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Chloride trends in Ontario's surface and groundwaters

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ABSTRACT

The Ontario Ministry of the Environment, Conservation and Parks leads 11 long-term monitoring programs at over 2,500 surface and groundwater sampling locations across the province that report chloride (Cl^-) concentration, some dating back to the 1960s. This study integrates these disparate datasets to provide comprehensive evidence relevant to spatial and temporal Cl^- trends in the Laurentian Great Lakes, and Ontario's inland lakes, streams and groundwaters. While the vast majority of historical Cl^- data indicate concentrations are well below the chronic exposure Canadian Water Quality Guideline (120 mg L^{-1}), many lake, stream and groundwater sampling locations proximal to roadways or in urbanized areas meet or exceed this guideline, and concentrations in these regional areas are persistently increasing. The current evidence implicates road salting activities for winter safety as a primary contributor to elevated Cl^- concentrations, and the trends may be exacerbated with urbanization and population expansion, particularly in southern Ontario.

Introduction

Lakes are often considered harbingers or sentinels of change in the environment (Adrian et al., 2009). Though lakes can vary across a spectrum in trophic status, mixing regime and morphometry, all are sensitive to climate, respond to changes in the environment, and importantly, integrate information about land-based activities within watersheds. Lakes can receive anthropogenic inputs, including contaminants, from both aquatic and terrestrial sources. To best understand and mitigate contaminant threats to ecosystem health in large water bodies, such as the Great Lakes, comprehensive spatial and temporal ecosystem-scale monitoring is required, inclusive of all source waters to the receiving lake environments. Information on trends in environmental conditions is essential background for the management of water quality on local to regional scales. In addition, monitoring through time allows for the identification of anomalous patterns or conditions that may signal adverse changes due to stressors of known or unanticipated impacts.

There exists increased awareness regarding historical trends and changes in Ontario surface and groundwater chloride (Cl^-) concentrations, as well as the impacts of elevated Cl^- on drinking water sources, aquatic organisms and their habitats (Ontario Ministry of the Environment, Conservation and Parks, 2019). Contributions of Cl^- from winter maintenance activities is an emerging issue in Ontario where evidence shows that the primary anthropogenic source of Cl^- to aquatic environments is runoff from deicers or road salts, commonly applied as sodium- ($NaCl$), magnesium- ($MgCl_2$), calcium- ($CaCl_2$) or potassium chloride (KCl) (Environment Canada and Health Canada, 2001). Additionally, Cl^- (as $CaCl_2$) is often used in water softeners in regional areas where drinking and tap waters are enriched in minerals (Health Canada, 2008).

Through various routine monitoring programs, the Ontario Ministry of the Environment, Conservation and Parks (MECP) has amassed a large pool of data on Cl^- concentrations in the Laurentian Great Lakes, inland lakes, streams, rivers and groundwaters dating back to the 1960s from over 2,500 monitoring stations representative of the diverse land use and physiographic features of the province of Ontario. These programs include Ontario's largest

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historical surface and groundwater monitoring networks. The historical monitoring data span important time periods where advancements in our understanding of anthropogenic activities and surface and groundwater quality have led to bi-national proactive measures, including the Great Lakes Water Quality Agreement (GLWQA) in 1972. Despite historical initiatives that were implemented to maintain water quality and ecosystem health, contaminants remain ubiquitous in the Laurentian Great Lakes basin; many of these contaminants accumulated for decades before reaching levels of concern or for negative ecosystem symptoms to emerge (Cornwall et al., 2015).

This study summarizes decades of Cl^- monitoring across the province of Ontario to provide critical insights into the current state of Cl^- levels in surface and groundwaters. We present long-term trends, identify specific time periods of change in aquatic Cl^- conditions, and compare Cl^- concentrations in aquatic environments across land use and physiographic features and to the Canadian Council of Ministers of the Environment Canadian Water Quality Guidelines for the Protection of Aquatic Life (CWQG; CCME, 2011). Concise reporting on Cl^- in Ontario surface and groundwaters will provide the scientific evidence required to best inform policy and mitigation efforts to preserve and protect aquatic ecosystem health in the Laurentian Great Lakes basin.

MECP programs and methods

Cl^- data presented herein are compiled from multiple monitoring programs that are summarized in Table 1. As MECP collects Cl^- data from ambient monitoring programs, concentrations presented may underestimate the full range of conditions in aquatic environments as event-based runoff periods were not specifically targeted for sample collection; seasonal base flow and event flow in streams and rivers, and seasonal periods of thermal stability and instability in lakes were captured according to program routine sampling schedules, described herein.

Streams and rivers

The Provincial (Stream) Water Quality Monitoring Network (PWQMN) was created under the Ontario Water Resources Commission and is currently delivered by MECP, in collaboration with Conservation Authorities and the Severn Sound Environmental Association. Over 400 stream locations are currently sampled by partners on an approximately monthly basis and analyzed for a suite of water quality indicators, including pH, dissolved oxygen, conductivity, nutrients, metals and Cl^-. Data from the PWQMN are used for tracking long-term trends in water quality and for special studies in support of source protection planning and nutrient, pesticide and road salt (Cl^-) management. Spatial and temporal patterns in Cl^- in rivers and streams were evaluated at a broad scale using this large network of monitoring sites across the province and also at a watershed scale along a gradient of increasing population density in the Muskoka River.

Data from the PWQMN were screened to include only samples collected during the ice-free season (May to November); sites and years with three or more samples; sites that were sampled in the latest available data year (2019); and sites that had at least 30 years of data (although not necessarily a continuous yearly record). Data from a total of 214 sites were used. Annual mean Cl^- concentrations were calculated for each site and generalized additive models (GAMs) were used to fit the annual mean concentration time series with the smoothing parameter (k) set to the number of years of data for each site divided by four. GAMs were used to analyze temporal trends because they allow for identification of time periods of statistically significant change, that were identified using the first derivative of the fitted model (Simpson, 2018). Land use in the upstream watershed of each sampling site was derived from Ontario Flow Assessment Tool (2021) and used to group sites; k-mean clustering identified three groups, that were named accordingly (agriculture, urban and treed/mixed).

The Muskoka River watershed has an area of 5,000 km^2 and a population of 60,000. Eight sampling locations are placed along the river length (120 km) that drain into Georgian Bay of Lake Huron. Water quality samples, including Cl^- concentration, were collected monthly at eight selected locations. Three locations are on the less-developed upper reach of the river, two are on the middle reach and three are on the lower reach that has relatively higher development. Samples were collected in 1983, and again in 2015 – 2017 for comparison purposes. The annual mean values for each collection period and the percent change between collection periods were calculated.

Inland lakes

Two datasets with broad geographic coverage were used to demonstrate the spatial patterns in Cl^- concentration in inland lakes: the Lake Partner Program and Broad-Scale Monitoring Program. Data from more intensive monitoring in the Boreal Shield ecozone, an area with a relatively high density of lakes, was used to evaluate spatial patterns in Cl^- with respect to possible sources. Long-term trends were also evaluated in Lake Simcoe, a large lake that has experienced increasing human activity in its watershed (Palmer et al., 2013).

The Lake Partner Program (LPP) is Ontario's community science lake monitoring program that provides lake water quality and trophic status information for cottage and recreational lake users. LPP volunteers collect lake water samples for chemistry, including Cl^- concentration, during the spring turnover period (May) and water clarity measurements (e.g., Secchi disc readings) bi-weekly throughout the summer for lakes "on" the Canadian Shield. For lakes "off" the Canadian Shield, one water sample is collected for chemistry each month from May to October and water clarity measurements are collected bi-weekly over the same time period.

Broad-Scale Monitoring (BSM) is an Ontario Ministry of Northern Development, Mines, Natural Resources and Forestry (MNDMNRF) program in collaboration with MECP. BSM provides detailed fisheries and water quality information from hundreds of inland lakes that support a recreational sport fishery. Water samples are collected for chemistry (including Cl^- concentration) from selected lakes during the spring months each year of the BSM sampling cycle. Additional sampling through the BSM includes netting for fish, checking for the presence of invasive species, conducting angling effort counts and collecting bathymetric depth data for the lake.

Spatial patterns in Cl^- concentration across the province of Ontario were evaluated using data from LPP and BSM. For both programs, samples used for analysis were collected in the spring, often April or May, for the years 2015 – 2019 from the deepest spot of the lake using an integrated water sampler lowered to a depth equivalent to the Secchi depth. Average Cl^- concentrations for 2015 – 2019 were calculated for each lake. If a lake was sampled only once in any of these years, then that year's data point was used to represent the final Cl^- concentration. If a lake had multiple sampling sites, then the mean Cl^- concentration for all sampling sites was calculated to represent the whole-lake Cl^- concentration. The initial pooled dataset included a total of 1,480 lakes across Ontario: 689 LPP and 791 BSM lakes. A map was generated from these data to examine the spatial patterns in Cl^- concentrations across the province and possible association to roads (highways).

Trends and spatial patterns were also evaluated at a smaller spatial scale in lakes near the city of Sudbury, Ontario monitored

Table 1
Summary of monitoring programs from which chloride data were compiled.

Geography monitored	Program or data source	Years active[1]	Frequency of sampling [2]	Data and information links
Great Lakes, nearshore	Great Lakes Nearshore Long-Term Monitoring Program	1994 – ongoing	Seasonal on 3-year cycle	https://data.ontario.ca/dataset/water-chemistry-great-lakes-nearshore-areas
Great Lakes and Lake Simcoe, via intake pipes for drinking water facilities	Great Lakes Intake Program (GLIP)	1976 – ongoing	Weekly – bi-weekly (year-round)	https://data.ontario.ca/dataset/lake-water-quality-at-drinking-water-intakes
Great Lakes, inland lakes, tributaries and groundwater, via intake pipes for drinking water facilities	Drinking Water Surveillance Program (DWSP)	1986 – ongoing	Yearly (year-round)	https://data.ontario.ca/dataset/drinking-water-surveillance-program
Lake St Clair, Detriot and Thames rivers	Lake St. Clair	2016 – 2019	Monthly	Data available on request
Streams and rivers (province-wide)	Provincial (Streams) Water Quality Monitoring Network (PWQMN)	1964 – ongoing	Monthly – seasonally	https://data.ontario.ca/dataset/provincial-stream-water-quality-monitoring-network
River, Boreal Shield ecozone	Muskoka River monitoring	1983, 2015 – 2017	Monthly	Data available on request
Inland lakes (province-wide)	Lake Partner Program (LPP)	2015 – ongoing	Monthly – bi-weekly	https://data.ontario.ca/dataset/ontario-lake-partner
Inland lakes (province wide)	Broad Scale Monitoring (BSM)	2008	Annual over 2–3 year cycle	Data available on request https://www.ontario.ca/page/broad-scale-monitoring-program
Inland lakes, Boreal Shield ecozone	Inland Lakes Monitoring and Assessment (DESC A, Sudbury area)	1976 – ongoing	Bi-weekly - monthly	https://data.ontario.ca/dataset/inland-waters-lakes-and-streams-water-chemistry
Lake Simcoe	Lake Simcoe Monitoring Program	2000 – ongoing	Bi-weekly	https://data.ontario.ca/dataset/lake-simcoe-monitoring
Groundwater (province-wide)	Provincial Groundwater Monitoring Network (PGMN)	2001 – ongoing	Annual	https://data.ontario.ca/dataset/provincial-groundwater-monitoring-network

[1] Duration of routine chloride monitoring indicated; timespan of other parameters and among locations may vary.
[2] All programs except those indicated sample in the ice-free season only.

by the MECP and the Laurentian University Vale Living with Lakes Centre. Lakes in this area of the Boreal Shield ecozone were historically impacted by nearby smelting operations, but lake acidification has improved following sulphur emission control programs. Recently, monitoring in this area has specifically focused on lakes with pH < 5.5 and a set that did not historically acidify that acts as a regional reference. Additional intensive monitoring complements the core monitoring by sampling more frequently at a subset of lakes for a broader suite of limnological and biological attributes. This intensive sampling is conducted monthly or bi-weekly from May to October. Data presented here contain a set of one mid-summer Cl^- concentration for each of 82 lakes, including 16 urban lakes and 66 non-urban (remote) lakes. Lakes were classified as urban when near urban or residential centres or major roadways (Fig. 1). Remote lakes included wilderness areas or those with minimal direct human influence.

Additional monitoring, part of MECP's inland lakes monitoring and assessment program, in the Boreal Shield ecozone takes place in eight lakes (nine basins) and their watersheds in the Muskoka-Haliburton region of Ontario (Palmer et al. 2014). The lakes, coined the "Dorset A lakes", have been sampled intensively since the 1970s for physical, chemical and biological data, including lake Cl^- concentrations. Water samples are collected at discrete depths using a pump and hose method to allow samples to be analyzed in a volume-weighted fashion, based on lake bathymetry. Data represent whole-lake, volume-weighted numbers that are averaged from bi-weekly (Dickie, Harp and Plastic lakes) to monthly (all remaining lakes) samples to create an ice-free mean for each year.

To investigate anthropogenic impact, the Dorset A lakes can be intrinsically sub-divided into two broad categories: 1) remote lakes without any winter-maintained roads within their watersheds (Blue Chalk, Red Chalk, Chub, Crosson and Plastic lakes) and 2) lakes with surrounding roads that are maintained by local municipalities throughout the winter (Dickie, Harp and Heney

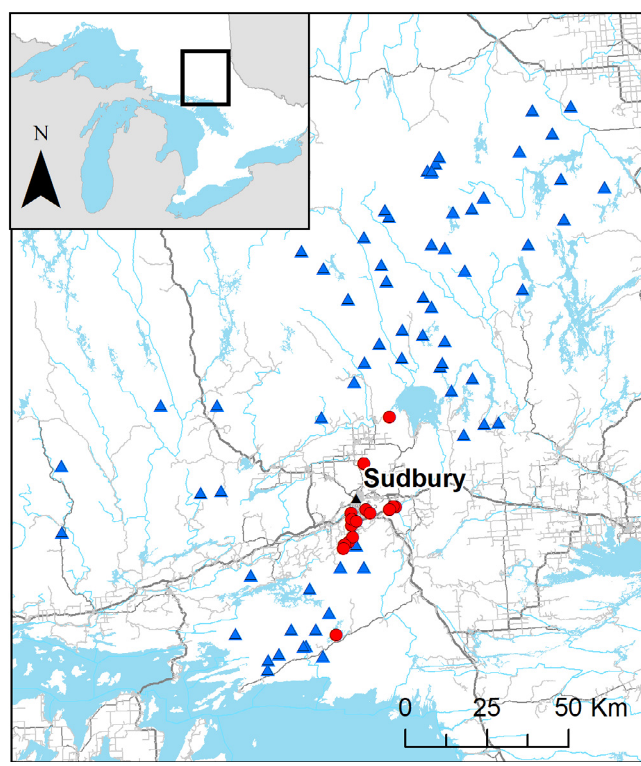

Fig. 1. Location of 66 non-urban (blue triangles) and 16 urban lakes (red circles) in the Sudbury area of south-central Ontario, Canada surveyed in 2018. The light grey and dark grey lines indicate collector roads and highways, respectively (ORN, 2018). Inset map shows the location of the Sudbury area within Ontario. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

lakes). For the comparison of remote lakes vs. those with winter-maintained roads, volume-weighted ice-free Cl^- data were standardized using Z-scores.

Lake Simcoe is a large inland lake (722 km^2) in the Boreal Shield Ecozone that has undergone significant change in response to increasing human activity in its watershed (Palmer et al., 2013). Following concerns in the 1970s of water quality deterioration and coldwater fish declines, the MECP and MNDMNRF began research and monitoring of the lake's water quality and biota. These efforts continue today along with management actions in collaboration with many partners under the 2009 Ontario Lake Simcoe Protection Plan.

Data presented here are annual (May through October) Cl^- average from samples collected monthly at the Lake Simcoe outflow (Atherly Narrows) since 1971, weekly from the water intake pipe at two drinking water facilities (since early 1980s), and bi-weekly at Cook's Bay since 2000, collected as part of Provincial Water Quality Monitoring, Great Lakes Intakes and Lake Simcoe Protection Plan Monitoring programs, respectively. Years when sampling frequency was less than four months were excluded and a Mann-Kendall trend test was used to detect significant monotonic trends in historical Cl^- concentration. A Mann-Kendall test was selected as it is a robust test for trends in time series with missing values (Hirsch and Slack, 1984).

Laurentian Great lakes

Data from several programs were compiled to assess the state of and the trends in Cl^- concentrations in the Laurentian Great Lakes. Spatial patterns were assessed with the Great Lakes Nearshore Long-Term Monitoring Program, which has a large network of stations along the Canadian shore. Long-term trends were assessed using data from two programs that have year-round sampling at a weekly or quarterly frequency from the intake pipes of drinking water facilities that draw water from the Great Lakes. Additional data from a multi-year study in Lake St. Clair were used to describe gradients in Cl^- into and out of Lake St. Clair.

Land use and physiography along the shoreline of the Laurentian Great Lakes is varied, so the monitoring approach of the Great Lakes Nearshore Long-Term Monitoring Program includes both index and reference stations; reference stations are locations where background conditions for a lake sub-area prevail; index stations are located in areas with a natural integration of stressors such as delta zones, depositional zones, an embayment and other areas where prevailing circulation patterns focus stressors. Surveys are conducted on a cyclical basis: the lower lakes (Erie and Ontario) and their connecting channels are generally surveyed every three years (reflecting the higher level of anthropogenic stress in the lower lakes), while the upper lakes are surveyed approximately every six years. Three surveys (spring, late summer and fall) collect data for water and sediment quality, and benthic invertebrate composition. Water column sampling pertaining to the Cl^- data consists of three depth-integrated samples collected over the euphotic component of the epilimnion, approximated as surface to the lesser of 2 $\times$ the Secchi depth, or to the top of the metalimnion. Average ($n = 3$) water column Cl^- concentrations collected in the most recent cycle of the Great Lakes Nearshore Long-Term Monitoring Program are presented. Most recent lake sampling years were: Lake Superior (2019); Lake Huron and Georgian Bay (2015); Lake Erie (2019) and Lake Ontario (2018). Standard deviations were calculated for each season's replicate samples at each index and reference station.

Cl^- values from two MECP programs were pooled into a combined dataset to look at long-term trends in the Great Lakes nearshore: the Great Lakes Intake Program (GLIP) and the Drinking Water Surveillance Program (DWSP). Both programs collect samples of untreated lake surface water from the intake pipes of drinking water facilities to track long-term changes in lake conditions as they are represented at a fixed location and depth. A long-term record of trophic indices (i.e., phytoplankton) and water quality (e.g., nutrients, chloride, dissolved organic carbon) is available for 18 locations in the Laurentian Great Lakes and Lake Simcoe that are monitored by the GLIP. Of these locations, 16 have been sampled year-round at least bi-weekly for the past 20 years, in some cases since the 1960s. Sites in this program are located away from localized sources of anthropogenic disturbance to strategically represent nearshore conditions in the broader area (i.e., lake basin). Long-term monitoring of surface water Cl^- concentrations is also available for at least the last 20 years at 44 locations across Ontario (representing nine inland lake locations, 15 locations in the Great Lakes and connecting channels, and 20 locations on rivers) through DWSP. Between two to nine sets of samples are collected per year at each DWSP sampling location and analysed for radiological, organic and inorganic parameters, including Cl^-. While 24 drinking water systems with groundwater sources are sampled under DWSP, only surface water sources in the Great Lakes and connecting channels were included in this analysis for simplicity. To prevent duplication, DWSP data were excluded at locations that also participate in GLIP. For both programs, only sites with $\geq$ 20 years of data (up to 2019) were included in plots. Recent trends (2010 – 2019) in Cl^- concentrations at each of the 16 Great Lakes intakes with bi-weekly sampling were examined for monotonic trends using Mann Kendall (MK) trend tests using the R package 'Kendall' (McLeod, 2011).

Lake St. Clair is a large shallow mesotrophic lake and is part of the Laurentian Great Lakes. From 2016 to 2019, the MECP in partnership with the Water Quality and Monitoring Surveillance Division of Environment and Climate Change Canada (ECCC) undertook a water quality and harmful algal bloom assessment of the Lake St. Clair, the Thames River and the upper Detroit River to characterize and examine the water quality patterns and drivers of harmful algal blooms. Over 1,700 discrete water samples across 200 sampling sites, typically from 0.5 to 1.5 m sample depths, were collected for Cl^- analysis. Samples were collected between May and October from 2016 to 2019. To examine the gradient of Cl^- into and out of the lake, sampling sites were broadly classified according to geographic location: Lake St. Clair, Thames River or Detroit River.

Groundwater

The Provincial Groundwater Monitoring Network (PGMN) is designed to collect, analyze and report on ambient groundwater quantity and quality conditions across Ontario. MECP leads and coordinates the PGMN in collaboration with all 36 Ontario CAs and 10 municipalities outside CA jurisdictions. The PGMN currently consists of 397 active wells that are selected for long-term groundwater quality sampling.

PGMN monitoring wells are not used for private or public drinking water supplies but do provide information on groundwater aquifers that may supply drinking water. The data provide an opportunity to assess conditions against standards and guidelines, recognize trends and assess potential correlations with changes in weather or land use activities. Sampling is completed on an annual basis (fall season) and analyses are completed at the MECP Laboratory Services Branch for general chemistry, major ions including Cl, nutrients and metals.

Trend analysis of the PGMN Cl^- data was completed by MECP hydrogeologists and examined using the MK trend test using the R package 'Kendall' (McLeod, 2011), followed by manual inspection of the data. Strong increasing or decreasing trends were identified where tau (trend direction and strength) was < -0.5 or > +0.5 and p

(probability of null hypothesis being valid, or the likelihood that a monotonic trend exists) < 0.05. Where concentrations were generally < 10 mg L^{-1} it was noted that the trend was likely to be statistically non-significant. Where -0.5 > tau > 0.5 and p > 0.05, wells were classified as 'stable non-trend' or 'variable' via manual inspection.

Sample analysis and data quality control

Three analytical methods were used to measure Cl^- concentration in surface and groundwater samples. All samples from Great Lakes programs, as well as provincial-scale groundwater and stream monitoring (PGMN and PWQMN) were analysed using colorimetric methods by MECP Laboratory Services Branch according to MECP (2020b). An additional method using ion chromatography was used for a portion of the samples in the Lake St. Clair study by the National Laboratory for Environmental Testing (NLET 01–1081, as in Chapra et al. 2009). With the exception of Lake Simcoe monitoring, all remaining inland lake and river monitoring programs analysed samples using ion chromatography by the MECP Dorset Environmental Sciences Centre according to MECP (2019b). Samples collected from intake pipes of drinking water facilities in Lake Simcoe and at the lake outflow were analyzed according to MECP (2020b) methods, and samples collected in the lake (i.e., Cook's Bay) were analyzed according to MECP (2019b).

All labs that generated data have participated in regular proficiency testing and interlaboratory comparison. Calibration ranges for methods using ion chromatography were 0.01 – 90 mg $L^{-1}Cl^-$ (NLET) and 0.05 – 110 mg $L^{-1}Cl^-$ (MECP Dorset Environmental Sciences Centre). The colorimetric method used by the MECP Lab-

Table 2
Mean ± standard deviations of Cl^- concentrations measured in the Provincial (Stream) Water Quality Monitoring Network (PWQMN) by decade. n = number of annual data points (site × year) and watershed land cover.

Decade	Land Use Mean ± SD (n = number of annual data points)		
	Agriculture	Treed/Mixed	Urban
1960–1969	38.64 ± 47.20 (n = 201)	7.65 ± 10.18 (n = 106)	N/A*
1970–1979	37.64 ± 79.16 (n = 917)	9.35 ± 16.11 (n = 416)	110.96 ± 52.46 (n = 15)
1980–1989	43.04 ± 100.86 (n = 1301)	11.11 ± 17.12 (n = 413)	129.6 ± 66.76 (n = 44)
1990–1999	42.58 ± 36.63 (n = 1162)	10.63 ± 14.3 (n = 412)	169.09 ± 86.69 (n = 44)
2000–2009	52.58 ± 44.49 (n = 1321)	14.20 ± 20.35 (n = 464)	217.68 ± 86.34 (n = 56)
2010–2019	54.97 ± 46.90 (n = 1544)	17.36 ± 24.81 (n = 566)	272.71 ± 126.92 (n = 60)

* No urban streams were monitored from 1960 to 1969 (N/A).

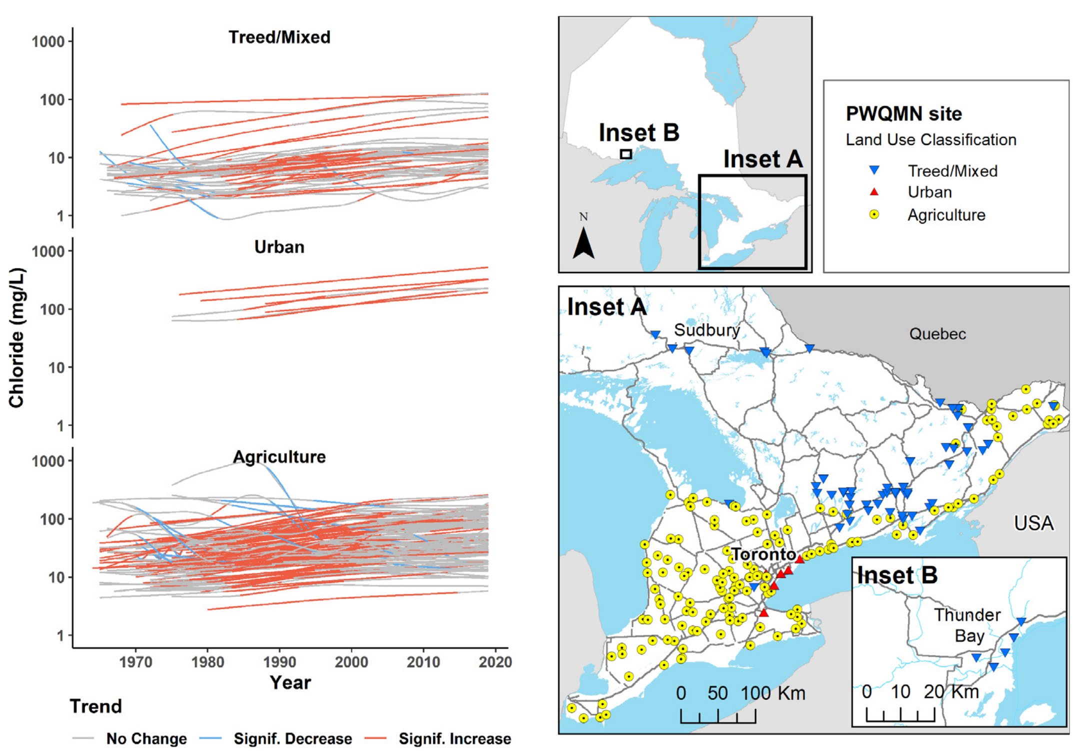

Fig. 2. Temporal trend lines fit using a generalized additive model of Cl^- concentration over time in surface water samples collected under the Provincial (Stream) Water Quality Monitoring Network (PWQMN) at 214 sites meeting data inclusion criteria, grouped by dominant land cover. Maps on the right indicate location of the sites in Ontario, Canada in the south of the province (Inset **A**) and near Thunder Bay (Inset **B**).

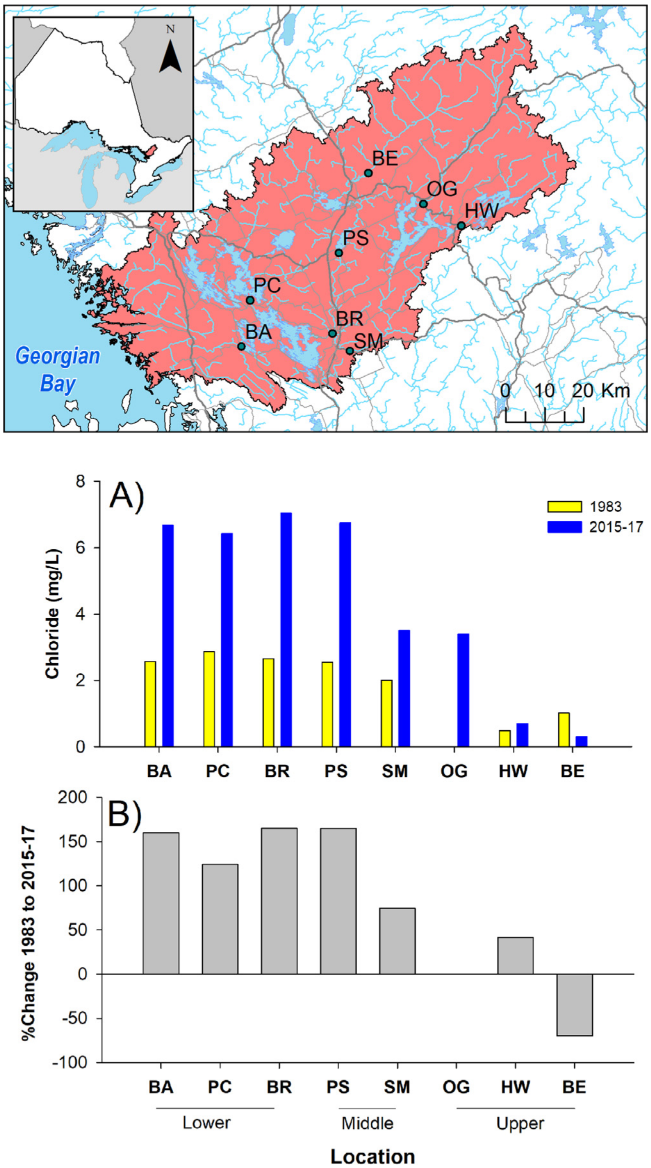

Fig. 3. Location of eight water chemistry sampling sites along the Muskoka River, central Ontario, showing proximity to development as represented by highways, collectors and arterial roads. Inset map shows the location of the Muskoka River watershed within Ontario. Sites are grouped into the upper reaches: Big East River (BE), Hollow River (HW) and Oxtongue River (OG); the middle reaches: Southern Muskoka River Branch at Martinsville (SM) and Northern Muskoka River Branch at Port Sydney (PS); and the lower reaches of the Muskoka River: Bracebridge (BR), Port Carling (PC) and Bala (BA). Annual mean Cl^- concentrations at each site in 1983 and 2015 – 2017 are shown in panel **A** and percent changes of Cl^- concentration between two periods (1983 vs. 2015 – 2017) are shown in panel **B**.

oratory Services Branch had a calibration range of 1 – 100 mg L^{-1}-Cl^- until 2014, when the range increased to 200 mg L^{-1}Cl-. Samples outside the calibration ranges were diluted to within calibration ranges prior to analysis. Analytical methods used by NLET and the MECP Laboratory Services Branch were compared using a subset of samples from the Lake St. Clair study. A paired *t*-test indicated no significant difference in results (N = 297, p = 0.02).

QA/QC was performed at the laboratory level for all datasets. Program-specific screening for outliers varied, but all considered historic ranges in addition to possible causes for extreme values. Maps indicating the proximity of sampling locations to road maintenance activities were generated in the software ArcMap version 10.6.1 (ESRI, 2020) with spatial data from the Ontario Road Network (ORN, 2018).

Results

Streams and rivers

Ontario streams monitored under the Provincial (Stream) Water Quality Monitoring Network (PWQMN) showed increasing Cl^- concentrations from the 1960s to 2019 (Table 2). Stream sites with upstream land use dominated by treed or mixed land use, typically located in the central, less developed area of the province (Fig. 2), showed the lowest overall Cl^- concentrations. Mean concentrations increased from a mean of 7.65 mg $L^{-1}Cl^-$ in the 1960s to 17.36 mg $L^{-1}Cl^-$ in the 2010s. Sites dominated by agricultural land, which were typically located in southern Ontario, increased from 38.64 mg L^{-1} to 54.97 mg L^{-1} over the same time period. Urban sites, primarily located in the Greater Toronto Area, increased from 110.96 mg L^{-1} in the 1970s to 272.71 mg L^{-1} in the 2010s (Table 2). Periods of significant change were most frequently observed in the period between 1980 and 2000 (Fig. 2). Overall, Cl^- concentrations in the streams monitored increased by approximately 2-fold from the 1960s to today.

Within the Muskoka River watershed, increasing spatial Cl^- concentrations were observed along a gradient from the less-developed upper river reach to the more developed middle and lower river reaches. Cl^- concentration increased from 0.3 mg L^{-1} at site BE to 6.7 mg L^{-1} at BA, a longitudinal increase of 21 × along the monitored river length (Fig. 3A). Temporal changes within the Muskoka River were noted where in 1983, Cl^- concentration was ∼ 2 mg L^{-1} at the middle and lower reach sites (the OG site was not measured) and increased to ∼ 6 mg L^{-1} in 2015 – 2017, with only BE experiencing a reduction in Cl^- concentrations from 1983 to 2015 – 2017. Cl^- concentration increased by 40 – 160% at the six sampling sites over a period of 35 years (Fig. 3B).

Inland lakes

The spatial analyses of LPP and BSM provincial-scale data indicates that the lakes with the highest Cl^- concentrations were commonly situated near urban centres, adjacent to highways, and tended to be in more populated regions of the province (Fig. 4). Most lakes (79%) had Cl^- concentrations below 5 mg L^{-1}; 19.5% had concentrations between 5 and 40 mg L^{-1}; and 0.5% had concentrations above 40 mg L^{-1}.

In the five remote Dorset A lakes, volume-weighted Cl^- concentrations were consistently below 1 mg L^{-1} throughout the data record (Fig. 5A). In contrast, in the three Dorset A lakes where nearby roads are maintained by local municipalities through the winter, Cl⁻ concentrations increased over the data record, peaking in the early-to-mid 2000s and declining slightly thereafter (Fig. 5B). Peak Cl^- concentrations observed in the early-to-mid 2000s ranged from ∼ 1 to 7 mg L^{-1}. The remote Dorset A lakes displayed inter-decadal variability that was consistent across lakes while lakes with winter-maintained roads within their watersheds displayed a rise in Cl^- concentrations in the 1980s and 1990s, followed by a decline in Cl^- concentrations since the mid-2000s (Fig. 5B).

Similar to the Dorset A lakes, low pH (<5.5) lakes in the less-developed area near Sudbury show low Cl^- concentrations, below 1 mg L^{-1}, with little variation among lake Cl^- concentrations in this lake category (Fig. 6A). The few lakes with Cl^- concentrations between 1 and 3 mg L^{-1} in this area have major roadways elsewhere within their watersheds and not in direct proximity, or have

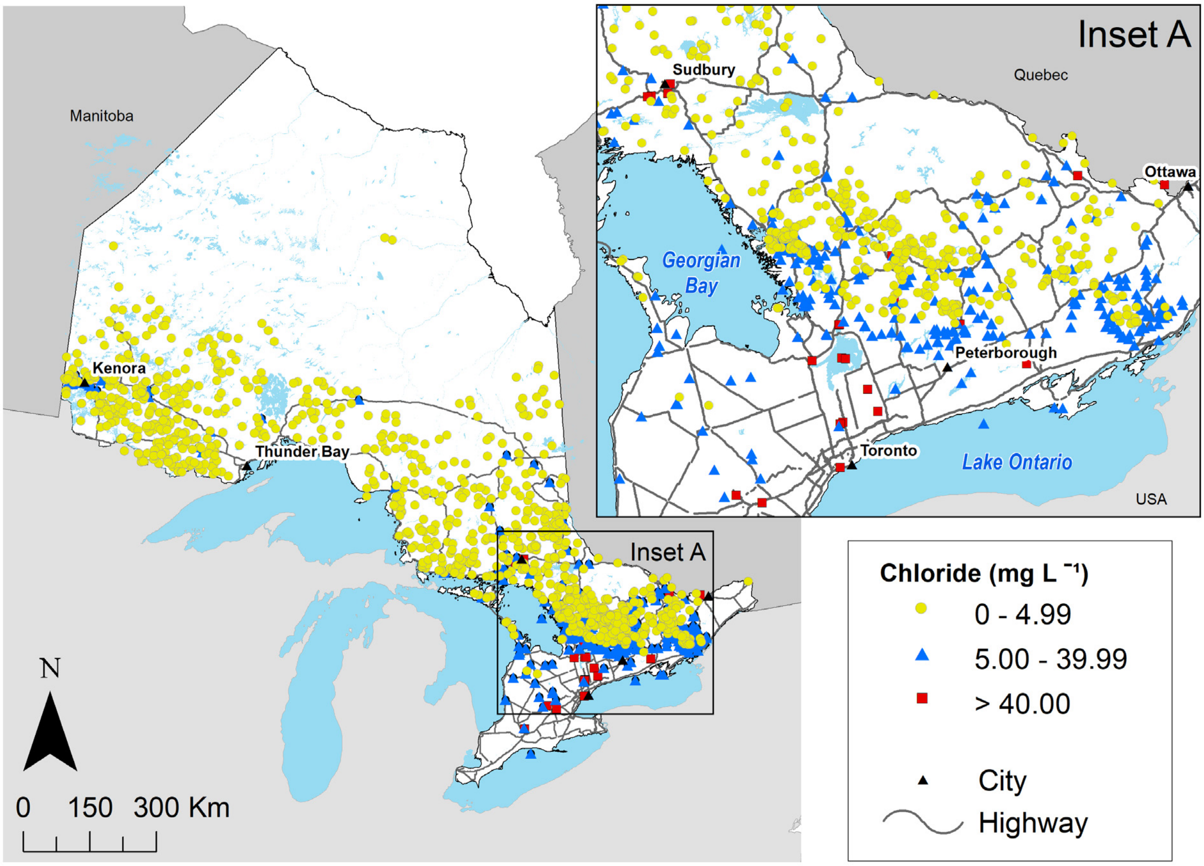

Fig. 4. Ontario, Canada map depicting the distribution of lake Cl^- concentrations sampled by the Lake Partner Program (LPP) (n = 689) and Broad-Scale Monitoring (BSM) (n = 791) in the spring (April and May) of the years 2015 – 2019. The area inside inset **A** is enlarged to detail the distribution of lake Cl^- concentrations for south-central Ontario and the proximity to highways. Selected cities are labelled for geographic reference.

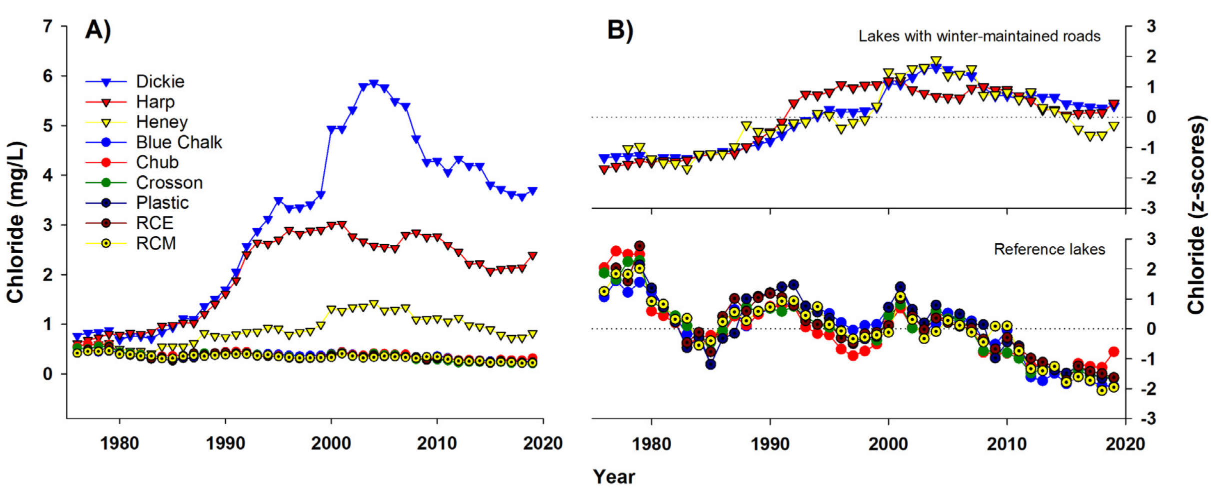

Fig. 5. A) Ice-free mean, volume-weighted Cl^- concentration data for eight Dorset A lakes from 1976 to 2019; **B)** Standardized ice-free mean, volume-weighted Cl^- concentrations for Dorset A lakes with winter-maintained roads vs. remote lakes from 1976 to 2019.

some degree of shoreline development (e.g., road access cottages). Cl^- concentrations in urban lakes near Sudbury were more variable but generally higher than in remote lakes and, in some cases, orders of magnitude higher with a range of 0.5 – 117 mg L^{-1}. The long-term record from Sudbury area lakes indicates that lakes with the highest Cl^- concentrations have also experienced steep increasing trends (Fig. 6B). Mid-summer Cl^- concentrations vary from year-to-year and in the highest concentration lakes (e.g., Silver and Hannah Lake), concentrations have approached the CWQG for the protection of aquatic life against effects of chronic exposure (120 mg L^{-1}).

Lake Simcoe has experienced statistically significant ($p < 0.01$) increasing trends in Cl^- concentration at all sites sampled in the lake. The highest concentrations of Cl^- occurred at station C1 in Cook's Bay (Fig. 7), which is relatively shallow and receives stormwater runoff from nearby developed areas. Samples collected

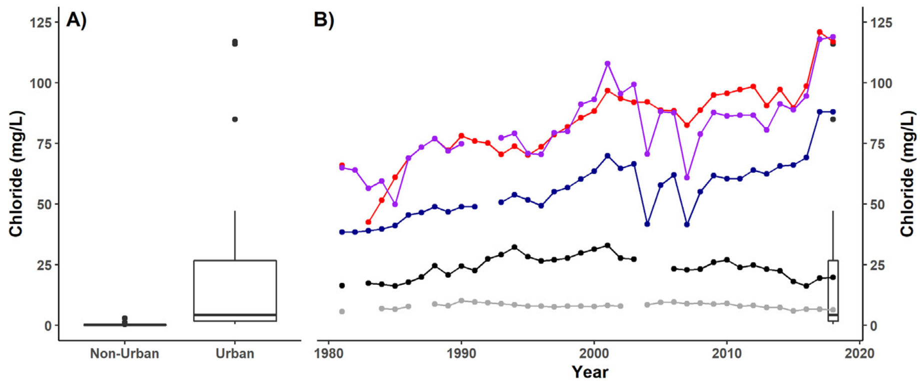

Fig. 6. Mid-summer Sudbury Lake dissolved Cl⁻ concentrations in A) a box plot comparing 66 non-urban and 16 urban lakes included in a broad spatial survey conducted in 2018, and **B)** the same box plot of the urban lakes from 2018 paired with long-term trends for select lakes with Cl⁻ concentrations above the urban lake average, including Silver (purple), Hannah (red), middle (blue), Whitson (black), Clearwater (grey). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

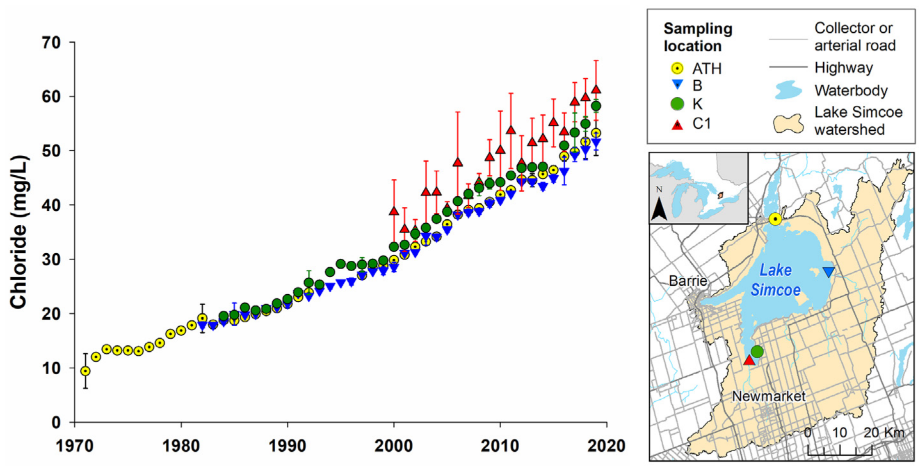

Fig. 7. Annual mean Cl⁻ concentration (May to October inclusive) from 1971 to 2019 in Lake Simcoe at intake pipes for the drinking water treatment systems at Beaverton (B) and Keswick (K), and from lake samples collected in Cook's Bay (C1) and at the lake outlet (ATH), near Atherley Narrows. Bars show ± 1 standard deviation of the annual average. Only years with at least 4 months of monitoring are shown. The map (right) shows these sampling locations in Lake Simcoe and the location of the watershed within Ontario, Canada (inset).

at Atherley Narrows, the Lake Simcoe outlet, since 1971 indicate that Cl⁻ concentration has risen from just above 10 mg L^{-1} to more than 50 mg L^{-1} over the past five decades (Fig. 7).

Laurentian Great lakes

Data from the monitoring at Great Lakes index and reference stations (Fig. 8) and samples of untreated water collected at drinking water facility intakes (Fig. 9) highlight differences among lakes that reflect the anthropogenic nature of Cl⁻ in the Laurentian Great Lakes. Overall concentrations of Cl⁻ at index stations and reference stations were lowest in Lake Superior (mean 2.5 mg L^{-1}; 1.4 to 6.4 mg L^{-1} range) with increases along the downstream hydrologic flow path (Fig. 8). The highest concentrations in Lake Ontario index and reference stations (mean 29 mg L^{-1}; 13 to 133 mg L^{-1} range) were an order of magnitude higher than the Cl⁻ concentrations observed in Lake Superior. Within each lake basin, index stations (i.e., areas known to be influenced by natural or anthropogenic disturbances) generally showed wider variability in Cl⁻ concentrations compared to reference locations (i.e., areas targeted to reflect background conditions). Samples collected at 15 drinking water intakes corroborated the results of the Great Lakes index and reference station monitoring. Chloride concentrations were higher in more developed basins (Erie, Ontario) than in less developed basins (Huron, Superior) (Fig. 9). Mean concentrations over the data record ranged from 3.3 mg L^{-1} in the Lake Superior basin to 22.4 mg L^{-1} in the Lake Ontario basin. Trends at DWSP locations showed a high degree of intra-annual variability, but were based on many fewer samples than GLIP locations (i.e., sampled quarterly versus weekly to bi-weekly, respectively).

Trends in Cl⁻, assessed using annual averages from 16 GLIP locations, were highly variable over time but were similar within basins. An increasing trend was most apparent in the Lake Huron basin, where mean concentrations increased from 7.3 ± 1.8 mg L^{-1} in the 1970s to 14.1 ± 22.7 mg L^{-1} in the 2010s. In contrast, locations in Lake Erie and Lake Ontario show a levelling of increases that occurred prior to 2010. Trends in GLIP data from 2010 to 2019 assessed using Mann-Kendall tests were only significant ($p < 0.05$) at one location in Lake Superior (Terrace Bay) and two locations in Lake Huron (Grand Bend, Amhurstburg).

Monitoring in Lake St. Clair measured similar annual Cl⁻ concentrations in each of the monitoring years from 2016 to 2019 with median concentrations around 10 mg L^{-1} (Fig. 10). These concentrations were similar to those measured in the lake outlet, the Detroit River, over the same monitoring years. However, Cl⁻ concentrations from one of Lake St. Clair's major tributaries in Ontario,

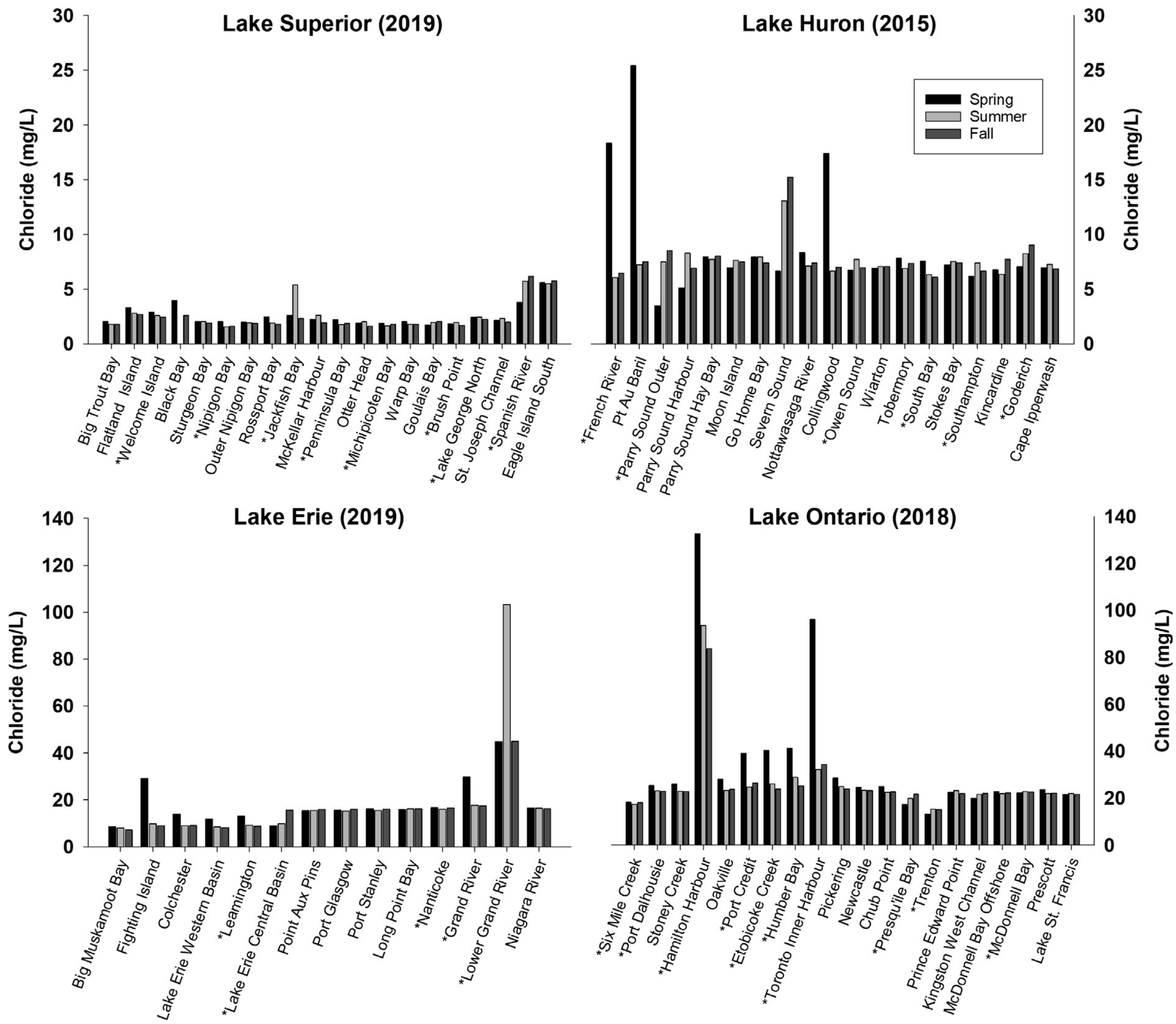

Fig. 8. Great Lakes Nearshore Program index and reference station seasonal mean Cl^- concentrations in the most recent year sampled by the Ontario Ministry of Environment, Conservation and Parks. Please note difference in $[Cl^-]$ range (y-axis) for top and bottom panels. Sites are arranged from west to east and sampling sites located in interconnecting channels are grouped into plots based on sampling year. Index stations are denoted by '*'.

the Thames River, were relatively higher ranging from around 68 – 95 mg L^{-1} from 2016 to 2019.

Groundwater

Fig. 11 presents the recent Cl^- concentrations from 507 PGMN wells. Most data (347 wells) are from the 2019 – 2020 time period, followed by 51 wells sampled during 2017 – 2018 and the remaining 109 wells sampled from 2002 to 2016. Cl^- concentrations ranged from < 0.5 to 7,490 mg L^{-1}, with a median concentration of 20 mg L^{-1}.

Cl^- concentration ranges and trends in PGMN wells with at least 10 Cl^- samples are summarized in Table 3. As a simplified analysis of the data, results are presented for overburden (unconsolidated sediments) or bedrock (consolidated rock) aquifers based on key fundamental differences in physical properties and composition. Approximately 84% of overburden and 75% of bedrock samples had Cl^- concentrations less than the chronic CWQG standard (120 mg L^{-1}). Approximately 3% of overburden and 4% of bedrock samples had Cl^- concentrations higher than the CWQG acute guideline (640 mg L^{-1}).

At least 41% of PGMN overburden wells exhibited unstable Cl^- concentrations, identified as a statistically increasing or decreasing trend or variable concentrations over the period of study. Similarly, at least 51% of PGMN bedrock wells exhibited unstable Cl^- concentrations.

Discussion

The current evidence and state of the knowledge gleaned from MECP's monitoring programs that include Cl^- monitoring points to two key findings: Cl^- concentrations in Ontario surface and groundwaters are highest where there are more people, and these concentrations are increasing. This has been previously suggested by Todd and Kaltenecker (2012), who found that warm season median Cl^- concentrations in some southern Ontario PWQMN streams were positively and significantly correlated to human population (Spearman's $\rho = 0.78$) and road density (Spearman's $\rho = 0.83$). They also reported that concentrations in these streams have been increasing since the 1970s. The spatial and temporal extension of these findings across Ontario show that this pattern holds true in the Great Lakes, inland lakes, streams, rivers and groundwaters of the province.

Streams and rivers are the artery networks of watersheds. They are primary transport vectors for water and materials flowing

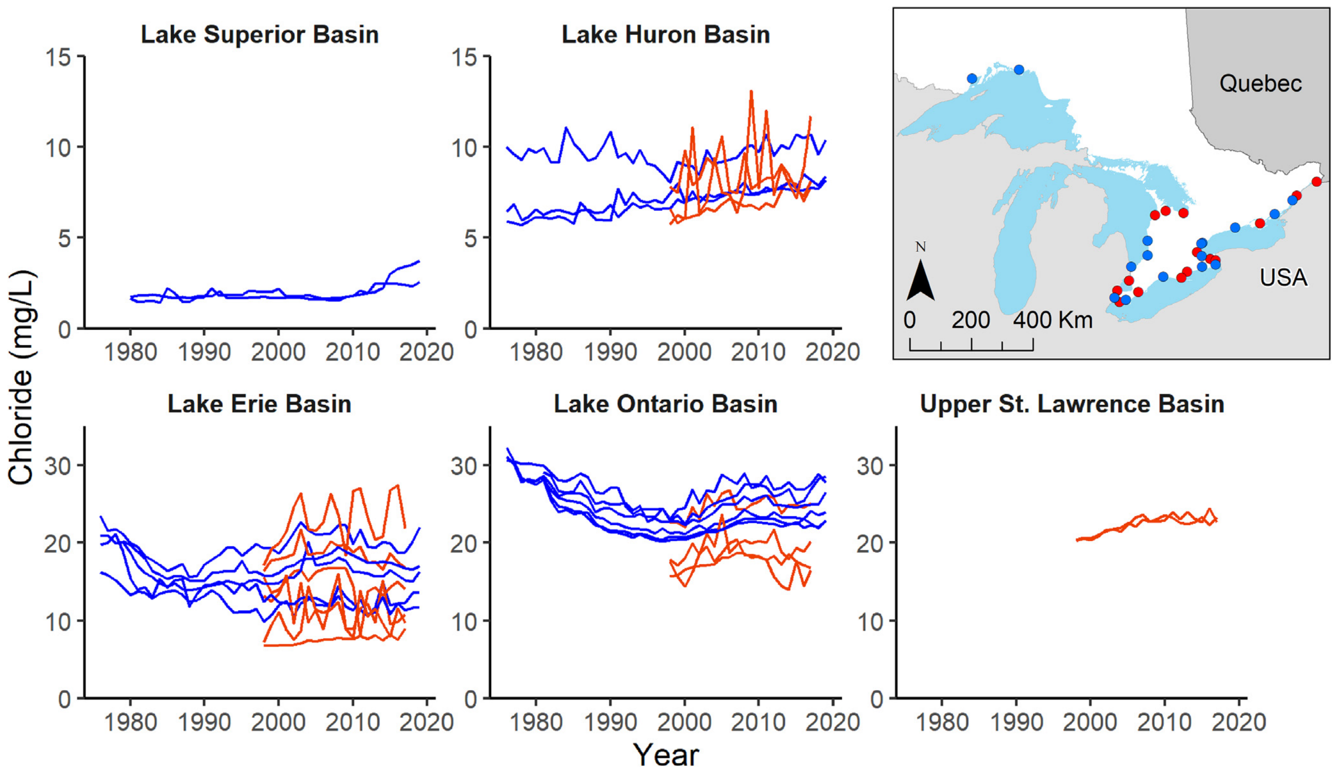

Fig. 9. Long-term changes in annual mean Cl⁻ concentrations from the untreated water collected from drinking water facility intake pipes located in the Great Lakes (including embayments and connecting channels) for all years with data, up to 2019. Locations of these facilities with long-term (greater than 20 years) data are shown on the inset map of Ontario, Canada. Program sampling frequency is indicted by colour: blue (GLIP, weekly-biweekly) and red (DWSP, quarterly). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

directly off landscapes and from groundwater to receiving lakes. This direct receipt of materials from landscapes and groundwater, coupled with comparatively lower water volumes than lakes, results in streams and rivers having higher measurable nutrient concentrations that enable a more direct opportunity to explore impacts of land-based activities on surface water quality (Wetzel, 2001). Streams historically monitored under the PWQMN categorized into agricultural, mixed/treed and urban land use clearly demonstrate the influence of land management on stream Cl⁻ concentration. Cl⁻ in streams dominated by urban land use show increased concentrations since the 1960s to concentrations well above 120 mg L^{-1}, the chronic CWQG standard (Fig. 2), which is consistent with Todd and Kaltenecker (2012). Streams in agricultural and mixed/treed land use also show increases over the data record, but concentrations have remained well below the chronic CWQG standard over the past six decades (Table 2). For the urban streams, that were primarily located in southern Ontario, the sharpest increase in Cl⁻ concentrations occurred in the 1990s, possibly corresponding to accelerated urban expansion and population growth in the lower Great Lakes basin, as similarly shown by Lawson and Jackson (2021). This pattern of increased Cl⁻ in areas impacted by human activity is also supported by data from intensive monitoring in the remote setting of the Muskoka River. Cl⁻ concentrations increased along a gradient of human impact from the upper to the lower reaches of the Muskoka River (Fig. 3). Although concentrations increased by more than 65% over the past 35 years, they remained < 10 mg L^{-1}, well below the chronic CWQG standard of 120 mg L^{-1}.

Inland lakes represent important sites of nutrient retention and transformation as water moves along the hydrologic flow pathway from headwaters to the Great Lakes or oceans downstream (Bouwman et al., 2013). Inland lakes provide an opportunity to more directly explore the effects of land-based activities on surface water quality, due to their close association with surrounding landscapes, smaller contributing watersheds and well-defined land uses. For example, some of the small inland lakes monitored in the Dorset and Sudbury areas are relatively remote, allowing simple categorization of lakes as unimpacted or impacted by human activity through the presence of winter-maintained roads or urban areas (Figs. 5, 6). The evidence of anthropogenic impact on Cl⁻ is clear: relatively higher and increasing historical Cl⁻ concentrations are measured in lakes that have winter-maintained roads or urban land use within their watersheds. Furthermore, Cl⁻ mass balance analyses of four of the Dorset A Lakes (two impacted vs. two unimpacted) found that the impacted lakes had increased Cl⁻ stream loads, lake export and lake concentrations despite long-term declines in atmospheric depositional inputs, whereas the unimpacted lakes showed declines in all of these Cl⁻ metrics over time (Yao et al., 2020).

On a large spatial scale spanning all regions of the province, a similar pattern in Cl⁻ concentrations in lakes emerged, despite the varied geography, geology and mixed land use across the province. The expansive LPP and BSM datasets show that inland lakes proximal to urban centres, major roadways and populated regions have relatively higher Cl⁻ concentrations (Fig. 4) compared to more remote areas. Similarly, Lake Simcoe (Fig. 7) and Lake St. Clair and its tributaries (Fig. 10) receive water from highly populated watersheds and show relatively high and increasing Cl⁻ concentrations. A pattern of increasing Cl⁻ concentrations has previously been observed in Lake Simcoe watersheds (Oswald et al., 2019). Concentrations in Cook's Bay, at the southern end of Lake Simcoe that receives stormwater input from cities of Newmarket and Aurora, are as high as ~ 60 mg L^{-1}, half the chronic CWQG standard (120 mg L^{-1}). The historical rate of increase in Lake Simcoe

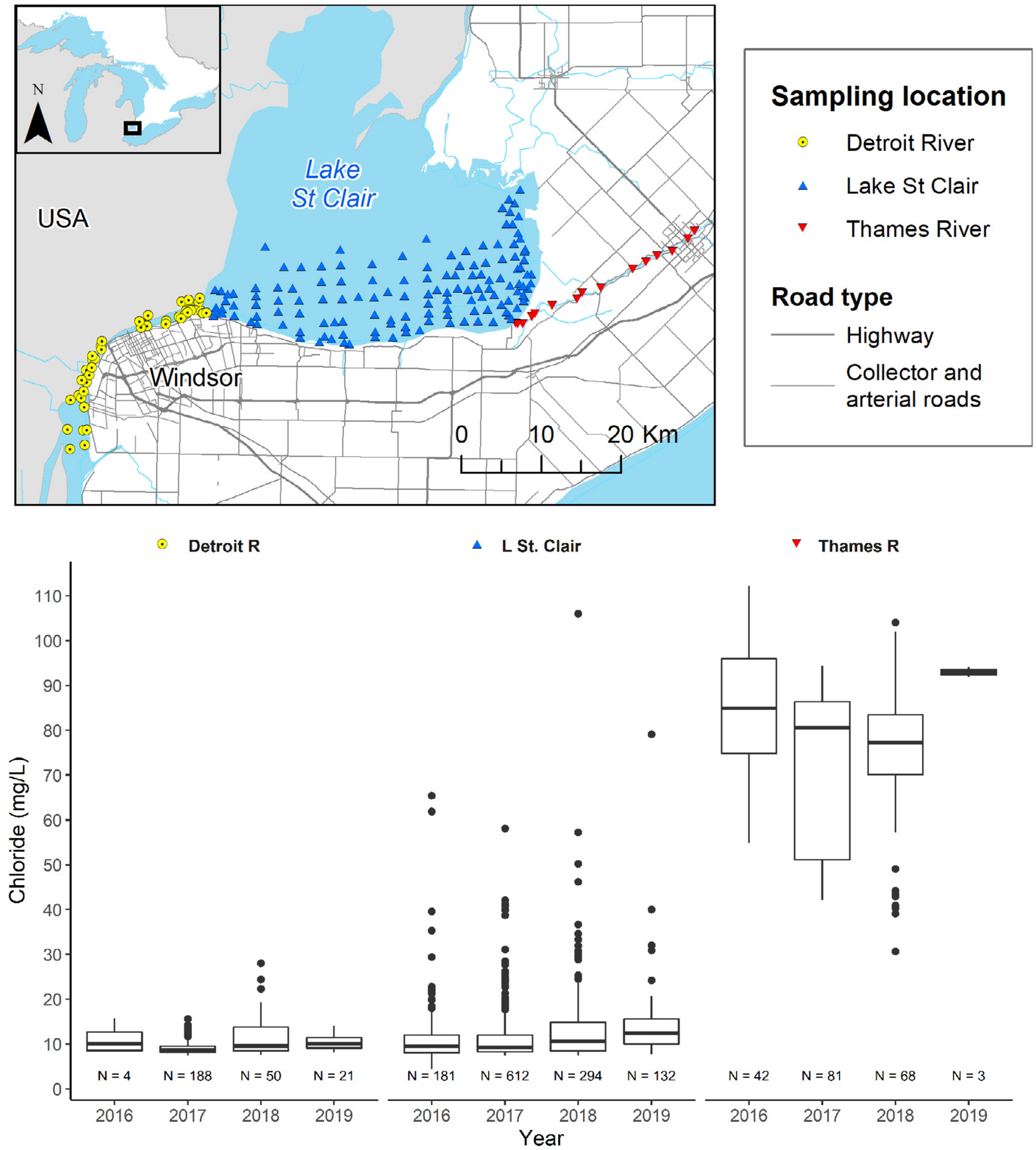

Fig. 10. Cl^- concentrations in Lake St. Clair at locations shown in the map (top) including one of its major tributaries (the Thames River) and outlet (the Detroit River) measured during a study from 2016 to 2019. The inset map shows the study location within Ontario, Canada.

is of concern, as Cl^- concentrations are expected to meet the chronic CWQG standard over the next few decades if the current trajectory continues (Winter et al., 2011; MECP, 2020). Some of the other locations monitored by MECP downstream from urbanized areas (e.g., Lake St. Clair, Toronto and Hamilton harbours, some Sudbury area lakes and the Thames and Grand rivers) are already at concentrations approaching the chronic CWQG standard.

The Great Lakes have been sentinels of change when considering the human population distribution among each of the monitored basins for Lake Superior (673 thousand total; 229 thousand in Ontario (NOAA, 2000)), Lake Huron (3 million total; 1.4 million in Ontario (ECCC & USEPA, 2018a)), Lake Erie (13 million total; 2.7 million in Ontario (ECCC & USEPA, 2019)) and Lake Ontario (11 million total; 9 million in Ontario (ECCC & USEPA, 2018b)). The trend of increasing human population downstream along the hydrologic flow pathway, from the headwaters of Lake Superior through Lakes Huron, St. Clair and Erie to Lake Ontario, follow the same trend of increasing Cl^- concentrations among these lakes (Figs. 8, 9, 10). Monitoring from MECP programs consistently show low Cl^- concentrations in the Great Lakes that have the least-populated watersheds and higher concentrations downstream from large urban watersheds draining cities such as Toronto and Hamilton. For example, Index monitoring in the mouth of the Grand River measured over 2 × the Cl^- concentration of any other sampling station in the lake (40 mg L^{-1}), and summer season concentrations were over 5 × higher (~100 mg L^{-1}) in 2019 (Fig. 8). In Lake Ontario, Cl^- in Hamilton and Toronto harbours was 4 – 5 × higher than any other Index or Reference sampling station in 2018 (80 – 140 mg L^{-1}) and 3 × higher than background levels typically reported in the lake (Howell and Benoit, 2021), specifically in the spring when hydrologic connectivity between landscapes and lakes are made strongest through vernal snowmelt and precipitation events.

There exists inter-lake variability in the time series trends for each of the Great Lake's water intake stations dating back to the 1980s, where the lower Great Lakes show an initial decline in Cl^- concentrations with an inflection point of increasing concentra-

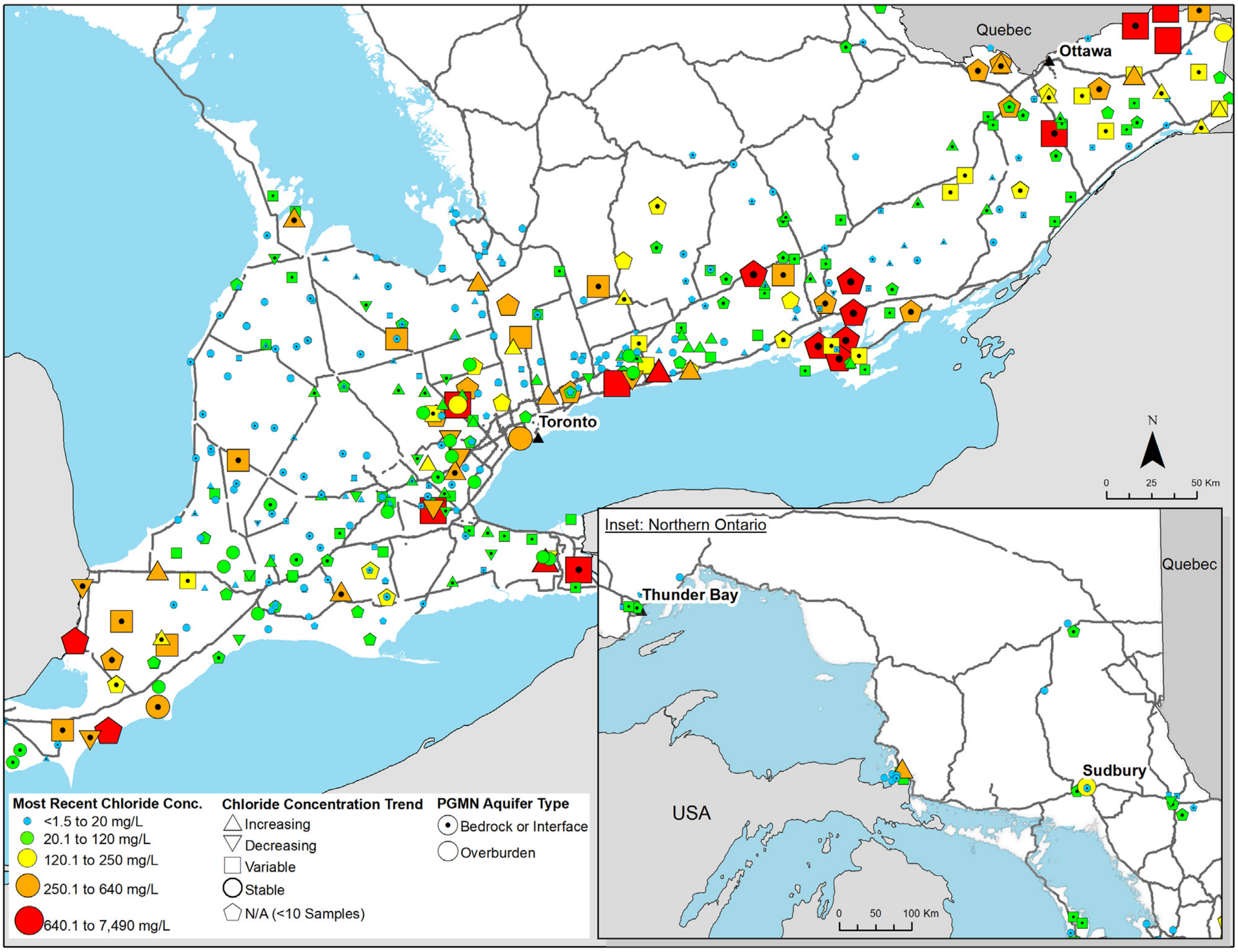

Fig. 11. Map depicting locations of 507 Provincial Groundwater Monitoring Network (PGMN) groundwater wells in Ontario, Canada. Symbols indicate trends in groundwater (overburden and bedrock) Cl^- concentrations (2002 – 2020) and most recently measured concentrations at each well; 2019 – 2020 (n = 347), 2010 – 2018 (n = 107); 2002 – 2009 (n = 53). Trend analysis consisted of visual interpretation and Mann-Kendall analysis for wells with greater than 10 measurements.

Table 3
Latest chloride concentration (2002 – 2020) and trend by aquifer type at 507 Provincial Groundwater Monitoring Network wells.

	Overburden (# of wells)	Overburden (%)	Bedrock (# of wells)	Bedrock (%)
Concentration (mg L^{-1})				
<0.5 to 20[1]	159	57	99	43
20.1 to 120[2]	74	27	74	32
120.1 to 250[3]	20	7	23	10
250.1 to 640[4]	18	6	22	10
640.1 to 7490[5]	8	3	10	2
Chloride Trend				
Unstable[6]	113	41	116	51
Increasing Trend	57	20.4	39	17.1
Decreasing Trend	15	5.4	12	5.3
Variable	41	14.7	65	28.5
Stable	78	28	55	24
N/A (<10 samples)	88	31	57	25

Notes: [1]50th percentile of all wells; [2]Canadian Council of Ministers of the Environment Canadian Water Quality Guideline for Protection of Aquatic Life (CCME CWQG) – chronic exposure; [3] Ontario Drinking Water Quality Standards Aesthetic Objective; [4]CCME CWQG – acute exposure; [5]Maximum concentration; [6]Unstable refers to trends exhibiting increasing, decreasing, or variable chloride concentrations.

tions in many locations starting in the mid 1990s (Fig. 9). This pattern of initial decline followed by increases in Lakes Erie and Ontario have been noted previously (Chapra et al. 2009; Winter et al. 2012) but a trend in Cl^- in these lakes is not apparent in monitoring of nearshore areas over the last decade. In contrast, significant increasing trends are now apparent at locations in Lakes Superior and Huron that have previously shown relatively less evidence of anthropogenic impact. While Cl^- concentrations recorded from the Great Lakes were well below guidelines to protect aquatic life, the increasing trends and trajectories observed in sampling from intake pipes of the drinking water systems are concerning because of the costs associated with the removal of, and potential damages caused by, Cl^- in municipal water treatment and drinking water facility infrastructure (Pieper et al., 2018).

In the most recent year of sampling, concentrations at some nearshore stations in Lakes Erie and Ontario and Lake St. Clair approached or exceed the chronic CWQG (120 mg L^{-1}) standard. This may be further exacerbated with human population increases. Southern Ontario is home to the largest and fastest growing urban population in Canada (Statistics Canada, 2017; Ontario Ministry of Finance, 2020). The population of Ontario is projected to grow by approximately one third over the next three decades (Ontario Ministry of Finance, 2020), with most growth occurring in the southern Ontario region. With the expansion of paved surfaces and road networks to support growing communities, winter road salting activities can also be expected to increase to keep pace with road safety measures and thereby introduce new avenues of Cl^{-} exposure to surface and groundwaters.

Amidst these spatial patterns in human population and Cl^{-} concentrations in lakes and tributaries, the basis for Cl^{-} supply to surface waters from groundwaters must be considered. Anthropogenic sources of Cl^{-}, including road salt, septic system effluent and agricultural activities, can also influence Cl^{-} concentrations in groundwater (Hem, 1959). The degree of hydrological connectivity between groundwater and surface water plays a key role in how effectively an anthropogenic surface source of Cl^{-} can influence concentrations in groundwater. The hydraulic properties of the geological materials overlying an aquifer of interest will also play a role in determining whether the aquifer is confined, unconfined or semi-confined and its vulnerability to anthropogenic activities. Cl^{-} concentrations in groundwater can also be natural in source, resulting from the dissolution of the minerals that comprise the aquifer matrix (Freeze and Cherry 1979). The composition of the aquifer matrix and the residence time of the groundwater in the pores or fractures of the aquifer will influence the natural composition of the groundwater, including the Cl^{-} concentration. How quickly groundwater moves through an aquifer is determined by physical properties of the aquifer.

There are a few areas of elevated groundwater Cl^{-} identified in the PGMN dataset (Fig. 11). To date, terminal receptors of the groundwater (e.g., surface water) have not been evaluated. PGMN Cl^{-} concentrations are elevated in areas of historical oil and gas production (e.g., Sarnia-Windsor corridor, Niagara area and Prince Edward County), which could be the result of improper handling of waste brines in early years (e.g., saline water being discharged to ground, surface or injected into shallow aquifers). Elevated Cl^{-} is also observed in the eastern Ontario region, where remnant waters of the ancient Champlain Sea have been trapped since the last ice age. These areas, and other heavily urbanized regions, may also be influenced by road salting. To date, none of the PGMN Cl^{-} concentrations have been explicitly linked to any of the previously mentioned activities, however studies to determine the sources of Cl^{-} in PGMN are currently in the developmental stage. There are many other factors to consider, including but not limited to location along flowpaths (e.g., recharge or discharge area, unconfined or confined aquifer, depth of aquifer, or hydraulic connection to the surface) and secondary porosity (e.g., bedrock fractures, root channels, etc.) that will contribute to the chemical evolution of groundwater in the multitude of aquifers present in Ontario.

It is anticipated that PGMN wells exhibiting an unstable Cl^{-} concentration could potentially be impacted by an anthropogenic source or other geochemical processes. The variability identified in overburden and bedrock wells indicates that groundwater monitored under the PGMN in Ontario is part of a highly dynamic system. While more than 75% of PGMN data had Cl^{-} concentrations less than the chronic exposure CWQG (120 mg L^{-1}), nearly half of the monitoring stations exhibited unstable Cl^{-} concentrations over the data record (Table 2). These sites with unstable concentrations, and the 3 – 4% of sites that exceeded the acute CWQG, are of particular concern and will be of focus within future PGMN monitoring studies to better understand the geogenic and anthropogenic drivers of groundwater chemical evolution, and the potential impacts those drivers may have on receptors such as surface waters.

In over six decades of MECP Cl^{-} monitoring, the current evidence from more than 2,500 surface and groundwater monitoring stations across the province of Ontario is clear: Cl^{-} concentrations are highest and are increasing in urbanized and populated areas and those that are proximal to roadways. Most monitoring stations have reported Cl^{-} concentrations that are well below the CWQG for the protection of aquatic life from chronic exposure to Cl^{-} (120 mg L^{-1}). However, recent studies on specific freshwater aquatic invertebrates show the current CWQGs may not be protective of all taxa, namely cladoceran zooplankton, particularly in low nutrient and soft-water lakes such as those on the Canadian Shield of which Ontario has many (Arnott et al., 2020; Valleau et al. 2020). Despite satisfying current CWQGs, projected Cl^{-} trends and trajectories must be considered, as even the most remote lakes and rivers in the province with present day Cl^{-} concentrations < 10 mg L^{-1} have exhibited strong increasing trends over the past decades. Areas with Cl^{-} concentrations and trends approaching the chronic CWQG must be considered in future monitoring studies and mitigation efforts to preserve aquatic ecosystem health in these waterways.

Actions to track and address anthropogenic sources are underway in some areas. For example, through the Lake Simcoe Protection Plan, the MECP has been funding the Lake Simcoe Region Conservation Authority to research and promote the adoption of best practices for winter salt application to reduce sources of Cl^{-} into Lake Simcoe and its tributaries (MECP, 2020). Some nearshore areas of the lower Great Lakes, urban streams and groundwater stations have met or exceeded chronic and/or acute CWQGs. These areas are most deserving of provisional efforts to curb the harmful effects of Cl^{-} on aquatic organisms and their habitats. Identification of priority areas can be made possible through extensive spatial and temporal monitoring data and continual assessment to remain at the forefront of the current state of the science through MECP monitoring.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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