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Assessing the Impacts of Land-Use Change and Climate Variability on Cyanobacterial Abundance and Toxicity in Shallow Lakes

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ABSTRACT

1. The global increase in the frequency and intensity of cyanobacteria blooms has been widely attributed to changes in land-use practices and climate variability, yet little is known of how toxicity has varied historically relative to cyanobacteria abundance.
2. Fossil pigments from cyanobacteria and algae were quantified from shallow lake sediment core records using high-performance liquid chromatography, whilst past concentrations of microcystin congeners were measured using liquid chromatography high-resolution mass spectrometry. These metrics were combined with measures of sedimentary geochemistry ($\delta^{13}C$, $\delta^{15}N$, %N, %C, C:N ratio) to estimate how lake production and abundance of toxigenic cyanobacteria varied during the past ~300 years in two small lakes in New Brunswick, Canada. Harvey Lake is an impacted site with a history of intensive catchment land use, whilst Wheaton Lake is a relatively undisturbed reference site.
3. Stratigraphically constrained cluster analysis (CONISS) revealed that primary production increased steadily in both lakes since the second half of the 20th century, whilst microcystin production increased by an order of magnitude after ca. 2000 CE. Fossil pigment concentrations were initially lower in Harvey Lake but shifted to more productive conditions after initial forest clearance and settlement and again after agricultural intensification during the 20th century. Although Wheaton Lake exhibited higher overall fossil pigment concentrations, including a pre-colonial eutrophic interval (ca. 1680–1750 CE), this reference basin also underwent enrichment since ca. 1980, possibly reflecting longer growing seasons in the last 50 years.
4. Although cyanobacterial pigments and microcystin concentrations were elevated in sediments deposited since ca. 2000 CE in both lakes, these variables were uncorrelated over the entire 300-year record, with the pre-colonial eutrophic interval in Wheaton Lake having low toxin concentrations. This pattern suggests either that cyanobacterial dominance and toxicity are regulated by different factors or that the preservation of microcystins and pigments is under unique controls.

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5. Statistical analyses showed that these small shallow maritime lakes are sensitive to relatively small land-use perturbations within their catchments and that even undisturbed basins may be vulnerable to toxic cyanobacteria blooms in a warming climate.

1 | Introduction

Occurrences of cyanobacteria blooms are increasing in freshwater environments globally (Huisman et al. 2018) resulting in heightened public and scientific concern due to the harmful effects of phycotoxins (i.e., anatoxins, microcystins and saxitoxins) produced by some cyanobacteria species (Dawson 1998; Birk et al. 2023). Cyanobacteria and their toxins negatively impact aquatic ecosystem services, devalue properties, increase the prevalence of anoxia, cause animal fatalities and, in rare instances, induce human illness (Qin et al. 2010; Johnston et al. 2024). A better understanding of the drivers of toxigenic cyanobacterial bloom formation is critical for the effective implementation of management strategies that safeguard freshwater resources (Paerl et al. 2016). Although cyanobacteria are influenced by complex controls, nutrient availability and water temperature are well documented as significant drivers of bloom development (Schindler 1974, 2006; Paerl and Huisman 2008; Taranu et al. 2015). Other factors affecting bloom formation include water column stability (Jöhnk et al. 2008), point-source contaminants (Álvarez et al. 2017), nutrient influx from diffuse sources (Bunting et al. 2016), extreme weather events (Stockwell et al. 2020), climate variability (i.e., changing precipitation patterns, winter lake ice cover and light attenuation) (Paerl and Huisman 2009; Paterson et al. 2017; Qin et al. 2021) and invasive species (Reid et al. 2024). Unfortunately, less is known of the controls of cyanobacterial toxicity or how it may vary at the decadal to centennial scales (Hayes et al. 2020).

Changes in freshwater nutrient status are often closely related to variation in land-use practices within lake catchments (Patterson and Kumar 2002; Roe et al. 2010; Bunting et al. 2007, 2016; Gushulak, Mezzini, et al. 2024). Agriculture and forest clearance can significantly influence nutrient cycling regimes and catchment hydrology, enhancing nutrient runoff into lakes (Patterson et al. 2002; Kakouei et al. 2021). Moreover, the advent of chemical fertilisers during the post-war era (aka 'Great Acceleration' of the 1950s) has resulted in the excessive application of nutrients in many agricultural regions (Steffen et al. 2015). This surplus application has saturated soils and increased release of diffuse sources of phosphorus (P) and nitrogen (N) into rivers and downstream lakes (Carpenter et al. 1998) causing eutrophication (Bunting et al. 2007, 2016; Gushulak, Mezzini, et al. 2024). Despite the close correlation between land-use change and freshwater nutrient loading, it has been difficult to predict the conditions that lead to major changes in the composition and toxicity of phototrophic communities, in part due to differences in antecedent conditions amongst lakes (O'Neil et al. 2012). Cyanobacterial proliferation has been observed across a wide gradient of lake nutrient status (i.e., mesotrophic to hypereutrophic lakes), potentially due to the physiological differences amongst species allowing nutrient uptake from different habitats (benthos, water column) (Cottingham et al. 2015). Furthermore, in systems with excess P present, diazotrophic cyanobacteria (i.e., Nostocales) can fix dissolved N_2 gas into bioavailable forms (i.e., ammonia) thereby partly alleviating N limitation (Cottingham et al. 2015). In systems where both N and P are in excess, non-diazotrophic cyanobacterial genera, such as *Microcystis* and *Planktothrix*, can dominate the phytoplankton community (Paerl and Otten 2013). However, as these communities also differ in their intrinsic toxicity, it has proven difficult to evaluate how land-use change may alter toxin accumulation in ecosystems.

Climate change since the late 1800s is a major driver of lake ecosystem functional change that has disproportionately favoured the proliferation of cyanobacteria over eukaryotic algae (Taranu et al. 2015; Erratt et al. 2023). During the 20th century, average global surface temperatures increased on average by ~1°C due to accumulation of radiatively active gases (IPCC 2021). Warming surface waters enhance cyanobacterial abundance and their subsequent toxin production and are expected to deteriorate water quality globally (Hayes et al. 2020; Erratt et al. 2023). Furthermore, increased precipitation associated with atmospheric warming may enhance nutrient runoff into lakes and influence cyanobacterial bloom proliferation (Stockwell et al. 2020; Erratt et al. 2022, 2023).

Understanding the occurrence, controls and consequences of past toxigenic cyanobacteria blooms is often limited by the absence of long time series of toxin identity and abundance (Christensen et al. 2022). This issue is exacerbated in regions of low population density or remote regions where routine water monitoring is expensive and logistically challenging (Pírez et al. 2013). In principle, palaeolimnological studies can be used to extend the records of cyanobacterial abundance and toxicity, both complimenting and extending the available instrumental data (Bennion and Battarbee 2007). Traditionally, sediment core analyses have used fossil pigments (i.e., carotenoids, chlorophylls and derivatives) (Leavitt 1993; Leavitt and Hodgson 2001) or morphological remains (i.e., frustules, scales, palynomorphs, akinetes and heterocytes) to reconstruct the abundance and composition of primary producer communities (Bunting et al. 2007; Gushulak, Chegoonian, et al. 2024). These approaches have defined the timing and extent of past blooms, as well as provided environmental baselines for restoration targets (Bennion et al. 2011), but are unable to distinguish between toxic and non-toxic cyanobacteria. Recent advancements in sedimentary DNA extraction and quantification have made it possible to discern if toxigenic genes have been present historically (Heathcote et al. 2023), although often these techniques cannot quantify the amount and type of toxin present (e.g., varieties of microcystin). Instead, targeted extraction and quantification of sediment-bound cyanobacterial toxins (Chen et al. 2006; Wu et al. 2012; Zastepa et al. 2015) have been proposed to reconstruct historical changes in toxin abundance from sediment core analyses (Zastepa, Taranu, et al. 2017; Waters et al. 2021; Clift and Waters 2024). In principle, combining analysis of fossil toxins and other proxies such as pigments, stable isotopes and diatoms can provide new insights into the effects of human activities on the composition and toxicity of phytoplankton assemblages, as well as the relationship between lake productivity and cyanobacterial risks (Waters et al. 2021).

In this study, we present a multiproxy analysis of sediment cores from two contrasting shallow lakes in southwestern

New Brunswick (NB), Canada. Both sites experienced well-documented toxigenic cyanobacteria blooms in the summer of 2015 and were part of a broader regional trend of persistent cyanobacterial outbreaks (Valadez-Cano et al. 2022; NB Public Health Advisories 2024). The reference ecosystem, Wheaton Lake, is located in a coastal area near Passamaquoddy Bay and is primarily surrounded by forested shoreline. In contrast, Harvey Lake is situated ~70 km inland and is impacted by a lakeshore village, abundant seasonal cottages and homes and past intensive agriculture on the eastern shore and southwest shorelines. Despite their proximity, Harvey Lake experiences a more continental climate, whilst that of Wheaton Lake is moderated by the adjacent marine bay (Fortin and Dubreuil 2019). At both lakes, we analysed sediments for microcystins (Microcystin-LA, -LR, -RR and -[Dha^7] LR), sedimentary phototrophic pigments and geochemical parameters (stable isotopes of C and N, %N, %C, C:N ratio). Microcystin congeners were chosen as a proxy for toxigenic cyanobacterial blooms as they are the most commonly reported cyanotoxins produced by freshwater cyanobacteria and have a high preservation potential in lake sediments (Zastepa et al. 2014).

By contrasting toxin availability with cyanobacterial abundance from fossil pigments, we sought to evaluate whether controls of cyanobacteria abundance differed from those of toxicity. The objectives of the study were to (1) reconstruct historical changes in lake production using taxonomically diagnostic fossil pigments and stable isotopes; (2) investigate the timing and magnitude of past toxigenic cyanobacteria using sediment-bound microcystin congeners; (3) explore the influence of land-use change as a driver of lake cyanobacteria occurrence in the two contrasting lake systems and (4) evaluate the influence of climate change (e.g., warming temperatures, increased open water season) on lake production and cyanobacterial toxicity. We hypothesised that impacted Harvey Lake should exhibit a greater degree of water quality degradation (increased production, higher toxicity) than reference Wheaton Lake due to its more extensive record of land-use change. This study represents one of the first examples of combining sedimentary geochemistry, past primary production and fossil toxins to estimate how differences in land use and climate affect the abundance of toxigenic cyanobacteria in two contrasting shallow lakes.

2 | Methods

2.1 | Study Sites

Harvey and Wheaton lakes were selected for multiproxy sediment core analysis owing to their contrasting land-use histories despite both experiencing first recorded incidences of cyanobacterial blooms in 2015 (NB Public Health Advisories 2024). Harvey and Wheaton lakes lie within the traditional territory of the Passamaquoddy (Peskotomuhkati) First Nation, a subgroup of the Maliseet (Wolastoqiyik) and part of the Wabanaki Confederacy (Bourque 2004). These seasonally nomadic indigenous populations lived and fished along the rivers and shorelines of both lakes for centuries before European colonisation, as evidenced from the presence of a shell-midden south of Wheaton Lake (Sanger 1986) and numerous archaeological artefacts near Harvey Lake (Harvey Lake Association, pers. comm., 2024).

Harvey Lake (45.72905° N, −67.02235° W) is a relatively shallow lake with a maximum depth of 13.6 m and a surface area of ~7 km^2 (Figure 1, Table 1). Adjacent to the village of Harvey (Station) within the greater Harvey Rural Community, the lake is within the Southern Highland climate region (van Groenewoud 1984; Fortin and Dubreuil 2019) of the Eastern Canada temperate climate zone (ECTCZ; Bailey et al. 1997). There are ~160 year-round homes and seasonal cottages established along the lake shore, concentrated around Herbert's Cove and the eastern margin along Highway 636 (Figure 1; Atasiei et al. 2022). Established in 1837 as an agricultural community, Harvey Settlement saw rapid forest clearance for farming and continued forestry along its shoreline. After 1869, the western extension of the European and North American Railway was completed, which created a crossroads with the ‘Great Road’ between Fredericton and St. Andrews at the southeastern corner of the lake and ran along the lake's western shore. After the completion of the railroad, the village (aka Harvey Station) progressed rapidly with around two dozen homes and businesses by 1878 (Elliott 2005). The growth of Harvey Station from 1869 onwards, with stores and hotels, impacted the lake, and mid-20th-century prosperity spurred a ‘cottage culture’ around it. Since the 1990s, increased development of year-round homes with lawns and infrastructure has placed further pressure on the lake's environment.

Wheaton Lake (45.166283° N, −66.99755° W) is a small and shallow lake, with a maximum depth of 12 m and a surface area of ~1 km^2, located less than 1 km from the moderating influence of Passamaquoddy Bay, an inlet of the Bay of Fundy (Figure 1, Table 1) within the Southwest Climate Region of the ECTCZ (van Groenewoud 1984; Fortin and Dubreuil 2019). The lake lies within a predominately low-relief catchment with only a few lakeshore homes. At present, the catchment is largely forested, although the region may have been subject to agriculture as early as the late 1700s (see below). The most significant recent landscape disturbance was the regional Bocabec forest fire in 1968, which affected the lake's northern flank, as well as recent limited lakeside property development (Figure 1).

It is unclear what impact either the native populations or the initial Acadian settlers (ca. 1604) may have had on Wheaton Lake, though population densities were likely very low. Regional settlement began in 1784, following the end of the Revolutionary War, when Loyalist and demobilised British soldiers arrived and were granted land in the Passamaquoddy region, including the area surrounding Wheaton Lake (Fellows 1971; Wynn 1981). However, historians suggest that there were few signs of human occupancy near Passamaquoddy Bay during this period (Wynn 1981).

2.2 | Core Collection

Sediment cores were collected from the study lakes in September 2021 using a modified UWITEC gravity corer (USC 09000) (UWITEC 2020), with SCUBA assistance. This approach allowed for the retrieval of both longer and precisely positioned cores, which could be capped at the lake bottom. The sediment cores were collected from water depths of ~6 m in both sites (Table 1). Sediment cores were maintained in a vertical position

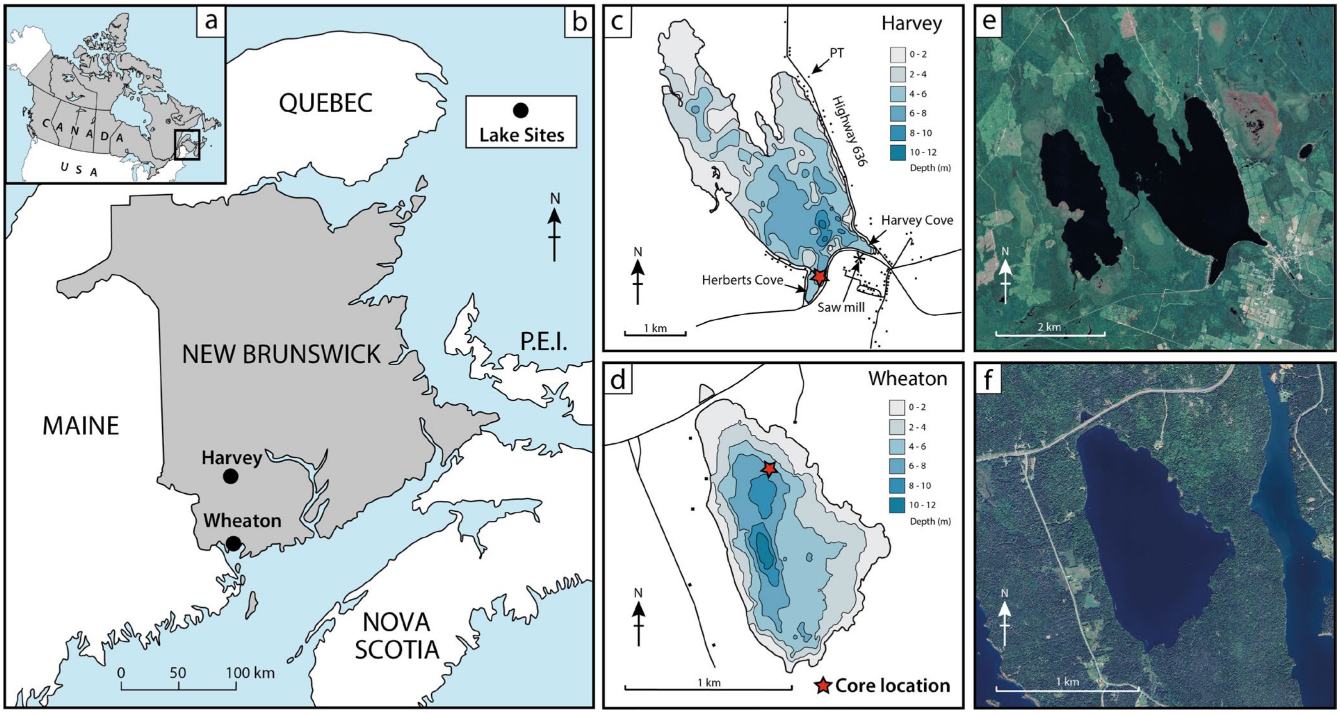

FIGURE 1 | Map of the study area showing (a) overview of Canada; (b) the province of New Brunswick with labelled study sites; (c) map of Harvey Lake, including bathymetric data (data from the Government of New Brunswick), core location and selected land-use attributes; (d) map of Wheaton Lake, including bathymetric data (data from the Government of New Brunswick) and core location; (e) aerial imagery of Harvey Lake and (f) Wheaton Lake (Image from Google Earth).

TABLE 1 | Physical and chemical parameters of studied lakes in September 2021.

	Harvey Lake	Wheaton Lake
Core location	45.72905° N, −67.02235° W	45.166283° N, −66.99755° W
Core collection depth	6.4 m	5.8 m
Max depth	13.6 m	11.98 m
Surface area	695.6 ha	104 ha
Ammonia (as N)	< 0.05 mg/L	< 0.05 mg/L
Kjeldahl Nitrogen	0.2 mg/L	0.4 mg/L
pH	7	7.1
Alkalinity (as $CaCO_3$)	8 mg/L	12 mg/L
Nitrate	< 0.05 mg/L	< 0.05 mg/L
Nitrite	< 0.05 mg/L	< 0.05 mg/L
Phosphorus—Total	0.01 mg/L	0.023 mg/L
Carbon—Dissolved organic	4.3 mg/L	3.2 mg/L
Chlorophyll-*a*	2.2 µg/L	14.4 µg/L
Solids—Total dissolved	24 mg/L	30 mg/L

and subsampled on-site using a portable extruder (Patterson, Nasser, Tremblay, and Galloway 2022). Samples were sectioned at 0.5-cm intervals to a depth of 20 cm and at 1-cm intervals thereafter. Following this, samples were frozen (−21°C) prior to subsampling for pigment, geochemical and microcystin analyses.

2.3 | Radiometric Dating

Subsamples from the sectioned sediment cores were measured for ^{210}Pb activity by gamma spectrometry at the University of Ottawa, Ontario, Canada. Chronologies for each lake core were generated using *rplum*, an R studio package for the

Bayesian age-depth modelling of ^{210}Pb activity (Aquino-López et al. 2018). Like traditional ^{210}Pb chronologies, *rplum* assumes a constant rate of supply (CRS) of atmospheric ^{210}Pb deposition (Appleby 2001). However, a self-adjusting Markov chain Monte Carlo (MCMC) algorithm separates the ^{210}Pb decay equation from the age-depth modelling process to produce more accurate age-depth estimates (Aquino-López et al. 2018). For the cores analysed in this study, the supported activity was calculated for each sediment segment using the measured ^{226}Ra activity.

2.4 | Stable Isotopes

Stable isotope ratios were estimated using an Elementar vario EL cube elemental analyser, interfaced to an Elementar VisION IRMS (Elementar Analysensysteme GmbH, Langenselbold, Germany) at the University of California-Davis Stable Isotope Facility. Stable carbon (δ^{13}C) and nitrogen (δ^{15}N) isotope value ratios were expressed as per mille (δ‰) units against authentic international standards of Vienna Pee Dee Belemnite and atmospheric nitrogen, respectively.

2.5 | Microcystin Analysis

The detailed protocol for the extraction and analysis of microcystins in sediment core subsamples can be found in Appendix S1. Briefly, sediment-bound microcystins were extracted from each sub-sample by first centrifuging whole sediment and decanting porewater. Microcystins within the sediment pore water were not quantified to reduce bias associated with post-depositional diffusion, enabling a more reliable comparison with the lipid-soluble fossil pigments. Sediment samples were extracted twice with 75% aqueous methanol and resulting extracts were combined. Methanol was removed by rotary evaporation, and extracts were concentrated by solid phase extraction (SPE) using Waters Hydrophilic–Lipophilic-Balanced (HLB) SPE cartridges. Eluted extracts were dried under a stream of nitrogen gas and stored at −20°C before liquid chromatography high-resolution mass spectrometry (LC-HRMS) analysis.

Certified reference materials of microcystin-LR (CAS: 101043-37-2), [Dha^{7}] microcystin-LR (CAS: 120011-66-7), microcystin-LA (CAS: 96180-79-9), nodularin-R (CAS: 118399-22-7) and microcystin-RR (CAS: 111755-37-4) were procured from the National Research Council of Canada's (NRC) Metrology Research Centre (Halifax, Nova Scotia). Blank sediment and microcystin-spiked sediment were run in parallel with study samples for method performance validation (Appendix S1). Total microcystin concentrations (TMC) reported were calculated as the sum of microcystin-LA, -LR, -RR and -[Dha^{7}] LR as ng/g. Nodularin-R was the internal standard.

LC-HRMS analysis was performed using an Agilent 1290 high-performance liquid chromatography (HPLC) system coupled to a Q-Exactive Orbitrap mass spectrometer (Thermo Fisher Scientific). Extracts were resolved with a C18 Eclipse Plus RRHD column (2.1 × 50 mm, 1.8 μm; Agilent Technologies) maintained at 35°C using gradient elution with water and acetonitrile mobile phases, both containing 0.1% formic acid. HPLC chromatographic and mass spectrometry settings and quantification parameters are reported in Appendix S1. Method limits of detection (LOD) and quantification (LOQ) are summarised in Appendix S1. Briefly, microcystin-LR and RR were detected at concentrations ≥ 0.02 ng/g, whereas microcystin-LA and -[Dha^{7}] LR were detectable at concentrations ≥ 0.08 ng/g.

2.6 | Fossil Pigment Analysis

Fossil pigments were extracted and quantified using HPLC, at the University of Regina, Saskatchewan, Canada, following protocols outlined by Leavitt and Hodgson (2001). Pigments were isolated and quantified using an Agilent 1100 HPLC autosampler system, fitted with a photodiode array detector, calibrated using authentic standards (Leavitt and Hodgson 2001). Spectral characteristics and chromatographic positions were used to tentatively identify carotenoids, chlorophylls and their derivative pigments (Leavitt and Hodgson 2001). Pigment concentrations are expressed as nmols g^{-1} organic matter, a metric which is directly comparable to microscopic estimates of phytoplankton abundance in whole lake experimental calibration (Leavitt and Findlay 1994).

2.7 | Climate Records

Sub-regional scale climatic records for each lake were derived from ERA5 monthly averaged data on single levels from 1940 to present (Hersbach et al. 2023). The nearest 0.25° grid cells to each lake were selected and '2 m temperature', 'total precipitation', '10 m wind speed', surface pressure' and surface net solar radiation', downloaded from the Copernicus database. Meteorological seasonal temperature means, and total seasonal precipitation were generated accounting for differing month length and leap years.

Ice-out, the date of annual ice disappearance from a body of water, was used as a proxy for climate warming and longer phytoplankton growing seasons (Patterson and Swindles 2015). Due to the limited ice-out data for Harvey Lake (available only from 1975 onwards), an extended record of ice-out dates was estimated back to 1876 using data from three lakes in New Brunswick—Oromocto Lake, Skiff Lake and Harvey Lake—as well as West Grand Lake in Maine (Walsh and Patterson 2022). Since no ice-out data were available for Wheaton Lake, the ice-out record from nearby coastal Lake Utopia (~20 km away, same climatic zone) was used to approximate ice-out dates for Wheaton Lake (Patterson and Swindles 2015; Walsh and Patterson 2022). In general, 'ice in' occurs later in the autumn and 'ice-out' occurs earlier in the spring for lakes in the coastal Southwest Climate Region (Wheaton) relative to those in the inland continental Southern Highland Climate Region (Harvey) (Patterson and Swindles 2015).

2.8 | Regional Records of Human Occupation

To quantify how human activities on the landscape affected water quality, regional records of human occupation from the nearest census subdivision to each lake were derived from Canadian census data obtained from Statistics Canada (1871–2021) (Appendix S2). These subdivision population values are presented as an indicator of human activity. Whilst other metrics of human activity were considered (i.e., number of dwellings), none of them were complete for the entire analytical period other than population. For Harvey Lake, the population data was derived from Harvey Village and Manners Sutton Parish subdivisions, as Harvey Village was part of Manners Sutton Parish prior to 1966. To account for this, the population data presented here are the sum of the two subdivisions (See Appendix S2). The population record presented for Wheaton Lake is derived from the Saint Patrick Parish subdivision.

2.9 | Numerical Analysis

Stratigraphically constrained cluster analysis (CONISS) (Grimm 1987) was performed to identify zones and points of change in the records. The analysis was restricted to $\log_{10}$ transformed geochemical signatures ($\delta^{13}C$, $\delta^{15}N$, %N, %C, C:N ratio) and chemically stable pigment concentrations: diatoxanthin (diatoms), alloxanthin (cryptophytes), pheophytin *b* (chlorophytes), lutein-zeaxanthin (chlorophytes + cyanobacteria), echinenone (total cyanobacteria), canthaxanthin (Nostocales cyanobacteria), aphanizophyll (N_2-fixing bacteria) and pheophytin *a* (total phototrophs). Sample intervals with erroneously low pigment concentrations (e.g., zeros) were omitted but accounted for $< 2\%$ of observations. Preliminary comparison of clustering patterns of fossil pigments and geochemical data with and without microcystins did not identify any significant differences in the number or duration of zones.

The analysis of sediment core observations with climate and population records required the datasets to be subsampled at comparable intervals. The medium *rplum* derived age for each sediment interval was matched to the corresponding census and climate variables year. Due to the wide error terms associated with ^{210}Pb dating, a 5-year moving average was applied to each climate variable prior to subsampling.

Principle component analysis (PCA) was performed on the chemically stable pigment concentrations (previously described) from each lake, using the fviz_pca_biplot function in the *factoextra* package (version 1.0.1) R Studio (Kassambara and Mundt 2017). Principle component axis 1 (PCA 1) was extracted and presented here as a bulk signature of total lake production.

Spearman rank correlation analysis was performed to test the significance of the relationship between the proxy data (i.e., PCA 1 and microcystins) and the drivers of environmental change (i.e., population data and climate variables). Correlations were calculated using the rcorr function in the *Hmisc* package (version 5.1-0) R Studio (Harrell 2023). The strength of the correlation was determined by using grading standards outlined by Yan et al. (2019).

3 | Results

3.1 | ^{210}Pb Chronology

Analysis of ^{210}Pb and ^{214}Pb activity provided a depositional timeframe for each sediment core, allowing ages to be estimated for each sediment section (Appendix S3). In Harvey Lake, ^{210}Pb activity declined in a monotonic fashion throughout the record. Due to the significant age uncertainty beyond the ^{210}Pb/^{214}Pb equilibrium (~21 cm), estimated ages past this depth should be treated with caution. The ^{210}Pb and ^{214}Pb activity from the Wheaton Lake core declined in a non-monotonic pattern, suggesting limited mixing, disturbance or variation in deposition rates in the upper ~10 cm of the sediment core.

3.2 | Microcystins

3.2.1 | Harvey Lake

In Harvey Lake, sediment-bound microcystins were absent or below detection limits between 36 and 8 cm (ca. 1722–1992) (Figure 2a). Toxins were first detected between 8 and 6.5 cm, with an average TMC of 0.5 ng g^{-1} dw (dry weight) (ca. 1992–2001). Concentrations increased ~100-fold in the interval 6.5–3.5 cm (ca. 2001–2013) to average values of 58.7 ng TMC g^{-1} dw. This interval was solely comprised of MC-LR. Between 3.5 and 0 cm (ca. 2013–2021) microcystins reached the highest levels detected (mean 187.8 ng TMC g^{-1} dw). This interval had the greatest diversity of microcystins detected, with MC-LA, -LR and -RR present. The maximum TMC concentration (615.5 ng TMC g^{-1} dw) was detected in the surface interval (1.5–1 cm; ca. 2020).

3.2.2 | Wheaton Lake

In Wheaton Lake, microcystins were detected intermittently throughout the ~35 cm sediment core record (Figure 2b). MC-LA was the only congener present beneath 5 cm depth, occurring in five discrete intervals with an average concentration of 21.1 ng g^{-1} dw. The deepest interval with microcystins present was 32–31 cm (ca. 1714), which had a concentration of 16.1 ng g^{-1} dw. In the interval between 5 and 3 cm, MC-LR was the sole congener, with concentrations ranging between 48.5 and 64.8 ng g^{-1} dw. Between 2.5 and 0 cm (ca. 2014–2021), both the diversity and concentration of microcystin congeners increased. In this interval, MC-LR and -RR were detected in contiguous samples and reached an average concentration of 144.2 ng TMC g^{-1} dw.

3.3 | Pigment and Stable Isotope Zonation

Cluster analysis (CONISS) based on the most stable pigments and geochemical proxies ($\delta^{13}C$, $\delta^{15}N$, %N, %C, C/N ratio) identified two principal zones (H-1 and H-2), with four subzones (H-1a, H-1b, H-2a and H-2b) in the Harvey Lake core (Figures 3a and 4a). The most significant change in phytoplankton assemblages and geochemical signatures occurred after ca. 1960, at the boundary between Zone H-1 (ca. 1722–1963) and Zone H-2 (ca. 1963–2021). In Wheaton Lake, CONISS indicated two principal zones, Zone W-1 (ca. 1681–1976) and Zone W-2 (ca. 1976–2021), distributed into six distinct subzones (W-1a, W-1b, W-1c, W-1d, W-2a and W-2b) (Figures 3b and 4b).

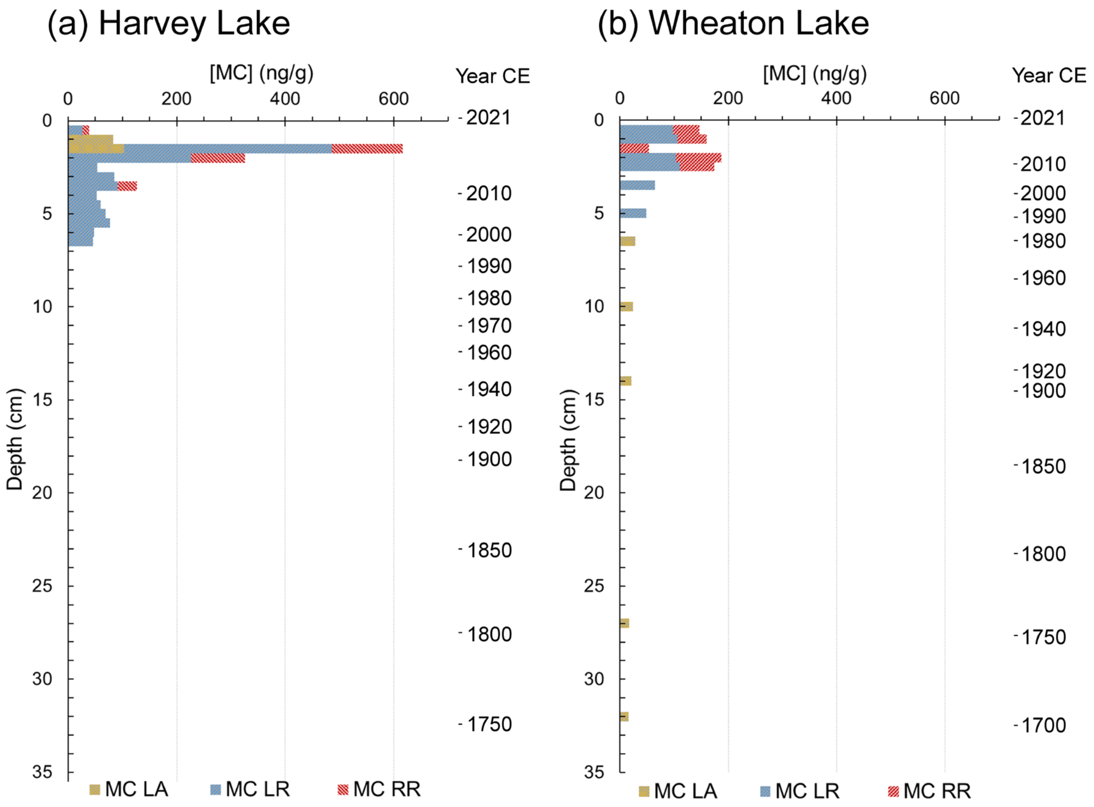

FIGURE 2 | Microcystins (MC-LA, -LR and -RR) congener concentrations expressed as ng MC/g sediment (ng/g) collected from (a) Harvey Lake and (b) Wheaton Lake. Concentrations are plotted against sediment core depth and *rplum* derived ^{210}Pb ages.

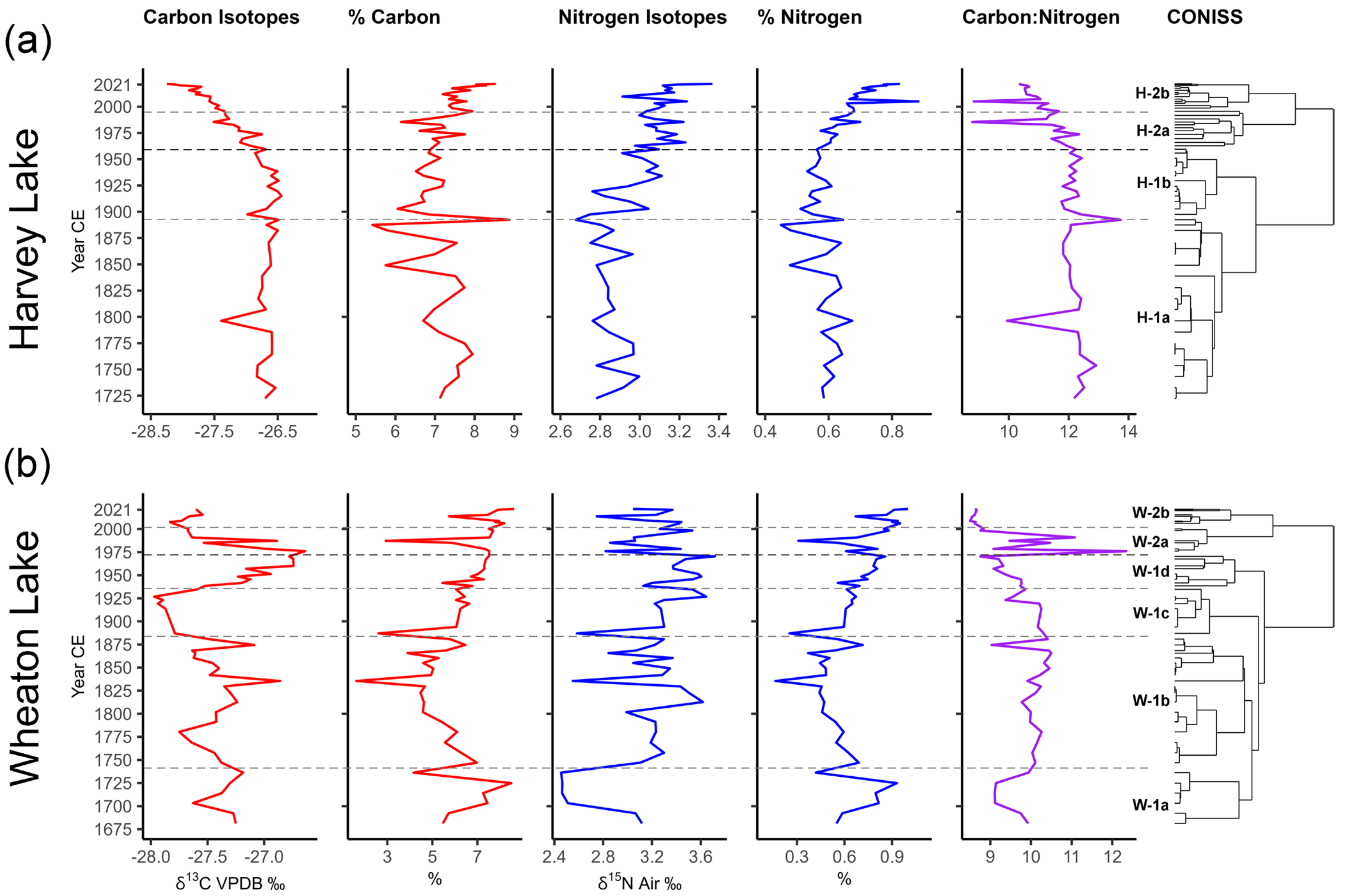

FIGURE 3 | Lake geochemistry ($\delta^{13}C$, $\delta^{15}N$, %N, %C, C:N ratio) for (a) Harvey Lake and (b) Wheaton Lake. Geochemical signatures are plotted against *rplum* derived ^{210}Pb ages. Horizontal lines delineate CONISS cluster zones.

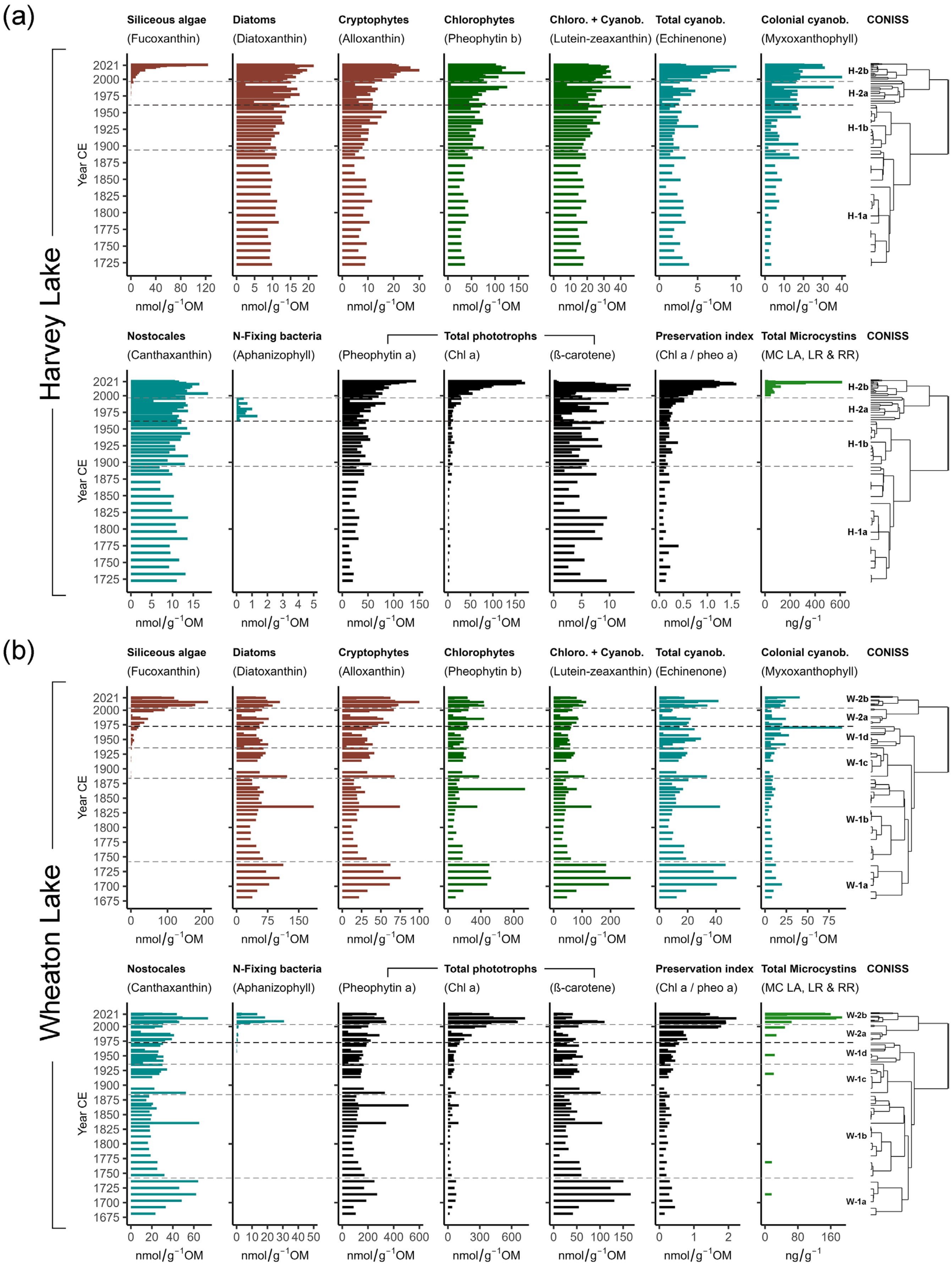

FIGURE 4 | Legend on next page.

FIGURE 4 | Profile of phytoplankton pigments (nmol pigment g^{-1} organic matter) in (a) Harvey Lake and (b) Wheaton Lake. Concentrations are plotted against *rplum* derived ^{210}Pb ages. Horizontal lines delineate CONISS cluster zones. Total microcystin concentrations (TMC) are plotted for each lake (not included in the CONISS analysis).

3.3.1 | Harvey Lake

Subzone H-1a (ca. 1722–1897) was defined by relatively low and stable %N (~0.6%) and δ^{15}N (~2.8‰), isotopically depleted δ^{13}C signatures (~−26.7‰) and a C:N mass ratio of ~12 (Figure 3a). Sedimentary pigment concentrations were also low and stable, with the assemblage primarily comprised of compounds derived from chlorophytes (pheophytin *b*), diatoms (diatoxanthin) and cryptophytes (alloxanthin) (Figure 4a). Subzone H-1b (ca. 1897–1963) was characterised by modest shifts in the geochemical proxies. During this interval, signatures of δ^{13}C and C:N remained mostly stable, whilst δ^{15}N exhibited an overall increase (Figure 3a). In this subzone, fossil pigment concentrations increased across most taxonomic groups, with concentrations of pheophytin *b* from chlorophytes that increased on average by ~70% from Subzone H-1a (Figure 4a).

Subzone H-2a (ca. 1963–1999) was characterised by a small but significant increase in δ^{15}N, with a subzone average of ~3.1‰ (Figure 3a). Concurrently, signatures of δ^{13}C depleted by ~0.5‰, with a subzone average value of −27.2‰, whilst C:N ratios decreased as %C and %N content increased modestly. Subzone H-2a was marked by an overall increase in fossil pigment concentrations over several taxonomic groups, including common cyanobacteria-specific pigments (i.e., echinenone and myxoxanthophyll) (Figure 4a). In addition, there was a transient occurrence of the pigment aphanizophyll, indicative of diazotrophic (N_2-fixing) cyanobacteria, albeit only at concentrations marginally above detection limits. Finally, Subzone H-2b (ca. 1999–2021) was marked by a major shift in geochemical signatures, with enriched δ^{15}N (mean 3.1‰; range ~2.9‰–3.4‰), and a continued depletion of δ^{13}C which reached a minimum of −28.2‰ (Figure 3a). The C:N ratio also further decreased, with an average subzone value of 10.6 (range 8.8–11.3). Fossil pigment concentrations increased sharply in Subzone H-2a, with the highest concentrations of compounds from diatoms (diatoxanthin), cryptophytes (alloxanthin), chlorophytes (pheophytin *b*), total cyanobacteria (echinenone) and Nostocales (canthaxanthin) (Figure 4a).

3.3.2 | Wheaton Lake

Subzone W-1a (ca. 1681–1747) exhibited elevated fossil pigment concentrations that formed a bi-modal peak which represented the concentration maxima for several groups, including cryptophytes (alloxathin), total phototrophs (β-carotene), cyanobacteria and chlorophytes (lutein-zeaxanthin) and total cyanobacteria (echinenone) (Figure 4b). During this interval, δ^{15}N (~2.7‰) and C:N (9.5) values were low, whilst %C (6.4%) and %N (0.7%) signatures were relatively elevated (Figure 3b). In contrast, Subzone W-1b (ca. 1747–1887) exhibited a significant decrease in pigment concentrations (Figure 4b) and a shift in geochemical signatures from the elevated values observed in Subzone W-1a (Figure 3b). Subzone W-1b was the longest of the six subzones identified in Wheaton Lake (ca. 140 years) and exhibited the most geochemical variability. In this subzone, δ^{13}C and δ^{15}N values fluctuated between −26.9‰ and −27.8‰ and 2.6‰–3.6‰, respectively, whilst the C:N ratio remained generally stable at ~10 (Figure 3b). Fossil pigment concentrations decreased significantly from the maxima observed in Subzone W-1a, with a ~50% decrease in alloxanthin (cryptophytes), pheophytin *b* (chlorophytes) and canthaxanthin (Nostocales), and a > 60% decrease in lutein-zeaxanthin (chlorophytes and cyanobacteria), echinenone (total cyanobacteria) and β-carotene (total phototrophs) (Figure 4b). Subzone W-1c (ca. 1887–1938) recorded a subtle overall increase in pigment concentration across most taxonomic groups, whereas δ^{15}N, %N and %C values increased and C:N decreased marginally. Values of δ^{13}C also decreased in this subzone, reaching a profile minimum of −28‰. In Subzone W-1d (ca. 1938–1976), the concentration of pigments from colonial cyanobacteria (myxoxanthophyll) increased from the levels observed in Subzone W-1c, whereas those from chlorophytes (as pheophytin *b* and lutein-zeaxanthin), diatoms (diatoxanthin) and cryptophytes (alloxanthin) decreased (Figure 4b). Notably, this subzone marked the first appearance of fossils from diazotrophic cyanobacteria. Signatures of δ^{15}N, %C and %N continued to enrich, whilst the C:N ratio decreased. Decreased δ^{13}C observed in Subzone W-1c shifted values towards more enriched (less negative) signatures.

Subzone W-2a (ca. 1976–2006) saw the abundances of total cyanobacteria (echinenone) and colonial cyanobacteria (myxoxanthophyll) decrease, whereas chlorophytes (pheophytin *b*), diatoms (diatoxanthin) and cryptophytes (alloxanthin) increased relative to Subzone W-1d (Figure 4b). In this subzone, δ^{13}C values were depleted, whilst %C decreased, and average %N values remained similar to Subzone W-1d, albeit with fluctuations (Figure 3b). The C:N ratio exhibited a relatively large range (~8.8–12.3). Pigment concentrations increased significantly in Subzone W-2b (ca. 2006–2021), with peak concentrations for diazotrophic cyanobacteria (aphanizophyll), cryptophytes (alloxanthin) and Nostocales (canthaxanthin). Overall, the fossil pigment concentrations observed in subzone W-2b did not exceed the levels observed in Subzone W-1a (Figure 4b). At the same time, C and N contents increased, whilst the C:N ratio value continued to decline (Figure 3b).

3.4 | Climate Analysis

During 1950–2021, mean seasonal temperature showed a warming trend across all seasons for ERA-5 reanalysis grids that represent the locations of both Harvey and Wheaton lakes (Appendix S4). On average, the marine-moderated area around Wheaton Lake experienced warmer autumns and winters compared to the more continental Harvey Lake region. Summer seasonal average temperature increased by ~1°C in both sites between 1950 and 2021. Annual total precipitation did not vary significantly between sites, with a modest increasing trend observed between 1950 and 2021, although a higher proportion

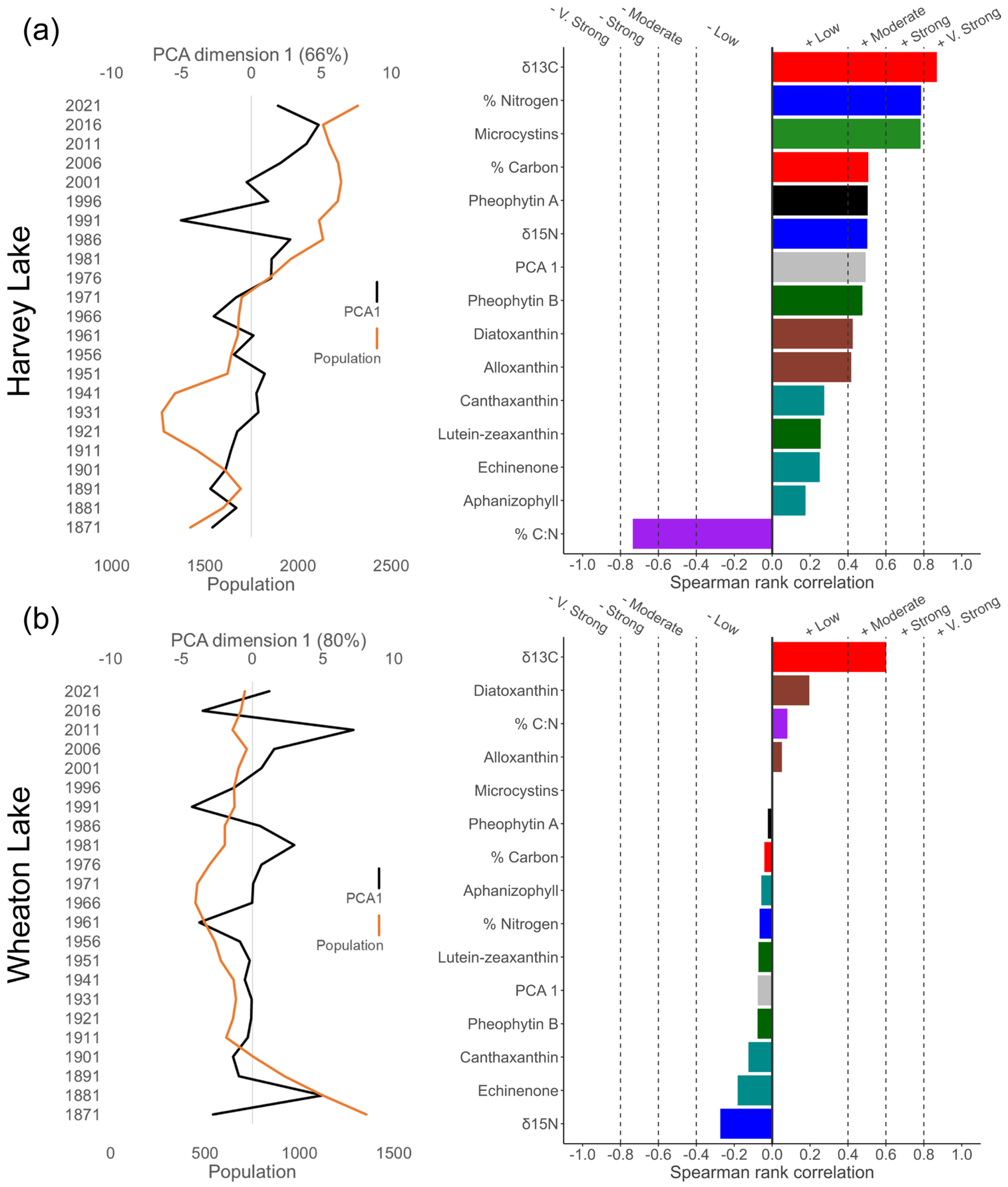

FIGURE 5 | Principle component analysis (PCA) dimension 1 and population data plotted against census year, with Spearman rank correlation scores between population and pigments, lake geochemistry (δ^{13}C, δ^{15}N, %N, %C, C:N ratio) and total microcystin concentration for (a) Harvey Lake and (b) Wheaton Lake. Colours follow those used in Figures 3 and 4.

of that precipitation would have fallen as snow at Harvey Lake (Appendix S4). Ice-out dates have become earlier at both sites, resulting in an open water season that is now approximately 10 days longer than in the late 19th century. Notably, the ice cover season at inland Harvey Lake is consistently longer than at coastal Wheaton Lake. Regression analysis indicates that, in general, ice-out dates are advancing more rapidly along the coast (Patterson and Swindles 2015) (Appendix S4). Average annual wind speed, solar radiation and air pressure were higher for more southerly coastal Wheaton Lake than inland Harvey Lake (Appendix S4).

3.5 | Relationship Between Climate-Land-Use and Lake Production

3.5.1 | Harvey Lake

Spearman rank correlation analysis revealed that human population was correlated positively with many fossil measures from Harvey Lake sediments (Figure 5a). Total microcystins ($\rho = 0.78$), % nitrogen ($\rho = 0.79$) and δ^{13}C ($\rho = 0.87$) were strongly and positively correlated with population, whilst pigment PCA 1 ($\rho = 0.49$) and δ^{15}N ($\rho = 0.5$) were moderately correlated with

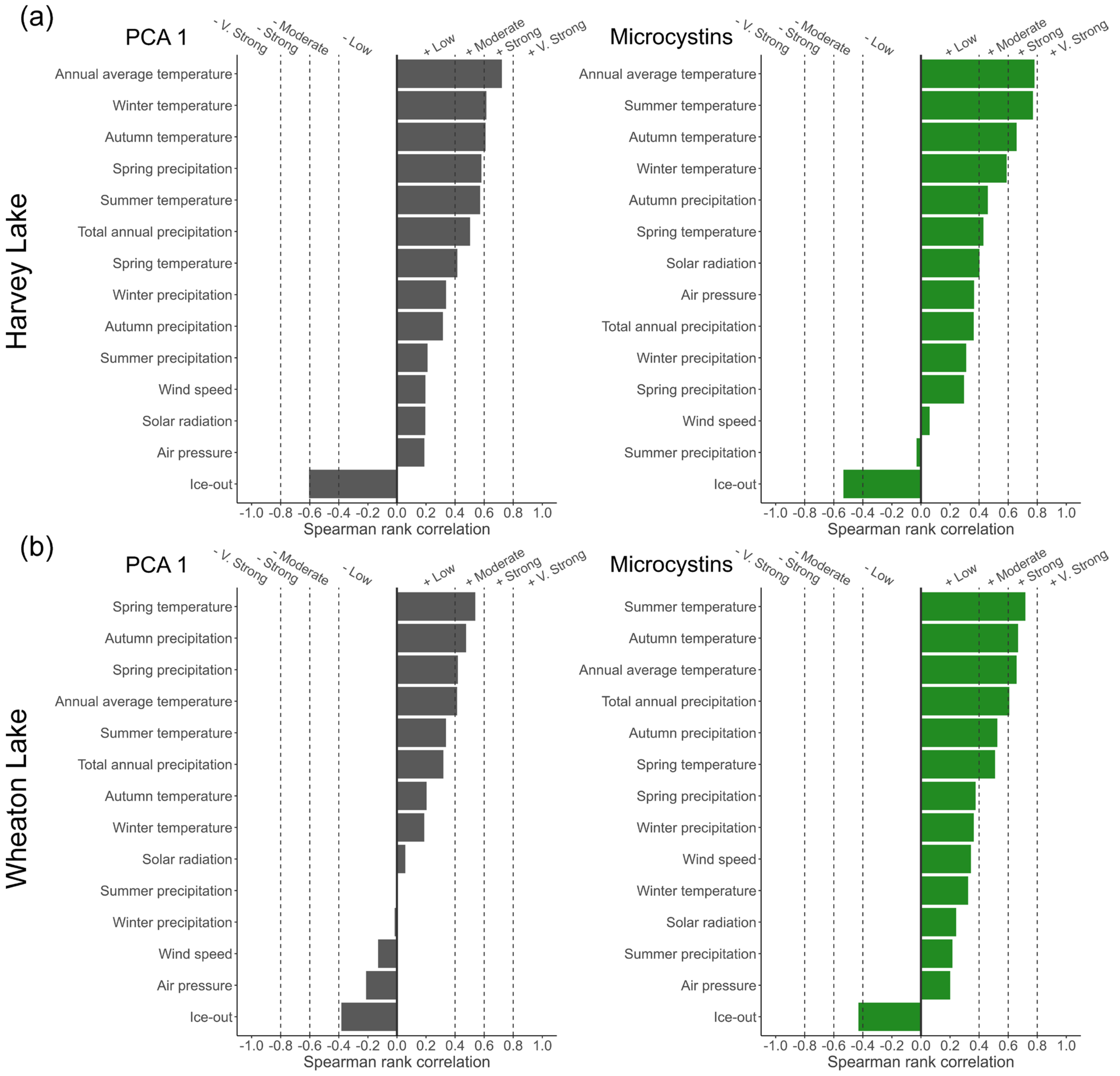

FIGURE 6 | Spearman rank correlation scores between climate variables and PCA 1 and microcystins for (a) Harvey Lake and (b) Wheaton Lake.

humans in the vicinity of Harvey Lake. Population correlations were positive but much weaker with measures of potentially N_2-fixing (canthaxanthin $\rho = 0.27$; aphanizophyll $\rho = 0.18$) and total cyanobacteria (echinenone $\rho = 0.25$), as well as the aggregate abundance of 'bloom-forming taxa' (lutein-zeaxanthin $\rho = 0.26$). The C:N ratio ($\rho = -0.74$) was strongly negatively correlated with population.

Spearman rank correlation between the climatic variables and PCA 1 and total microcystin concentration indicated moderate to strong positive correlations ($0.42 \leq |\rho| \leq 0.78$) between the temperature variables, whilst precipitation was generally weakly to moderately positively correlated ($0.21 \leq |\rho| \leq 0.58$) (Figure 6a). Ice-out date was strongly negatively correlated with pigment PCA 1 ($\rho = -0.6$) and moderately negatively correlated with fossil concentrations of microcystins ($\rho = -0.53$). Other climate variables, including wind speed, solar radiation and air pressure, were typically very weakly to weakly ($0.1 < |\rho| \leq 0.37$) positively correlated with PCA 1 and microcystins, with the exception of solar radiation which was moderately positively correlated with microcystin concentration ($\rho = 0.40$).

3.5.2 | Wheaton Lake

For Wheaton Lake, the Spearman rank correlations between population and sediment proxies were typically weakly negatively to weakly positively correlated ($-0.3 \leq |\rho| \leq 0.2$), with the exception of $\delta^{13}C$ ($\rho = 0.6$) which was strongly positively correlated to regional human populations (Figure 5a). Total microcystins were uncorrelated with population change, whilst aphanizophyll ($\rho = -0.06$), lutein-zeaxanthin ($\rho = -0.07$), canthaxanthin ($\rho = -0.13$) and echinenone ($\rho = -0.18$) were all very weakly negatively correlated with population change.

Spearman rank analysis indicated a moderate positive correlation between pigment PCA 1 for Wheaton Lake and local spring average temperature ($\rho = 0.54$), annual average temperature ($\rho = 0.41$), autumn total precipitation ($\rho = 0.48$) and spring total precipitation ($\rho = 0.42$) (Figure 6b). Similarly, microcystins were strongly and positively correlated with summer average temperature ($\rho = 0.72$), autumn average temperature ($\rho = 0.67$), annual average temperature ($\rho = 0.66$) and annual total precipitation ($\rho = 0.61$). Wind speed, solar radiation and air pressure were very weakly positively to weakly negatively correlated with PCA 1, whilst the same variables had a weak positive relationship with TMC. Ice-out date was weakly negatively correlated with PCA 1 ($\rho = -0.38$), whilst microcystins ($\rho = -0.43$) were moderately negatively correlated for Wheaton Lake.

4 | Discussion

Analysis of sedimentary pigments, stable isotopes and microcystins from the Harvey and Wheaton Lake sediment cores illustrated significant shifts in geochemistry, lake production, cyanobacterial abundance and phycotoxin levels over the past ca. 300 years. Despite differences in land-use histories, both lakes exhibited varying degrees of eutrophication since the mid-twentieth century, marked by elevated cyanobacterial biomass and TMC. In Harvey Lake, the timing of these changes corresponds with the early agricultural expansion that followed European settlement in 1837, the founding and subsequent construction of the village of 'Harvey Station' in 1869, post-WWII cottage development, the introduction of chemical fertilisers, and the shift to year-round home development in the late 20th century. Moderate positive correlations between pigment PCA 1, $\delta^{15}N$, and population increase suggest that water quality degradation resulted from increased nutrient influx from both point and diffuse sources (Bunting et al. 2007). Whilst Wheaton Lake also underwent recent, albeit slower, eutrophication, the overall weak correlations between population change and sediment core proxies suggest that human activities within the catchment have had a limited impact on lake production. Moreover, moderate positive correlations between PCA 1, temperature and precipitation variables, as well as a strong positive correlation between average temperature and microcystins, are consistent with the inference that regional climate change has regulated changes in production and toxicity of Wheaton Lake. Despite the lack of discernible anthropogenic forcing at Wheaton Lake, the ten-fold greater fossil pigment concentrations at that site compared to Harvey Lake suggest that Wheaton Lake has usually been the more productive of the two basins, potentially owing to its smaller size and shallower bathymetry. In light of these findings, we suggest that our impacted site (Harvey Lake) was more strongly influenced by anthropogenic processes than reference Wheaton Lake, but that both sites were affected by climate variability. Furthermore, the higher toxin concentrations in Harvey Lake relative to Wheaton Lake, despite greater primary production in the latter, reveal a complex relationship between historical trends in lake production and toxin occurrence in these systems.

4.1 | Changes in Lake Production Over Time (Objective 1)

Changes in past phytoplankton abundance and geochemical parameters suggest a trend towards increased primary production in both lakes during the 20th and 21st centuries. The pattern of production change at Harvey Lake is typical of other New Brunswick lakes (i.e., Daly et al. 2019), as well as other shallow lakes globally that have undergone settlement-related eutrophication (Jinglu et al. 2007; Taranu et al. 2015). In most cases, pigment concentrations start to increase in the late 1800s and accelerate during the 20th century. Although post-depositional diagenesis may explain some of the observed recent down core changes (e.g., loss of labile fucoxanthin from siliceous algae; Leavitt 1993), the relatively stable ratio of Chl *a* (precursor) to pheophytin *a* (product) (preservation index) prior to ca. 2000 suggests little variation in pigment preservation through most of the core record (Leavitt and Hodgson 2001). Consequently, stratigraphic records of fossil pigments are consistent with a general pattern of eutrophication in both lakes.

Despite a recent history of limited terrestrial disturbance, stratigraphic analyses suggest the presence of some substantial but transient eutrophication events. In particular, Wheaton Lake may have undergone a period of eutrophication between approximately 1680 and 1750, though these dates must be interpreted cautiously as they are based on an extrapolated *rplum* ^{210}Pb age model. This interval was followed by a phase of reduced production that lasted until around 1880, after which fossil concentrations increased steadily to the present day. Elevated abundance of algae and cyanobacteria during basal Subzone W-1a was not anticipated, either in terms of timing or magnitude. This subzone is characterised by the highest historical levels of primary production (e.g., β-carotene, secondarily pheophytin *a*) and total cyanobacterial abundance, as well as marked concurrent shifts in isotope values. Specifically, the notable increase in canthaxanthin from potentially diazotrophic Nostocales, combined with a sedimentary $\delta^{15}N$ value of 2.5‰, is consistent with a substantial increase in the net N_2 fixation rates that depletes sedimentary $\delta^{15}N$ (~3‰–3.5‰) through addition of atmospheric N_2 (~0‰; Fogel and Cifuentes 1993; Gushulak, Chegoonian, et al. 2024). It is noteworthy that this subzone appears to predate the first large-scale colonial settlement in the region (starting in 1784, with the arrival of the Loyalists), even allowing for chronological uncertainty associated with the extrapolated *rplum* ages.

Climatic variability may have been responsible for boosting the downcore primary production in Wheaton Lake. For example, changes in lake water level due to long-term variation in precipitation patterns can affect production in shallow lakes, with lower water levels or increased water clarity favouring the development of benthic mats (Catherine et al. 2013; Gushulak et al. 2021) that are typically very well preserved in sediments (Leavitt and Hodgson 2001). Equally, long-term climate variability associated with the Little Ice Age (ca. 1300–1850) has been suggested to have increased lake stability, favouring the increased growth of phytoplankton, including cyanobacteria, in some unproductive ecosystems (Shampain et al. 2024). However, the absence of a similar signal in the early part of the nearby Harvey Lake core record suggests that the Wheaton Lake productivity record is not linked to regional climate variability effects on lake ecosystems. Similarly, we assume that short episodic disturbances, such as forest fires, are unlikely to have continuously influenced Wheaton Lake for the ~70 years of elevated productivity in the early 18th century. Whilst such forest disturbance can lead to an extended period of succession recovery, its effects are typically shorter-lived (Leavitt et al. 2009; but see Scully et al. 2000). Additional ^{14}C chronological control is required to fully elucidate the possible mechanisms underpinning the early eutrophication event.

Interpretation of the geochemical data provided insights into the provenance of the sedimentary matrix, and the possible causative factors underpinning the observed phototrophic shifts. At both sites, decreasing C:N values in more recently deposited sediments indicate an increased contribution of autochthonous phytoplankton to the sedimentary organic matter (Kaushal and Binford 1999). Generally, freshwater phytoplankton have a C:N mass ratio between 4 and 10, whereas vascular plants have signatures greater than 20 (Meyers and Teranes 2001). The signatures observed in both study lakes indicate an increased abundance of phytoplankton as the predominant fraction of the sedimentary matrix since at least 1900 CE. Overall, temporal trends in C:N values observed here are consistent with those in other studies that have reported C:N declines during eutrophication (McGowan et al. 2012; Bunting et al. 2016). Similarly, the depletion of sedimentary $\delta^{13}C$ observed in Harvey Lake was correlated inversely with the pigment-inferred productivity increases. This pattern is consistent with increased phototroph biomass sequestering respired CO_2 during photosynthesis (Finlay and Kendall 2007). In general, respired CO_2 in the water column is isotopically depleted (−25‰ to −30‰) relative to atmospheric CO_2 (0‰) or dissolved carbon from terrestrial carbonates (−5‰ to −10‰), such that its incorporation into, and deposition as, phytoplankton can result in reduced $\delta^{13}C$ values relative to compared materials derived from other carbon sources (Meyers and Teranes 2001; Bunting et al. 2007; Shampain et al. 2024). Unexpectedly, the declining trend in $\delta^{13}C$ was not observed in Wheaton Lake due to a transient enrichment of $\delta^{13}C$ values during the mid-20th century. At present, there is no clear causal mechanism for this event, as historical changes in all other geochemical parameters were similar amongst lakes, and pigment profiles did not exhibit concomitant variation. We speculate that this relatively narrow shift in $\delta^{13}C$ values (~1‰) may reflect changes in carbon source, catchment hydrology and forest recovery following the Bocabec forest fire in 1968. Despite this deviation, the overall pattern of geochemical change in both lakes is consistent with progressive eutrophication since the late 1800s.

4.2 | Reconstruction of Past Microcystin-Producing Cyanobacterial Blooms (Objective 2)

LC-HRMS analysis of select sediment-bound microcystins from both lakes showed that microcystin-producing cyanobacteria have occurred prior to the public advisories first issued in 2015. In Harvey Lake, the microcystins (MC-LA, -LR, -RR and -[Dha^7]LR) were below detection thresholds until after ca. 1990. However, microcystins were detected intermittently throughout the entire ~300-year record of Wheaton Lake, including at low concentrations in intervals that appear to predate European settlement. The more persistent signal of the cyanotoxins in Wheaton Lake is also consistent with the generally higher concentration of fossil pigments from potentially diazotrophic Nostocales in the core record. Some sediment core intervals with elevated TMC appear to correspond with reported bloom events. For example, in Wheaton Lake two sediment core subsections with markedly elevated toxin concentrations corresponded with documented blooms that occurred in 2015 and 2021, whilst in Harvey Lake the only documented major bloom occurred in 2015 (NB Public Health Advisories 2024).

In Harvey Lake, the contemporaneous rise in sedimentary pigments and TMC in Subzone H-2b suggests a relatively abrupt transition in lake status during the past ~25 years. In general, this signal is consistent with the elevated abundance of total cyanobacteria (echinenone) as well as spikes in colonial cyanobacteria (myxoxanthophyll) and, to a lesser extent, the Nostocales varieties (canthaxanthin) that together may represent a recent surge in cyanobacterial abundance and cyanotoxin production. In general, these pigment biomarkers are known to be chemically stable and well preserved in lake sediments (Leavitt and Carpenter 1990; Leavitt 1993). However, the observed elevated TMC also correspond to the most recent interval with high concentrations of labile fucoxanthin (siliceous algae) and Chl *a*, suggesting a potential effect of post-depositional degradation on the recent Harvey Lake stratigraphy. Whilst temporal patterns in stable biomarkers also suggest a recent increase in lake production (e.g., pheophytin *a*), the close correspondence of TMC and Chl *a*:pheophytin preservation ratios in both lakes suggests that interpretations of recent changes in toxicity must also consider potential effects of post-depositional degradation or changes in the sedimentary environment which favour preservation of labile compounds (e.g., anoxia; Leavitt and Hodgson 2001; Leavitt 1993). Fortunately, rapid post-deposition degradation is usually limited to the most recent 10–20 years and typically does not affect interpretation of older fossil deposits, particularly when Chl *a*:pheophytin ratios are invariant (Leavitt and Hodgson 2001).

Recent degradation of water quality by cyanobacterial blooms can also be inferred from the analysis of fossil pigments and microcystins from less developed Wheaton Lake. Whilst sedimentary records suggest that total lake production (as pheophytin *a*) and total cyanobacterial (echinenone) abundance are within an envelope of conditions defined by historical events, several chemically stable metrics of colonial cyanobacterial abundance (i.e., myxoxanthophyll, canthaxanthin and aphanizophyll) are more consistent with the recent expansion of cyanobacterial blooms. In particular, aphanizophyll from diazotrophic cyanobacteria (e.g., *Aphanizomenon*) is only present after the mid-1950s, suggesting the presence of modern blooms of these taxa. As this pigment is absent from the surface sediments of Harvey Lake, yet present deeper, it appears that its presence in the most recent 70 years of Wheaton Lake sediments is not solely an artefact of preservation. Instead, aphanizophyll is present as early as Wheaton Lake Subzone W-1d, prior to changes in the Chl *a*:pheophytin preservation index. Thus, whilst the influence of post-depositional losses cannot be entirely ruled out, the most parsimonious interpretation is that Wheaton Lake has experienced an expansion of the toxigenic cyanobacteria which have been present in the lake since at least 1725 CE.

Microcystin analyses provide an important insight into the history and composition of toxigenic cyanobacteria communities in the study lakes, but should be interpreted with caution. Other studies have quantified sedimentary microcystin congeners (Zastepa, Pick, and Blais 2017); however, they too provide only a limited insight into the timing and magnitude of one possible class of toxin produced by cyanobacteria. Furthermore, microcystins have been demonstrated to be subject to post-depositional degradation, with their ultimate fate varying significantly amongst congeners (Zastepa et al. 2014). For example, MC-LA exhibits lower sedimentation rates than MC-LR or MC-RR (Zastepa, Pick, and Blais 2017), but may have a longer post-depositional half-life and thus may be more persistent in lake sediments (Zastepa et al. 2014). As noted above, variation in sedimentary preservation may have also influenced the records presented here. For example, the absence of microcystins for much of the Harvey Lake record and the intermittent spikes of MC-LA downcore in Wheaton Lake may represent post-depositional diagenesis or congener-specific differences in sedimentation and preservation (Zastepa et al. 2014, 2015; Zastepa, Pick, and Blais 2017). Moreover, by only extracting sediment-bound microcystins, the total concentrations recovered may be underestimated as some congeners may have dissolved into the sediment pore waters after deposition (i.e., MC-LA and -LR) (Zastepa et al. 2014). Whilst it is important to critically appraise the limitations of such new palaeoenvironmental proxies, the observation that MCs can persist in lake sediments for millennia (Waters et al. 2021) shows that they can provide unique insights as to historical changes in the toxicity of phytoplankton communities.

4.3 | Assessing the Influence of Land-Use on Water Quality (Objective 3)

In Harvey Lake, the observed changes in pigment concentrations and shifts in geochemical signatures corresponded closely with known land-use changes in the lake catchment. The $\delta^{15}N$ and pigment data suggest that the initial establishment of the dispersed Harvey Settlement in 1837 had minimal impact on Harvey Lake as proxy levels remained relatively stable from pre-settlement sediments through the entire span of Cluster Subzone H-1a, which ended around 1900. This stability is particularly remarkable as there was rapid and extensive regional forest clearance to prepare land for agriculture, including along the shores of Harvey Lake.

During the period within Cluster Subzone H-1b, changes to catchment hydrology and nutrient cycling led to small but increasingly significant shifts in phytoplankton abundance across all taxonomic groups. Elevated abundance of fossil pigments from diatoms (diatoxanthin), cryptophytes (alloxanthin) and chlorophytes (pheophytin *b*) suggests that nutrients mobilised by runoff after forest clearance and progressively more intense land tillage increased lake biological production (after Gushulak, Mezzini, et al. 2024). The seemingly slow response of the lake to such profound changes in the catchment can be attributed to the fact that, unlike short-term events such as fires or extreme weather, fundamental ecosystem transformations (e.g., forest to field) have centennial-scale impacts on catchment biogeochemistry (reviewed in Leavitt et al. 2009).

The observed shift in Harvey Lake production after ca. 1900 is consistent with multiple anthropogenic developments. By 1910, the village had a tourism trade, with limited waste management by hotels; it is likely that nutrients made their way from the village into the local stream that flows into the lake. Establishment of a lumber mill in 1911 may have also contributed to eutrophication, as the operation accumulated a large pile of wood wastes along the southern shore during 1911–1990. Although the geochemical composition of the sawdust-derived leachate is unknown, other research suggests such effluent can be enriched in phosphorus and nitrogen-based compounds such as ammonia (Kannepalli et al. 2016) that affect phototrophic community composition (Smith et al. 1999). Of interest, demand for lumber and expected waste production increased during the 1960s; however, milling ceased in 1990 when the site could no longer accommodate more waste, and dust from the operation began to affect the health of local residents (Court of Queen's Bench 1986). The sawdust pile was removed between 2009 and 2011 (B. Matheson, pers. comm.).

Harvey Lake appears to have been affected by the Great Acceleration of the mid- 20th century when lakeshore cottages and associated developments were expanded. Additionally, increased agricultural activity after WWII resulted in the application of industrial chemicals (Russel and Williams 1977) that replaced manure as the principal fertiliser. Prior to that time, the establishment of the creamery industry in the Harvey area by the 1930s allowed the expansion of dairy herds adjacent to the lake (Old Harvey Photos 2024). Unlike manure that releases N and P slowly, chemical fertilisers release nutrients quickly, providing a rapid boost to plant growth but are easily transported by runoff into water systems (Eghball et al. 2002), negatively affecting the lake. This Great Acceleration interval is apparently recorded in Harvey Lake Subzone H-2a by increased algal and cyanobacterial abundance in the early 1960s, as well as a near-synchronous enrichment of $\delta^{15}N$ which typically reflects elevated agricultural N runoff (Anderson and Cabana 2005; Bunting et al. 2007, 2016). Nitrogen isotope enrichment occurs due to isotopic fractionation of terrestrially applied fertiliser, wherein ^{14}N is preferentially lost to the atmosphere through the processes of nitrification, denitrification and ammonia (NH_3) volatilisation (Anderson and Cabana 2005); the more fertiliser applied, the more excess N is available for microbial processing and volatilisation, and the higher the degree of enrichment and N runoff (Bunting et al. 2007). The moderate positive correlation between $\delta^{15}N$ and population change supports the application of $\delta^{15}N$ as an agricultural N runoff proxy, evidencing the impact of catchment activities on lake production.

Additional development occurred later in the 20th century. For example, the Pentecostal Tabernacle (PT) was established on Harvey Lake's northeast shore in 1953, increased to over 100 structures across five acres and attracted thousands of visitors annually. The PT closed abruptly in January 1982 when snow load caused the main building to collapse, and it was later relocated to a site away from the lake to allow upgraded waste control standards to be adhered to (K. Kinney, pers. comm., 2024). Nutrient emission via septic leakage has been documented as a potential contributor to eutrophication (Withers et al. 2011) and shifts in phytoplankton assemblages (Jinglu et al. 2007).

Agricultural activities within the Harvey Lake catchment declined after 1970, although the human population adjacent to the lake has increased as the catchment has become a commuter community for the nearby City of Fredericton. Former seasonal cottages have gradually been replaced by large, year-round homes, most with suburban-style lawns, along the lake's southern and western shores. The growing density of cottages and permanent houses led to increased septic tank usage until 2014, when sewage system infrastructure was completed to service all dwellings on Harvey Lake. Consistent with increases in near-lake nutrient sources, the abundance of cyanobacteria and their toxins has increased since the 1990s (i.e., Subzone H-2b) likely due to a shift in land-use activities from more distal farming to localised activities directly on the lake shore (Gushulak, Mezzini, et al. 2024).

4.4 | The Influence of Climate Change on Recent Eutrophication and Toxicity (Objective 4)

Climate change, exacerbated by anthropogenic forcing, has resulted in warming atmospheric temperatures and changing weather patterns (IPCC 2021; Trancoso et al. 2024). Shallow lakes are highly sensitive to climate-related perturbations due to the close mechanistic links between atmospheric and terrestrial processes which influence aquatic ecosystem structure and function (Williamson et al. 2009; Meerhoff et al. 2012). The climate trends presented here for each lake suggest warmer and generally wetter conditions in southern New Brunswick since 1950, albeit at slightly differing rates between the inland and coastal sites. Equally, analysis of lake ice phenology indicates a trend towards later ice-on and earlier spring ice breakup between 1880 and present. The occurrence of this warming trend, concurrent with known land-use intensification and the modification of the terrestrial landscape, makes untangling the proportional significance of climate- and human-mediated stressors difficult (Hofmann et al. 2021). In Harvey Lake, temperature variables were moderately to strongly correlated with lake production and microcystin concentration, a pattern which suggests that, in addition to anthropogenic activity, climate warming may have influenced water quality degradation in this system. Furthermore, the strong negative correlation between pigment PCA 1 and Ice-out dates suggests that open water season length could be a significant control on production in this ecosystem. By comparison, the recent rise of cyanobacterial abundance and microcystin concentrations in Wheaton Lake did not correspond with known nutrient enrichment or land-use change, whilst instead moderate positive correlations with temperature and precipitation variables suggest that recent climate variability may have favoured increased production and cyanobacteria toxicity in this lake. Furthermore, the lower magnitude of climate-fossil correlations in Wheaton Lake may reflect the moderating influence of the Bay of Fundy. Overall, these findings are supported by those of Daly et al. (2019), in a similar impact/reference lake investigation, who suggested that shifts in lake thermal properties under climate change have impacted lake ecological structure and zooplankton communities in northern New Brunswick lakes.

There are several mechanisms by which regional climate change may influence phytoplankton communities to favour cyanobacteria. First, cyanobacteria have been observed to have a higher thermal optima compared to eukaryotic algae (Butterwick et al. 2005), potentially leading to a higher proportion of cyanobacterial pigments relative to those from diatoms and cryptophytes. Second, warmer atmospheric temperatures since the 19th century have resulted in thinner winter lake ice, a longer open water season and deeper or more intense stratification in lakes across Canada. Some bloom-forming cyanobacteria, such as *Microcystis*, can regulate their buoyancy, enabling them to float or sink in stratified waters, harvesting light and nutrients at various water column depths (Jöhnk et al. 2008; Paerl et al. 2011). Third, increased precipitation may enhance the mobilisation of nutrients from the catchment (Ho and Michalak 2020). Fourth, persistent increases in extreme weather events (i.e., hurricanes and tropical storms) may favour cyanobacterial development (Stockwell et al. 2020) due to significant disruptions to abiotic and biotic conditions by exceptional rainfall and winds (Klug et al. 2012; Yang et al. 2016).

A warmer future climate may increase the frequency and intensity of storm events, particularly at higher latitudes (Patterson, Nasser, Reinhardt, et al. 2022). Although circumstantial, we note that the first ever reports of cyanobacterial blooms in Harvey, Wheaton Lakes and other regional lakes emerged in 2015, the summer following Subtropical Storm Arthur, which passed directly over these sites. The storm significantly deepened the wave base of Harvey Lake, resuspended surficial sediments and increased turbidity, resulting in discoloured water for over a week and the release of sedimentary nutrients into the water column (Patterson et al. 2020; Atasiei et al. 2022; Patterson, Nasser, Reinhardt, et al. 2022). Consistent with this mechanism, Yang et al. (2016) noted that the passage of typhoons over Lake Taihu, China, was correlated positively with nutrient increases and subsequent cyanobacteria bloom events. Thus, we hypothesise that Hurricane Arthur may represent an ecological tipping point which led to the proliferation of microcystin-producing cyanobacterial blooms, although further research is needed to validate this.

5 | Conclusions

Although the influence of land-use change on lake productivity has been well documented in palaeolimnological research, its impacts on historical patterns of microcystin production have only been addressed in a few other studies. The analysis of microcystins, fossil pigments and geochemical markers presented herein provides the first historical insights into how toxigenic lake cyanobacteria blooms have responded to land-use and climate change from Atlantic Canada. Analysis of sediment-bound TMC from both studied lakes indicates cyanotoxin presence decades to centuries earlier than previously thought, although the frequency and magnitude of toxic events appear to have increased significantly in the last 20 years. For nearly 200 years, land-use changes surrounding Harvey Lake appear to have increased lake productivity due to a combination of agriculture, urbanisation, industrialisation, cottage development and, since the 1990s, permanent waterfront homes. Given the differences in the land-use histories between our two study lakes, we infer that the contemporary eutrophication signal and increased microcystin concentrations in reference Wheaton Lake arise from the effects of climate variability on lake structure and function. This research adds to the growing literature suggesting that modelled future warming temperatures and projected changes in major storm tracks present a substantial threat to freshwater resources, even in lakes which are relatively unimpacted by human activities. Further, the consistent presence of microcystins in the sediment records over the last two decades suggests that 'recent' increased cyanobacterial toxicity may be underestimated in many other systems globally due to the bias towards only testing lakes with active surface blooms. This observation makes regular water quality monitoring advisable, especially for lakes used for recreational activities and municipal drinking water.

Author Contributions

Conceptualisation: D.A.H., H.M.R., R.T.P., P.R.L., D.R.M. and S.A.B. Developing methods: D.A.H., H.M.R., R.T.P., P.R.L., E.T.M. and D.R.M. Conducting the research: D.A.H., H.M.R., R.T.P., P.R.L., E.T.M. and D.R.M. Data analysis: All authors. Data interpretation: All authors. Preparation of figures and tables: All authors. Writing: All authors.

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Conflicts of Interest

The authors declare no conflicts of interest.

Data Availability Statement

Data are available from the authors upon reasonable request.

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Supporting Information

Additional supporting information can be found online in the Supporting Information section.