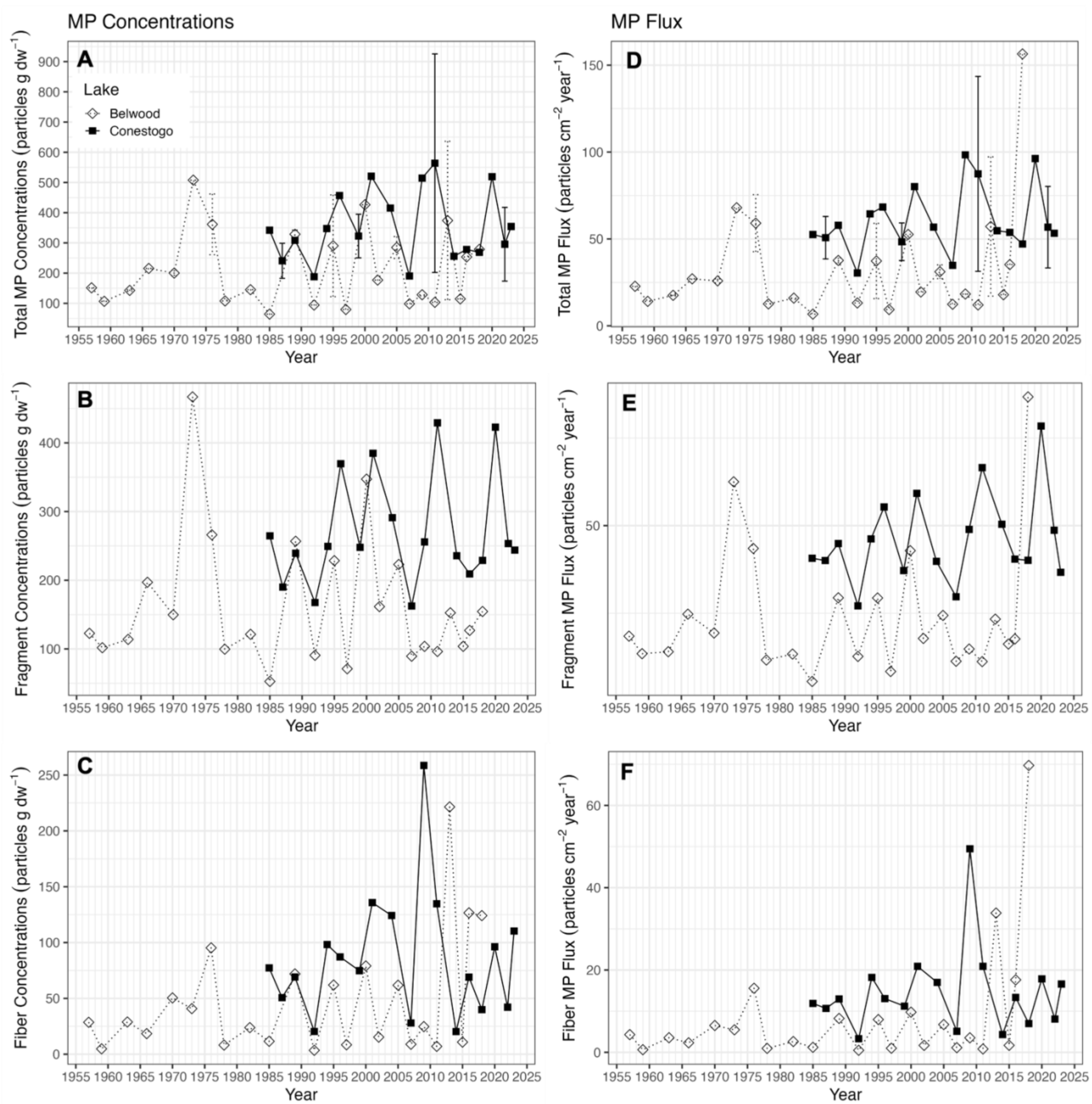

Figure 2.4: Stratigraphic variation in the concentration and flux of MP particles in sediment cores from Belwood Lake and Conestogo Lake. Graphs in the left column show [A] total concentration of MP particles, [B] concentration of MP fragments, and [C] concentration of MP fibers in particles per gram of dry sediment (particles g dw^{-1}) for both lakes. Graphs in the right column show [D] total flux of MP particles, [E] flux of MP fragments, and [F] flux of MP fibers. All flux estimates are corrected for sediment focusing and expressed as particles per square centimeter per year (particles cm^{-2} year^{-1}). Error bars represent the difference between duplicate samples processed at select intervals.

Composition of MPs, when based on particle abundance, was variable among samples in both reservoirs but three polymers tended to be numerically dominant (PA, RB, PE; Figure 2.5A). The same three polymers also had the highest relative mass (Figure 2.5B). The polymers ABS, SAN, and PET had higher relative mass in a few samples compared to their relative abundance (Figure 2.5A and B). Although this is partially due to higher densities of these polymers (ABS = ~1.05 g cm^{-3}, SAN = ~1.08 g cm^{-3}, PET = ~1.34 g cm^{-3}), larger than average particle size also plays a role. This includes ABS in samples deposited ~1966 and ~1976 in Belwood Lake, SAN in a sample deposited in ~2017 in Belwood Lake, and PET deposited in two samples deposited in ~2004 and ~2014 in Conestogo Lake. Conversely, particles consisting of PU and PTFE particles typically had lower relative mass (<2%) than relative abundance, suggesting they consisted of relatively small particle size as polymer densities of PU and PTFE are relatively high (PU = ~1.1 g cm^{-3}, PTFE = ~2.2 g cm^{-3}).

Relative abundance of the three most abundant polymers was highly variable among samples in both reservoirs without obvious trends over time (Figure 2.5C). Rubber may be an exception to this, as the relative abundance was generally higher in samples deposited since ~1990 (Figure 2.5C). The relative mass of PA declined after ~1980 at both reservoirs but the relative abundance did not (Figure 2.5C, D). This suggests that the average particle size of PA has increased since the 1980s. The relative abundance and relative mass of rubber particles have increased since ~1990 at both reservoirs, suggesting their supply has increased but with little to no change in average particle size.

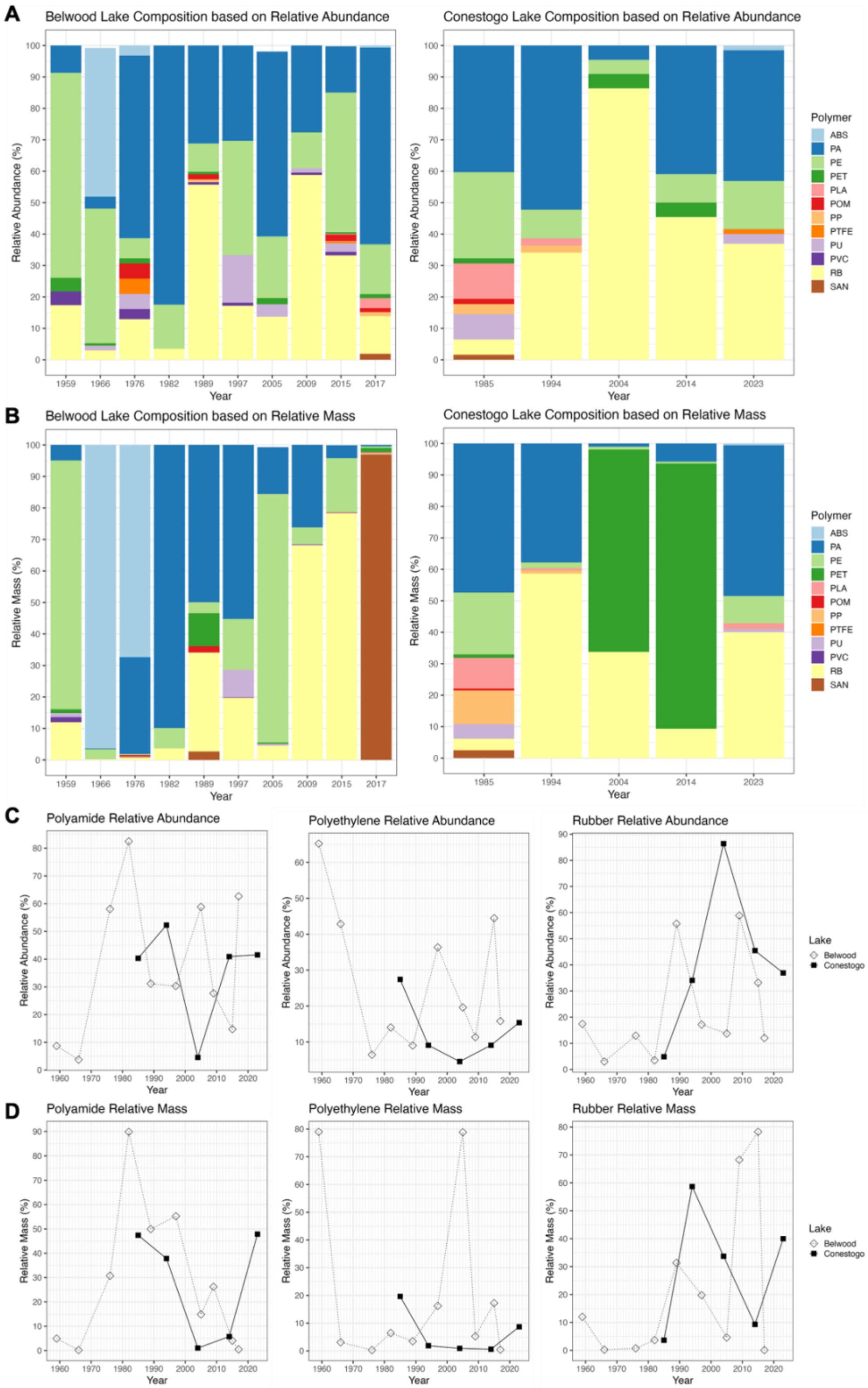

Figure 2.5: Temporal variation in MP composition, expressed as relative abundance and relative mass of particles at ~10-year intervals in sediments from Belwood Lake and Conestogo Lake. Row [A] shows relative abundance of the polymers that exceeded 1% in at least

one sample, row [B] displays relative mass of the same polymers, row [C] the relative abundance of the three most abundant polymers (polyamide, polyethylene, rubber), and row [D] shows the relative mass of the same three most abundant polymers.

4. Discussion

In Canada, plastic production has increased approximately 230-fold since the 1950s (Environment and Climate Change Canada, 2019), yet there is little to no evidence of a rising trend in the concentration, flux, and composition of MP particles in profundal sediment at Belwood Lake and Conestogo Lake, two headwater reservoirs within the upstream agricultural portion of the rapidly urbanizing watershed of the Grand River. Inter-sample variability of MP concentration, flux and composition was high in the stratigraphic profiles, and the concentrations were comparable in both reservoirs and dominated by the same three polymers (polyamide, polyethylene, rubber). The sedimentary concentrations varied between about 60 and 560 particles g dw^{-1}, with an average of 209.8 particles g dw^{-1} at Belwood Lake and 354.6 particles g dw^{-1} at Conestogo Lake (Figure 2.4A). These concentrations are high compared to values in aquatic sediment at other sites within the Laurentian Great Lakes and southern Ontario (Table 2.1). Differences among studies in sample processing, however, present challenge for comparisons of MP concentrations between sites. For example, previous studies have not used a consistent filter size, which means they enumerate different portions of the total pool of MP particles in aquatic sediment (Table 2.1). Most of them used a coarser minimum filter size (53-160 μm) than in our study (28 μm), which could lead to lower estimates of MP concentration than reported in this study because MP abundance typically increases with decreasing particle size (Hale et al., 2020). Fortunately, equations exist that can improve the comparability by converting MP concentrations generated using different filter sizes to estimates of concentrations that would be determined with the 28-500 μm size range assessed in our study (Koelmans et al., 2020). When the equations are applied, concentrations of MP particles in sediment in Conestogo Lake and Belwood Lake remain higher than those in nearshore, tributary, and beach sediments of Lake Erie and Lake Ontario (Ballent et al., 2016; Dean et al., 2018; Lenaker et al., 2021). The concentrations of MP particles determined in the two GRW reservoirs, however, are comparable to the range of concentrations determined in offshore sediments in Lake Huron (24.6-380.9 particles g dw^{-1}) and Lake Ontario (11.2-664.2 particles g dw^{-1}) (Belontz et al., 2024). This suggests that depositional zones with fine grained sediments, such as deep-water basins in the Great Lakes and reservoirs within the GRW, accumulate high-concentrations of MP particles, as has been measured elsewhere in fine-grained aquatic sediments within depositional zones (Tibbets et al., 2018; Enders et al., 2019; Corcoran et al., 2020; Wilson et al., 2021; Belontz et al., 2022).

Table 2.1: Comparison of MP particle concentrations reported in sediment from sites within the Laurentian Great Lakes and southern Ontario.

Study	Location	Sampling Location	Size Range (μm)	Conversion Factor	Concentration Reported	Converted to 28-500 μm (particles g dw^{-1})
Dean et al., 2018	Lake Erie	Nearshore, tributary, beach	63-2000	1.53	0-391 particles kg dw^{-1} (nearshore) 10-462 particles kg dw^{-1} (tributary) 50-146 particles kg dw^{-1} (beach)	0-0.59 (nearshore) 0.02-0.71 (tributary) 0.08-0.22 (beach)
Belontz et al., 2022	Lake Huron	Nearshore, offshore	63-2000	1.53	59-335 714 particles kg dw^{-1}	0.09-513.6
Ballent et al., 2016	Lake Ontario	Nearshore, tributary, beach	63-2000	1.53	40-27 830 particles kg dw^{-1}	0.06-42.6
Lenaker et al., 2021	Lake Erie	Nearshore, offshore	125-355	4.34	110-3200 particles kg dw^{-1}	0.48-13.9
Belontz et al., 2024	Lake Huron	Offshore	53-2000	1.36	18.1-280.1 particles g dw^{-1}	24.6-380.9
Belontz et al., 2024	Lake Ontario	Offshore	53-2000	1.36	8.2-488.4 particles g dw^{-1}	11.2-664.2

Absence of a rising trend in the concentration and flux of MP particles in sediment at Conestogo Lake and Belwood Lake was unexpected, because studies elsewhere have shown stratigraphic patterns in MP deposition that align with the rise of plastic production since the 1950s (Brandon et al., 2019; Simon-Sanchez et al., 2022; Our World in Data, 2023). This unexpected result could potentially be explained by improvements in plastic waste management and environmental conservation (i.e., erosion control) during the past several decades, in tandem with minimal land use changes in this region of the GRW as it remains primarily rural (Stewardship Ontario, 2013; Conservation Authorities Act, 1990). Additionally, most MPs identified in these reservoirs were secondary MPs (fragments, fibers) produced through the weathering of larger plastic waste. As generation of MPs from this larger plastic waste is influenced by many factors, such as polymer property or environmental conditions, a clear trend in association with global plastic production may be challenging to detect.

MP particles may migrate downward through the sediment column of lakes, which may compromise the use of sediment cores to track time trends in MP deposition (Dimante-Deimantovica et al., 2024). The processes that promote post-depositional migration of MPs in lake sediment, however, are not fully understood (Bancone et al., 2020). Leiser et al. (2021) suggested that aggregation of MPs to iron-organo flocs facilitates their downward migration. Bioturbation of sediment by benthic invertebrates is known to promote downward movement of MPs and other materials (Frank et al., 2023). MPs are more prone to downward migration when sediment grain size is large and MP particles are small and spherical rather than rod-shaped (Waldschager & Schuttrumf, 2020; Gao et al., 2021). Sediment core collection with open-barrel (i.e., gravity) corers and subsequent vertical sectioning, which are commonly employed by paleolimnological studies of human impacts on aquatic ecosystems, may also promote downward movement of MPs through the water space that occurs between the sediment and the inner core tube wall. From our experience, sediment consisting of fine-grained organic-rich sediment (i.e., gyttja) generates the most space between the sediment core and inner core tube wall due to its gelatinous consistency. This phenomenon allows for MPs that may be at highest concentrations in upper strata to migrate through the water space to lower strata, including strata deposited before industrial plastics production began. This could have occurred in the study by Dimante-Deimantovica et al. (2024) and may account for the detection of low concentrations of MP particles in sediment intervals deposited before 1950. Our sediment cores did not capture sediment that was deposited before plastics were produced, which makes it challenging to assess if the above processes may have confounded our stratigraphic records of MP deposition. However, the profundal sediments that were sampled from Belwood Lake and Conestogo Lake are fine-grained (clays and silts), highly minerogenic, and low in organic matter. Also, periodic deep-water anoxia occurs in these reservoirs which leads to a lack of sediment dwelling organisms and low rates of bioturbation. Although benthic invertebrates like chironomids and oligochaetes can be abundant in anoxic conditions, we did not observe any of these organisms in the sediment cores from Belwood and Conestogo lakes. Additionally, detectable decreasing ^{7}Be activity within the upper 2-cm of the sediment core from Conestogo Lake suggests sediment mixing was not substantial enough to account for an absence of the rising trend over time in the cores. Collectively, these factors are likely to minimize downward migration of MP particles through the sediment profiles we analyzed from Belwood Lake and Conestogo Lake, which adds confidence that the stratigraphic records are not strongly influenced by confounding factors. Consistent with this, stratigraphic profiles of MP abundance in sediment cores from Lake Huron and Lake Ontario also do not reveal a rising trend since the 1950s (Belontz et al, 2024). Instead, they display plateaus and declines in MP abundance which the authors suggest coincide with economic events that have affected plastic production in Canada (Belontz et al., 2024). For example, declines in MP abundance after 2010 (Lake Ontario) and 2012 (Lake Huron) were interpreted as reflecting influence of the 2007-2008 Global Financial Crisis that caused a drop in global plastic production during the late 2000s (Our World in Data, 2023). The marked decline in MP concentration observed after ~2011 (Conestogo Lake) and ~2013 (Belwood Lake) in our study coincides with the declines observed at Lake Huron and Lake Ontario (Figure 2.4; Belontz et al., 2024), suggesting sediments in the GRW reservoirs track to some degree influence of variation in global plastic production. Research on isolated lakes located in smaller watersheds, however, has revealed that stratigraphic profiles of MP concentration and flux typically are more strongly influenced by plastics usage at a regional scale and local hydrologic processes compared to sediment records from marine environments or basins in larger catchments which tend to reflect global patterns of plastic production (Turner et al., 2019; Bancone et al., 2020; Almas et al., 2022).

At Belwood Lake and Conestogo Lake, increases since the early 1980s in the relative abundance and relative mass of MP particles composed of rubber likely reflect the influence of processes operating at a local to regional scale (Figure 2.5). Rubber particles are often generated through mechanical abrasion of tires by vehicular traffic on paved roads (Sieber et al., 2020). Tire road wear particles (TRWPs) have been identified as a substantial source of MPs to the environment (Kole et al., 2017). The rise in relative abundance and relative mass of rubber MP particles in both reservoirs, thus, may be a consequence of the steady increase in paved roads in rural southern Ontario that occurred between 1975 and 1995 (Fenech et al., 2005), which promoted greater traffic volumes and driving speeds—factors known to increase TRWP generation (Rodland et al., 2023). Additionally, low permeability of pavement increases the delivery of TRWPs into nearby aquatic environments via runoff (Kole et al., 2017; Wagner et al., 2018; Rasmussen et al., 2023). Rural roads often lack infrastructure for runoff collection, which promotes delivery of TRWPs to nearby water bodies (Wagner et al., 2018), and it has been reported that Belwood and Conestogo lakes capture substantial water volumes of road-generated runoff from within their watersheds (Loomer & Cooke, 2011). Given that tire rubber accounts for nearly 30% of the MP particles (and ~25% of the total mass) at these reservoirs, effective mitigation of MP supply to aquatic environments could be achieved through better management of runoff from roads.

Microplastics have accumulated in these reservoirs for decades, and the stratigraphic records generated during our study support quantification of the total mass of MPs that have become stored in the bottom sediment of the profundal zone during the time intervals captured by the sediment cores. Using the average volume (1.72×10^{-6} cm^3) and density (1.19 g cm^{-3}) of the MP particles enumerated and the average depositional area of each reservoir (5.29 km^2 for Belwood Lake, 4.73 km^2 for Conestogo Lake; estimated using Google Earth historical imagery), we estimate that 84.4 tonnes of MPs have accumulated in the fine-grained bottom sediment at Belwood Lake since 1957, and 105.7 tonnes of MPs at Conestogo Lake since 1985. At Belwood Lake, we estimate that 55.9 tonnes of MPs have accumulated since 1985, which is approximately two-fold lower than at Conestogo Lake over the same time interval. When correcting for differences in surface area between the two reservoirs, 10.6 tonnes km^{-2} of MPs have been deposited since ~1985 at Belwood Lake compared to 22.4 tonnes km^{-2} of MPs in Conestogo Lake. Given that the sedimentary concentrations are comparable among the two reservoirs, the two-fold difference between these values corresponds roughly with the two-fold difference in the median sedimentation rate at these reservoirs since ~1985 (0.45 g cm^{-2} $year^{-1}$ in Belwood Lake; 0.98 g cm^{-2} $year^{-1}$ in Conestogo Lake). These findings reinforce that the supply of MPs may be reduced to the reservoirs via better control of sediment delivery to the reservoirs, and they suggest that greater improvements may be possible at Conestogo Lake given its higher sedimentation rate.

Pervasive contamination of landscapes across the Laurentian Great Lakes watershed has stimulated a recent call for long-term monitoring programs capable of tracking changes in the abundance and composition of MPs in water and sediment and informing management strategies (Hataley et al., 2023; McIlwraith et al., 2023). The estimates of the concentration, flux, and composition of MPs in sediment cores from Belwood Lake and Conestogo Lake provided by this study may serve as important benchmarks which can be used for comparison to future monitoring data in the Laurentian Great Lakes. Such benchmark data can be used to evaluate for emergence of trends in the future and to evaluate the success of mitigative actions, and they are needed for the Grand River and other substantial rivers flowing into the Great Lakes, because they have been identified as important pathways of MP delivery to the Great Lakes (Baldwin et al., 2016; Corcoran et al., 2015, 2020). We note, however, that the two reservoirs in this study are located upstream of the main urban centers within the GRW. Thus, the data from these reservoirs likely reflect contributions from rural and agricultural portions of the landscape and may underestimate the flux of MPs from urban centers within the GRW to downstream aquatic ecosystems. Unfortunately, there are few reservoirs and lakes in the GRW that can be sampled downstream of the urban centers. In the absence of these types of basins, we advocate for the analysis of sediments in urban storm-retention ponds and the nearshore zone of Lake Erie adjacent to the mouth of the Grand River, combined with use of modelling approaches, to add insight into the flux of MPs from urban landcover within the GRW and aquatic ecosystems located downstream, including Lake Erie.

Chapter 3:
Synthesis and Recommendations

As explored in Chapter 2, temporal trends of MP abundance, flux, and composition in Belwood Lake and Conestogo Lake sediment were surprisingly subtle, despite known changes in plastic production and use since the 1950s. Although concentrations and fluxes of MPs in Belwood Lake and Conestogo Lake have not increased substantially through time, there has been a steady delivery of MPs to reservoir sediments for decades, which has resulted in several tonnes of plastic stored in the bottom sediment. MPs were identified in sediments of both reservoirs at the start of each record (~1957 in Belwood Lake, ~1985 in Conestogo Lake), at concentrations ranging from ~50-550 particles g dw^{-1}, which is comparable to values determined by other studies in the Great Lakes basin. Despite similar concentrations between reservoirs, MP flux was higher in Conestogo Lake than Belwood Lake by approximately two-fold due to more rapid delivery of sediment. Composition of MP particles was variable, but MPs identified as rubber increased since ~1990, which coincides with an increase of paved roads upstream of the reservoirs that facilitate delivery of sediment and MPs derived from vehicular tire wear. Initiatives exist in the Grand River watershed to mitigate processes delivering sediment and associated contaminants to local water bodies (i.e., control of erosion, surface runoff), such as the Rural Water Quality Program (Holeton, 2013; Grand River Conservation Authority, 2024). Continued application of these initiatives will help decrease the delivery of MPs to waterbodies in rural regions. In addition to local initiatives, these findings contribute towards the growing body of literature characterizing time trends of MP deposition across the Laurentian Great Lakes region. Currently, a long-term MP monitoring framework is in development for the Laurentian Great Lakes by a working group for submission to the International Joint Commission (IJC)'s Great Lakes Science Advisory Board (IJC Great Lakes Science Advisory Board, 2024). Reconstruction of past trends in MP pollution over space and time within this region can be used to inform the design of a monitoring program and improve our ability to anticipate future changes in MP pollution across the Laurentian Great Lakes basin.

This research reported in this thesis has contributed to the Microplastics Fingerprinting Project at the University of Waterloo by advancing several objectives. The primary role of this research in the larger project was to reconstruct past changes in MP deposition using sediment core analysis (Objective 5). Additionally, this study has contributed to the assessment of factors influencing MPs sources and entry pathways along the river to lake continuum (Objective 2). For example, the findings support that MPs consisting of rubber have increased since the 1990s, likely via runoff from paved roads. However, the study sites are located upstream of urban centers and, thus, could not contribute new knowledge about temporal trends caused by urban activities which are known to be substantial sources of MP emissions (Grbic et al., 2020). To determine releases of MPs to the GRW from urban centres, others in the Microplastics Fingerprinting Project are assessing inputs, outputs, and retention of MPs stormwater ponds in the cities of Kitchener and Waterloo. Additionally, collection of sediment cores from the nearshore region of Lake Erie where the Grand River discharges into the lake may be helpful to evaluate MP supply downstream of urban centers; an endeavour currently in progress by the Ecohydrology Research Group at the University of Waterloo. Lastly, mass flux data generated by this study will be used to assess the accuracy of MP mass balance models that will be used to quantitatively link the sources, transport, and fate of MPs at the watershed scale (Objective 3).

Future research could analyze more samples from the Belwood Lake and Conestogo Lake sediment cores using LDIR spectroscopy to generate a higher temporal resolution data to characterize variation in the composition of MPs. Existing MP composition data from Belwood Lake and Conestogo Lake provide approximately decadal resolution, as the facility for LDIR spectroscopy utilized in this analysis was newly established. Consequently, a limited number of samples were able to be included in this thesis. Further spectroscopic analysis may reveal additional time trends in MP composition in these reservoirs. This may also allow for use of spectroscopically generated count data, which were not used in this thesis. Although most existing studies similarly use manual counting, emerging research suggests spectroscopic methods may the throughput, and possibly also the accuracy, of MP quantification (Song et al., 2015; Cheng et al., 2022).

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Appendix: Raw Data

Below, I present the raw data for key measurements made in MSc Thesis research.

Table A1: Results of loss-on-ignition analyses on the sediment core from Belwood Lake, as generated in 2018.

Mid-Point Depth (cm)	% Water	% Organic	% MM	% Carbonate	% Non-Carbonate MM
0.5	80.94	12.90	87.10	21.76	65.34
1.5	72.57	11.40	88.60	23.62	64.98
2.5	71.96	12.35	87.65	19.57	68.08
3.5	68.02	10.54	89.46	25.66	63.80
4.5	65.70	10.91	89.09	24.79	64.30
5.5	64.49	11.06	88.94	23.76	65.18
6.5	64.90	11.33	88.67	23.79	64.89
7.5	67.42	10.36	89.64	24.92	64.72
8.5	66.14	11.17	88.83	23.62	65.21
9.5	66.10	10.62	89.38	24.67	64.71
10.5	64.47	11.14	88.86	23.41	65.45
11.5	65.30	10.97	89.03	23.19	65.84
12.5	63.79	10.46	89.54	24.52	65.01
13.5	66.29	10.33	89.67	25.27	64.40
14.5	63.03	10.39	89.61	24.23	65.38
15.5	63.09	11.23	88.77	24.03	64.75
16.5	63.35	10.75	89.25	22.96	66.29
17.5	60.67	10.24	89.76	23.74	66.02
18.5	60.76	10.73	89.27	22.34	66.93
19.5	57.34	9.85	90.15	23.54	66.61
20.5	62.97	10.10	89.90	22.35	67.55
21.5	60.89	10.13	89.87	21.98	67.89
22.5	59.07	10.10	89.90	21.27	68.63

23.5	58.51	9.97	90.03	20.91	69.12
24.5	57.02	9.58	90.42	22.20	68.22
25.5	56.11	9.45	90.55	21.27	69.29
26.5	53.93	8.94	91.06	21.79	69.28
27.5	52.63	9.11	90.89	21.37	69.52
28.5	58.71	9.17	90.83	21.51	69.32
29.5	53.52	9.64	90.36	20.55	69.81
30.5	55.14	9.32	90.68	19.76	70.91
31.5	53.63	9.07	90.93	20.21	70.72
32.5	53.36	9.06	90.94	20.78	70.16
33.5	55.51	9.47	90.53	18.97	71.56
34.5	51.94	9.47	90.53	18.98	71.55
35.5	50.89	8.58	91.42	20.23	71.19
36.5	53.96	9.15	90.85	19.22	71.64
37.5	50.58	8.58	91.42	19.04	72.39
38.5	51.46	8.61	91.39	20.05	71.34
39.5	49.90	8.78	91.22	20.06	71.16
40.5	51.34	8.72	91.28	18.70	72.58
41.5	49.52	8.66	91.34	18.34	73.00
42.5	50.79	9.05	90.95	17.59	73.36
43.5	50.51	8.83	91.17	17.11	74.06
44.5	47.76	8.66	91.34	17.25	74.09
45.5	45.33	9.17	90.83	16.59	74.24
46.5	44.47	8.69	91.31	16.96	74.35

Table A2: Water content of the sediment core from Belwood Lake, as generated in this study.

Mid-Point Depth (cm)	% Water
0.5	64.07
1.5	60.48
2.5	68.42
3.5	65.91
4.5	61.60
5.5	68.12
6.5	66.45
7.5	60.55
8.5	58.53
9.5	59.57
10.5	59.31
11.5	58.89
12.5	61.30
13.5	58.18
14.5	54.03
15.5	63.51
16.5	58.64
17.5	57.51
18.5	64.21
19.5	60.83
20.5	60.26
21.5	50.84
22.5	60.04
23.5	57.98
24.5	54.56
25.5	50.13
26.5	57.57

27.5	55.06
28.5	52.54
29.5	53.23
30.5	53.50
31.5	51.77
32.5	53.38
33.5	55.09
34.5	52.21
35.5	50.66
36.5	52.38
37.5	53.11
38.5	51.16
39.5	50.48
40.5	48.41
41.5	48.73
42.5	47.69
43.5	46.33
44.5	44.09
45.5	44.53
46.5	41.65
47.5	42.14
48.5	38.02

Table A3: Results of loss-on-ignition analyses the sediment core from Conestogo Lake.

Mid-Point Depth (cm)	% Water	% Organic	% MM	% Carbonate	% Non-Carbonate MM
0.50	94.81	9.57	90.43	15.14	75.28
1.50	76.09	9.39	90.61	14.46	76.15
2.50	70.63	8.52	91.48	14.73	76.75
3.50	69.86	8.23	91.77	14.82	76.95
4.50	66.24	8.33	91.67	15.30	76.37
5.50	64.58	8.19	91.81	16.54	75.27
6.50	64.76	8.01	91.99	15.80	76.19
7.50	62.48	8.30	91.70	16.09	75.61
8.50	62.40	8.69	91.31	14.66	76.65
9.50	60.71	8.79	91.21	14.30	76.91
10.50	60.89	8.62	91.38	14.14	77.23
11.50	60.17	8.37	91.63	14.22	77.41
12.50	59.27	8.31	91.69	14.36	77.33
13.50	59.36	8.37	91.63	14.19	77.44
14.50	59.38	8.77	91.23	14.23	77.00
15.50	58.91	8.46	91.54	14.31	77.23
16.50	57.26	8.29	91.71	14.00	77.71
17.50	57.29	8.69	91.31	13.67	77.64
18.50	56.99	8.18	91.82	13.64	78.18
19.50	57.86	9.30	90.70	13.54	77.16
20.50	57.84	8.67	91.33	14.50	76.82
21.50	57.73	8.24	91.76	14.88	76.88
22.50	58.07	8.64	91.36	14.29	77.06
23.50	57.92	8.42	91.58	14.47	77.11
24.50					
25.50	58.85	7.80	92.20	15.22	76.98

26.50	56.56	7.65	92.35	16.01	76.34
27.50	57.41	8.12	91.88	15.80	76.08
28.50	57.43	8.58	91.42	14.77	76.65
29.50	57.07	8.43	91.57	15.21	76.36
30.50	56.68	8.37	91.63	15.33	76.31
31.50	55.99	7.64	92.36	16.18	76.18
32.50	56.71	8.94	91.06	15.10	75.95
33.50	56.33	7.46	92.54	17.73	74.81
34.50	54.43	7.98	92.02	16.75	75.28
35.50	55.60	8.86	91.14	15.53	75.61
36.50	56.47	9.28	90.72	15.54	75.18
37.50	56.96	8.78	91.22	16.58	74.64
38.50	58.35	9.03	90.97	14.60	76.37
39.50	57.02	8.97	91.03	14.66	76.37
40.50	57.84	8.66	91.34	15.02	76.32
41.50	57.60	8.64	91.36	15.07	76.29
42.50	58.11	8.33	91.67	15.50	76.17
43.50	57.41	9.20	90.80	14.81	75.99
44.50	56.37	9.19	90.81	15.26	75.55
45.50	57.03	9.75	90.25	13.93	76.32
46.50	58.03	8.24	91.76	15.63	76.14
47.50	58.42	8.46	91.54	14.58	76.96
48.50	58.12	8.50	91.50	13.90	77.60
49.50	56.30	8.70	91.30	11.92	79.38
50.50	55.55	8.70	91.30	12.65	78.65
51.50	57.34	8.69	91.31	15.63	75.68
52.50	59.59	8.22	91.78	15.44	76.33
53.50	60.36	8.49	91.51	15.62	75.89
54.50	55.94	8.73	91.27	14.66	76.61
55.50	56.28	8.22	91.78	15.33	76.45

56.50	55.62	8.51	91.49	15.22	76.27
57.50	53.76	8.45	91.55	14.68	76.87
58.50	53.83	9.54	90.46	13.59	76.87
59.50	52.97	8.48	91.52	14.53	76.99
60.50	52.91	8.24	91.76	14.63	77.14
61.50	51.82	8.28	91.72	15.16	76.56
62.50	51.87	8.19	91.81	15.31	76.51
63.50	52.18	9.01	90.99	15.02	75.97
64.50	51.52	8.60	91.40	14.59	76.81
65.50	51.39	8.87	91.13	14.36	76.78
66.50	51.02	8.93	91.07	13.34	77.73
67.50	49.44	9.20	90.80	13.10	77.70

Table A4: Results of radiometric analysis on the sediment core from Belwood Lake, including modelled ages.

Top depth (cm)	Bottom depth (cm)	Mid-Point Depth (cm)	^{210}Pb Activity (dpm g^{-1})	^{137}Cs Activity (dpm g^{-1})	^{226}Ra Activity (Weighted mean of ^{214}Bi & ^{214}Pb Activity) (dpm g^{-1})	CRS Dates with Linear Extrapolation	Mid-Depth Point CRS Dates with Linear Extrapolation	Dry Mass Sedimentation Rate (g cm^{-2} $year^{-1}$)
0	1	0.5	4.69	2.08	1.95	2017.97	2018.05	*1.96*
1	2	1.5	11.86	0.52	1.97	2017.23	2017.60	0.54
2	3	2.5	13.01	0.65	2.24	2016.43	2016.83	0.49
3	4	3.5	12.04			2015.62	2016.03	0.51
4	5	4.5	11.11	0.51	1.99	2014.76	2015.19	0.55
5	6	5.5	11.01			2014.10	2014.43	0.54
6	7	6.5	10.92	0.60	2.03	2013.31	2013.70	0.53
7	8	7.5	11.90			2012.34	2012.82	0.47
8	9	8.5	12.94	0.63	1.88	2011.20	2011.77	0.41
9	10	9.5	11.71			2010.12	2010.66	0.45
10	11	10.5	10.56	0.56	2.15	2009.24	2009.68	0.50
11	12	11.5	10.88			2008.35	2008.80	0.45
12	13	12.5	11.21	0.59	2.22	2007.14	2007.75	0.44
13	14	13.5	11.47			2005.99	2006.56	0.41
14	15	14.5	11.74	0.56	2.09	2004.73	2005.36	0.38
15	16	15.5	11.21			2003.29	2004.01	0.38
16	17	16.5	10.69	0.46	1.88	2002.04	2002.67	0.39
17	18	17.5	9.94			2000.70	2001.37	0.41
18	19	18.5	9.22	0.56	1.97	1999.48	2000.09	0.43
19	20	19.5	9.15			1998.10	1998.79	0.42

20	21	20.5	9.08	0.63	2.07	1996.86	1997.48	0.41
21	22	21.5	8.58			1995.70	1996.28	0.43
22	23	22.5	8.09	0.50	2.13	1994.41	1995.05	0.45
23	24	23.5	7.55			1993.11	1993.76	0.47
24	25	24.5	7.02	0.61	1.97	1991.90	1992.50	0.49
25	26	25.5	7.05			1990.38	1991.14	0.44
26	27	26.5	7.08	0.55	1.42	1988.55	1989.46	0.40
27	28	27.5	7.07			1986.68	1987.61	0.38
28	29	28.5	7.05	0.60	1.55	1984.95	1985.81	0.37
29	30	29.5	6.65			1983.28	1984.11	0.38
30	31	30.5	6.26	0.67	1.53	1981.61	1982.44	0.38
31	32	31.5	6.16			1979.77	1980.69	0.40
32	33	32.5	6.06	0.82	2.08	1978.17	1978.97	0.41
33	34	33.5	5.36			1976.75	1977.46	0.48
34	35	34.5	4.73	0.76	2.13	1975.69	1976.22	0.57
35	36	35.5	4.80			1974.10	1974.90	0.52
36	37	36.5	4.88	0.89	1.96	1972.82	1973.46	0.47
37	38	37.5	4.80			1971.21	1972.02	0.46
38	39	38.5	4.73	0.83	1.97	1969.73	1970.47	0.45
39	40	39.5	4.72			1968.03	1968.88	0.45
40	41	40.5	4.70	1.14	2.13	1965.88	1966.96	0.44
41	42	41.5	4.21			1964.43	1965.15	0.43
42	43	42.5	3.76	1.35	1.41	1962.43	1963.43	0.43
43	44	43.5	3.93			1960.98	1961.71	0.44
44	45	44.5	4.11	1.68	2.14	1958.96	1959.97	0.46
45	46	45.5	3.87			1957.39	1958.17	0.49
46	47	46.5	3.63	2.11	2.09	1957.03	1957.21	0.53

Table A5: Results of radiometric analysis on the sediment core from Conestogo Lake, including modelled ages.

Top depth (cm)	Bottom depth (cm)	Mid-Point Depth (cm)	^{210}Pb Activity (dpm g^{-1})	^{137}Cs (dpm g^{-1})	^{226}Ra Activity (Weighted mean of ^{214}Bi & ^{214}Pb Activity) (dpm g^{-1})	CRS Dates with Linear Extrapolation	Mid-Depth Point CRS Dates with Linear Extrapolation	Dry Mass Sedimentation Rate (g cm^{-2} $year^{-1}$)
0	1	0.5	11.72	0.30	2.42	2023.41	2023.43	0.91
1	2	1.5	12.75	0.27	2.42	2023.15	2023.28	0.82
2	3	2.5	11.05			2022.81	2022.98	0.98
3	4	3.5	10.73			2022.46	2022.64	1.01
4	5	4.5	10.41	0.19	2.60	2022.06	2022.26	1.05
5	6	5.5	10.37			2021.62	2021.84	1.03
6	7	6.5	10.34			2021.18	2021.40	1.03
7	8	7.5	10.30			2020.71	2020.95	1.02
8	9	8.5	10.27	0.25	2.59	2020.22	2020.47	1.01
9	10	9.5	10.26			2019.73	2019.98	0.99
10	11	10.5	10.24			2019.20	2019.46	0.98
11	12	11.5	10.23			2018.67	2018.94	0.97
12	13	12.5	10.22	0.28	2.59	2018.06	2018.37	0.96
13	14	13.5	9.93			2017.53	2017.80	0.98
14	15	14.5	9.65			2017.04	2017.28	1.00
15	16	15.5	9.37			2016.51	2016.78	1.03
16	17	16.5	9.10	0.28	2.63	2015.97	2016.24	1.05
17	18	17.5	8.84			2015.45	2015.71	1.08
18	19	18.5	8.60			2014.91	2015.18	1.11
19	20	19.5	8.35			2014.40	2014.65	1.14

20	21	20.5	8.11	0.32	2.65	2013.94	2014.17	1.17
21	22	21.5	8.47			2013.39	2013.66	1.08
22	23	22.5	8.84			2012.80	2013.09	0.99
23	24	23.5	9.22			2012.17	2012.48	0.92
24	25	24.5	9.62	0.30	2.57	2011.58	2011.88	0.85
25	26	25.5	9.14			2011.01	2011.30	0.89
26	27	26.5	8.69			2010.37	2010.69	0.94
27	28	27.5	8.25			2009.82	2010.09	0.99
28	29	28.5	7.83	0.38	2.55	2009.24	2009.53	1.04
29	30	29.5	7.71			2008.71	2008.98	1.03
30	31	30.5	7.61			2008.21	2008.46	1.02
31	32	31.5	7.51			2007.64	2007.92	1.01
32	33	32.5	7.40	0.26	2.18	2007.08	2007.36	0.99
33	34	33.5	7.71			2006.48	2006.78	0.93
34	35	34.5	8.03			2005.74	2006.11	0.86
35	36	35.5	8.36			2005.03	2005.38	0.80
36	37	36.5	8.70	0.18	2.32	2004.34	2004.68	0.75
37	38	37.5	8.40			2003.61	2003.98	0.77
38	39	38.5	8.12			2002.92	2003.27	0.79
39	40	39.5	7.83			2002.25	2002.59	0.81
40	41	40.5	7.56	0.25	2.35	2001.59	2001.92	0.84
41	42	41.5	7.50			2000.98	2001.28	0.83
42	43	42.5	7.44			2000.37	2000.67	0.83
43	44	43.5	7.39			1999.67	2000.02	0.82
44	45	44.5	7.33	0.25	2.40	1999.00	1999.33	0.82
45	46	45.5	7.26			1998.30	1998.65	0.82
46	47	46.5	7.19			1997.68	1997.99	0.82
47	48	47.5	7.13			1997.03	1997.36	0.82
48	49	48.5	7.05	0.32	2.54	1996.44	1996.73	0.82
49	50	49.5	6.72			1995.83	1996.13	0.86

50	51	50.5	6.40			1995.19	1995.51	0.91
51	52	51.5	6.08			1994.61	1994.90	0.96
52	53	52.5	5.78	0.31	2.35	1994.12	1994.37	1.01
53	54	53.5	5.85			1993.62	1993.87	0.98
54	55	54.5	5.92			1993.02	1993.32	0.95
55	56	55.5	5.99			1992.41	1992.71	0.91
56	57	56.5	6.06	0.30	2.43	1991.78	1992.09	0.88
57	58	57.5	5.87			1991.16	1991.47	0.91
58	59	58.5	5.68			1990.53	1990.84	0.95
59	60	59.5	5.49			1989.90	1990.21	0.98
60	61	60.5	5.31	0.32	2.41	1989.28	1989.59	1.02
61	62	61.5	5.18			1988.65	1988.97	1.05
62	63	62.5	5.05			1988.03	1988.34	1.08
63	64	63.5	4.93			1987.47	1987.75	1.11
64	65	64.5	4.80	0.25	2.39	1986.90	1987.19	1.15
65	66	65.5	5.00			1986.32	1986.61	1.06
66	67	66.5	5.22			1985.64	1985.98	0.97
67	68	67.5	5.65	0.29	2.58	1984.91	1985.27	0.84

Table A6: Results of manual analysis and counting of microplastic particles in the sediment core from Belwood Lake.

Sample	Fragment Counts	Fiber Counts	Total Counts	Wet Weight Used (g)	Dry Weight (g)	Average % Spike Recovery
0-1	168.0**	135.0**	304.0	3.93	0.78	0.77
2.0-3.0	115.0	119.0	234.0	3.40	1.10	0.80
4.0-5.0	147.0	22.0	170.0	4.40	1.74	0.80
6.0-7.0	146.0**	152.5**	298.5	6.35	2.20	0.74
8.0-9.0	111.0	15.0	126.0	5.17	1.43	0.80
10.0-11.0	112.0	33.0	148.0	4.52	1.46	0.72
12.0-13.0	144.0	21.0	168.0	4.82	2.18	0.72
14-15	208.5**	64.0**	273.5	2.55	1.04	0.79
16-17	162.0	22.0	184.0	2.93	1.28	0.77
18-19	306.0	76.0	382.0	2.80	1.22	0.72
20-21	54.0	13.0	69.0	2.28	0.96	0.75
22-23	202.5**	58.0**	166.0	2.26	1.46	0.79
24-25	83.0	10.0	93.0	2.45	1.11*	0.80
26-27	185.5**	59.0**	233.0	1.67	0.86	0.71
28-29	44.0	16.0	60.0	2.26	1.07*	0.73
30-31	166.0	39.0	206.0	2.42	1.87	0.72
32-33	89.0	14.0	103.0	2.58	1.20*	0.72
34-35	251.5**	93.0**	376.0	2.03	0.97*	0.76
36-37	324.0	35.0	359.0	2.09	0.99*	0.69
38-39	124.0**	48.0**	172.0	2.09	1.02*	0.74
40-41	132.0	19.0	151.0	2.03	1.05*	0.63
42-43	94.0**	30.0**	124.0	2.09	1.10*	0.69
44-45	89.0	11.0	100.0	2.08	1.17*	0.72
46-47	179.0	48.0	227.0	3.17	1.85*	0.78

* Theoretical dry mass calculated from LOI water content used

** Averaged from duplicate samples

Table A7: Results of manual analysis and counting of microplastic particles in the sediment core from Conestogo Lake.

Interval	Fragment Counts	Fiber Counts	Total Counts	Wet Weight Used (g)	Dry Weight (g)	Average % Spike Recovery
1-2	277.0	131.0	408.0	5.38	1.67	0.67
4-5	233.0**	37.5**	321.0	2.52	0.91	0.77
8-9	315.0	78.0	393.0	2.25	0.97	0.77
12-13	244.0	49.0	293.0	3.35	1.35	0.78
16-17	206.0	74.0	280.0	2.74	1.21	0.80
20-21	155.0	20.0	175.0	1.89	0.80	0.81
24-25	267.5**	72.0**	367.0	3.55	0.48	0.76
28-29	249.0	256.0	505.0	2.04	1.19	0.81
32-33	224.0	45.0	269.0	2.70	1.71	0.80
36-37	188.0	86.0	274.0	1.81	0.77	0.83
40-41	275.0	103.0	378.0	2.06	0.89	0.80
44-45	196.0**	59.5**	316.0	2.12	0.89	0.81
48-49	249.0	65.0	314.0	1.95	0.81	0.82
52-53	272.0	113.0	385.0	3.23	1.37	0.79
56-57	143.0	24.0	167.0	2.45	1.10	0.76
60-61	221.0	70.0	291.0	2.51	1.20	0.76
64-65	177.5**	46.0**	246.0	2.08	1.00	0.76
67-68	181.0	59.0	240.0	1.87	0.91	0.74

* Theoretical dry mass calculated from LOI water content used

** Averaged from duplicate samples