Development of Statistical and Mass Balance Approaches for Assessing and Predicting Chloride Concentrations in Halifax Lakes

By

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ABSTRACT

Lake chloride concentrations ([Cl^-]) in Halifax and other urban centres where road salt is used is a growing environmental issue. This thesis used Cl^- data available seasonally from 57 Halifax lakes to model lake [Cl^-] using both linear regression methods and a statistically averaged mass balance model. The watershed variables found to be most predictive of mean lake [Cl^-] were the % urban coverage (R^2 = 0.64), road density (m/m^2) (R^2 = 0.55), and stormwater pipe density (m/m^2) (R^2 = 0.51) in a watershed. The combination that best predicted mean lake [Cl^-] was stormwater pipe density, ramp density, and % rock (of the watershed's surficial geology) (R^2 = 0.71). A mass balance modeling approach was developed and used to estimate Cl^- loading rates for four land use categories (rural 1-3 g/m^2/yr, commercial 0-9 g/m^2/yr, residential 97-162 g/m^2/yr, and road 804-964 g/m^2/yr). These findings further support a growing body of literature that associates roads and development with elevated and increasing freshwater [Cl^-].

LIST OF ABBREVIATIONS AND SYMBOLS USED

[ ]	Concentration
Δ	Delta or change in
Cl^-	Chloride ion
$AlCl^2$	Aluminum chloride
BMP	Best management practices
Ca^{2+}	Calcium ion
$CaCl^2$	Calcium chloride
CART	Classification and regression tree
CCME	Canadian Council of Ministers of the Environment
CSRS	Canadian spatial reference system
DEM	Digital elevation model
EC	Environment Canada
ECCC	Environment and Climate Change Canada
$FeCl^3$	Iron chloride
HC	Health Canada
HCl	Hydrochloric acid
HRM	Halifax Regional Municipality
KCl	Potassium chloride
kg-salt/2-lane km	Kilograms of salt per kilometre of two-lane road
kg-salt/lane km	Kilograms of salt per lane per kilometre
kt	Kilotonnes
LC50	Lethal concentration killing half a test population
LiDAR	Light detection and ranging
Mg	Megagram (equivalent to metric tonne)
Mg^{2+}	Magnesium ion
$MgCl^2$	Magnesium chloride
N	Nitrogen
n	Sample size
n/km^2	Population per square kilometre
Na^+	Sodium ion
NaCl	Sodium chloride
NAD	North American datum
NO_3^-	Nitrate ion

OLS	Ordinary least squares
ON	Ontario
p	Probability under the assumption of no effect
pH	Potential hydrogen
ppm	Parts per million
Q3	Third quartile
Q4	Fourth quartile
QRF	Quantile Regression Forest
r	Correlation coefficient
R^2	Coefficient of determination
R^2-adj	Adjusted coefficient of determination
R^2-pred	Predictive coefficient of determination
RMSE	Root mean squared error
saltable area/m^2 district area	Saltable area per metre squared of district area
SC	Specific conductance
SRP	Soluble reactive phosphorus
t	ton
Tg	Teragram
Tg/yr	Teragrams per year
U.S.	United States
USGS	United States Geological Survey
UTM	Universal transverse mercator
VIF	Variance inflation factor
WWTP	Wastewater treatment plant

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CHAPTER 1 Introduction

Elevated freshwater chloride concentrations ([Cl^-]) have emerged as a major anthropogenic environmental stressor. Chloride (Cl^-) can have physical, chemical, and ecological impacts in freshwater lakes and rivers, many of these impacts are currently being studied (Sibert et al., 2015; Haq et al., 2018; Hintz et al., 2022). Increasing [Cl^-] has been observed in many North American lakes, with over 7500 that are estimated to have elevated [Cl^-] (Dugan et al., 2017). These elevated [Cl^-] have been correlated with development and de-icing salt use (Moore, et al., 2020; Yao et al., 2021).

The Halifax Regional Municipality (HRM) has been rapidly urbanizing over the past 40 years, with associated impacts to water quality (Doucet et al., *in review*). Of particular note, a combination of dense road networks and a high frequency of winter weather events leads to a high use of de-icing salts to ensure winter road safety (Environment Canada & Health Canada [EC & HC], 2001). Many of the densely developed areas in HRM (Halifax peninsula, downtown Dartmouth, and area around the Bedford Highway) are coastal and thus drain to marine environments. However, much of the stormwater from suburban and rural areas drains to freshwater systems, and in turn affects a portion of the county's many lakes.

Guideline concentrations for potential parameters of concern in the environment are set nationally by the Canadian Council of Ministers of the Environment (CCME) and receptor are based (aquatic life, human, wildlife, livestock etc.). Canadian freshwater aquatic life guidelines use a species sensitivity distribution method to assess the risk that contaminant concentrations pose to ecosystems. For chronic exposure to Cl^- this guideline value is set at 120 mg/L in order to protect 99% of species over indefinite exposure; the acute guideline, which is set to protect ecosystem function over transient exposures is 640 mg/L (Canadian Council of Ministers of the Environment [CCME], 2011). These guidelines are some of the lowest globally. However, emerging evidence suggests that these guidelines may not be stringent enough to protect ecosystem function

(Hérbert, 2022). The suggestion of significant impacts under chronic guideline levels is concerning as several urban lakes in HRM have regularly exceeded this value over the past three decades (Scott et al., 2019).

Road salt was deemed a toxic substance by Environment Canada and Health Canada in an assessment under the Canadian Environmental Protection Act of 1999, due to the deleterious effects it has at contaminant concentrations that regularly occur during its storage and use (2001). In response, The Road Salt Code of Practice was developed to encourage the use of best management practices (BMPs), the creation of salt management plans, and the identification of salt vulnerable areas to combat these effects (Environment and Climate Change Canada [ECCC], 2004). The latest program report indicated good uptake of BMPs, but poor fulfillment of the identification of salt vulnerable areas (ECCC, 2021). Additionally, BMPs and considerations for salt vulnerability have not been updated since the release of the original code, though much research has occurred in these areas over this time (Betts et al., 2014; Betts et al., 2015; Claros et al., 2021).

Synoptic decadal surveys of HRM lakes dating back to 1980 have observed elevated and increasing [Cl^-] (Clement & Gordon, 2019). Though the Road Salt Code of Practice has been active for nearly 20 years, many HRM lakes still experience spring exceedances of the chronic guideline level, and some of these lakes have experienced significant increases in spring [Cl^-] since 2011 (Doucet et al., *in review*). This pattern suggests that the mitigation methods recommended, or the levels of uptake seen, are not adequate to protect aquatic habitats.

Land use planning has been identified as one of the most effective ways of protecting freshwaters from high [Cl^-] without compromising road safety (Blaszczak et al., 2019; Dugan et al., 2020). Canadian studies that consider various degrees of watershed urbanization have found a development threshold at 25% of watershed area, over which [Cl^-] are likely to exceed current chronic guideline levels (Winter et al., 2011; Scott et al. 2019). Additionally, different types of development have been found to have varying de-icing salt loading rates or influences on lake [Cl^-] (Dugan et al., 2020; Lam et al., 2020). Thus, explicitly incorporating Cl^- loading potential and resulting lake impacts into urban land use planning will be critical to mitigating this environmental threat. However, land use planners currently do not have access to tools that could be used to assess how proposed development will change lake [Cl^-].

The objectives of this thesis are to address this gap and to develop information needed to conduct lake [Cl-] vulnerability assessments in the Halifax Regional Municipality. The specific objectives are: (i) to quantify relationships between lake [Cl^-] and lake and watershed characteristics to evaluate whether influential factors identified in other regions are relevant in HRM, and (ii) to produce a predictive Cl^- loading model based on land zoning categories that may be used to assess the potential impacts of development on lakes during urban planning processes.

CHAPTER 2 Literature Review

2.1 Properties of Chloride

Chloride is the anion of the element chlorine. Chlorine is a non-metal/halogen with seven valence electrons in the outermost orbital; due to this incomplete outer orbital, it is highly reactive and readily forms salts with metals to gain the final valance electron. Common Cl^- salts include $NaCl$, KCl, $MgCl_2$, $CaCl_2$, $AlCl_2$, and $FeCl_3$. These salts are highly soluble in water and remain in ionic form once dissociated. The Cl^- ion is non-reactive and highly mobile in water (CCME, 2011).

While Cl^- is used by organisms for osmotic regulation, it is lost as easily as it is taken up and its concentration is not generally affected by reactions, making it a conservative ion. It does not biodegrade, precipitate, volatilize, or adsorb readily to mineral surfaces or particulate matter (Wetzel, 2001; CCME, 2011). Because of its persistence in the environment, Cl^- is often used as a tracer in hydrological studies (Kaufman & Orlob, 1956; Wetzel, 2001). In the absence of further loading, Cl^- concentrations in aquatic systems are affected only by dilution from the addition of fresher water inputs, or concentration, as water evaporates (EC & HC, 2001).

2.2 Impacts to Freshwater Systems

The impacts of Cl^- to freshwater systems are numerous and interconnected (Figure 2.1). While some pf these impacts have been well documented and studied for 50+ years, evidence for others continues to emerge. The following sections discuss the physical, chemical, and ecological impacts that elevated Cl^- can have on freshwater systems, in particular, lake systems.

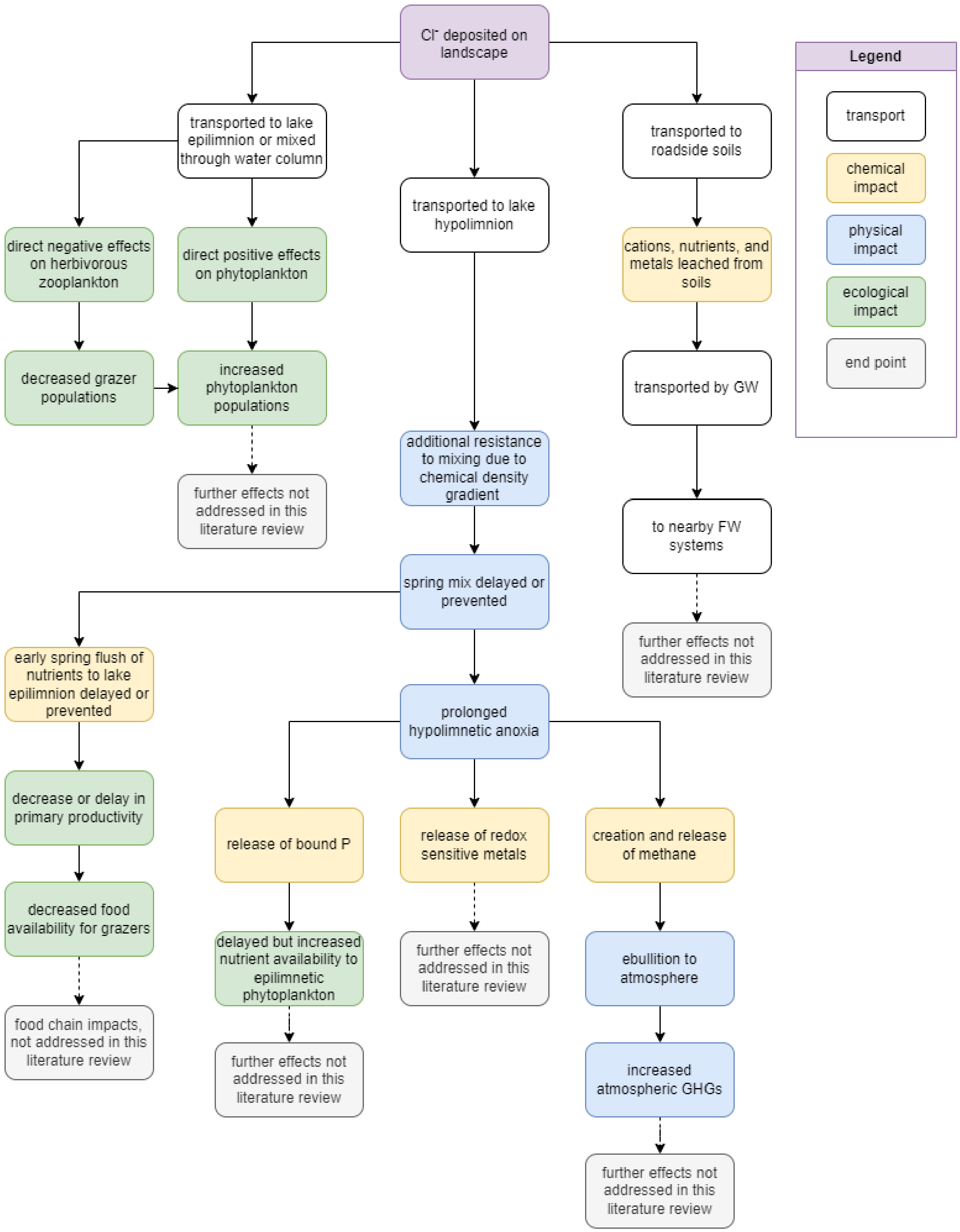


Figure 2.1 Conceptual model of potential Cl^- impacts to fresh surface water systems.

2.2.1 Physical Impacts

Large Cl^- gradients can cause changes in mixing regimes of dimictic lakes, which would normally undergo complete mixing twice a year in the spring and the fall, when the water column becomes isothermal (Wetzel, 2001). Salinity, driven by Cl^-, can create similar or stronger density gradients (haloclines) that are not resolved by temperature convergence (Judd, 1970). Haloclines can create monomictic conditions, where a lake only mixes once a year, or meromictic conditions, where full turnover never or rarely occurs (Wetzel, 2001). Monomixis can occur annually, or only in years with particularly heavy salting seasons, as a fall mix erodes the gradient (Judd, 1970; Novotny et al., 2008). A transition to monomixis can disrupt nutrient cycling and food web dynamics as it prevents the usual spring flush of nutrients to the epilimnion (Yang et al., 2016; Mesman et al., 2021). Meromixis creates two distinct layers of water: a mixolimnion that mixes when isothermal, and a monimolimnion that is perennially isolated at the bottom of the lake (Wetzel, 2001). Transitions to meromixis have been observed and studied in many urban Northeastern North American lakes due to large inputs of Cl^- (Smoll et al., 1983; Judd et al., 2005; Novotny et al., 2008; Sibert et al., 2015; Dupuis et al., 2019). Due to persistent anoxia, the monimolimnions of these lakes exhibit strong redox gradients, which can result in elevated nutrient and methane concentrations (Sibert et al., 2015; Dupuis et al., 2019).

The effect of a halocline is dependent on the magnitude of the [Cl^-] gradient; high surface [Cl^-] does not necessarily indicate altered mixing patterns (Judd & Stegall, 1982) nor are acceptably low [Cl^-] indicative of regular mixing (Smoll et al., 1983). Factors that may make lakes susceptible to changes in mixing regime may include direct, piped, stormwater inputs, high road salt application rates in their watersheds, low fetch or depth to surface area ratios, midwinter thaws, and basins that are sheltered from high winds (Judd, 1970; Judd & Stegall, 1982; Smoll et al., 1983; EC & HC, 2001; Wetzel, 2001; Sibert et al., 2015; Dupuis et al., 2019; Ladwig et al., 2021). However, gradients that do not fully prevent mixing can still prolong stratification by delaying full spring mixing as they contribute to increased water column stability at depth (Ladwig et al., 2021).

2.2.2 Chemical Effects

Though Cl^- is considered conservative and largely inert in aquatic systems, there is evidence that high concentrations of Cl^- salts can alter geochemical processes in soils and sediments, which may ultimately alter the chemistry of freshwater systems. Dissolved Cl^- salts can facilitate mobilization of metals from soils and sediments to pore water or surface water. The mechanisms for this mobilization are competition with cations for soil binding sites, the formation of chloro-metal complexes, and/or enhanced colloidal transport due to increased ionic strength (Warren & Zimmerman, 1994; Bäckström et al., 2004; Norrström, 2005; Mayer et al., 2008; Acosta et al., 2011; Kim & Koretsky, 2013; Galella et al., 2021).

Cadmium in particular has been found to shift towards the dissolved phase in the presence of high [Cl^-] (Warren & Zimmerman, 1994; Bäckström et al., 2004; Norrström, 2005; Mayer et al., 2008; Acosta et al., 2011). This tendency is likely due to its low charge to radius ratio which results in a weak binding affinity (Strumm and Morgan, 1981, as cited in Warren and Zimmerman 1994). Weak binding to particulates makes cadmium more susceptible to forming chloro-complexes in the presence of Cl^- ligands, which readily move to a dissolved state (Warren & Zimmerman, 1994). Cadmium is highly toxic, especially in its free ion form, but dissolved species bound to ligands, like chloro-complexes, can also be harmful to aquatic life (CCME, 1999). However, the fate of heavy metals released from soils subjected to high [Cl^-] and their transport to surface water is still poorly understood (Schuler & Relyea, 2018).

Chloride containing salts have also be found to mobilize cations from soils (Kim & Koretsky, 2013; Snodgrass et al., 2017; Haq et al., 2018; Sutherland et al., 2018), and in some cases, transport them to surface water systems (Shanley, 1994; Green et al., 2009; Sutherland et al., 2018; Galella et al., 2021). Kelting and Laxson (2021), observed significantly higher concentrations of Ca^{2+}, Mg^{2+}, and Na^+ in lakes whose watersheds receive road salt, compared with those that do not. Release of nitrate (NO_3^-), dissolved nitrogen (N), and soluble reactive phosphorus (SRP) from soils and sediments subjected to high [Cl^-] water has been observed in lab experiments (Kim & Koretsky, 2013; Haq et al., 2018). Haq et al. (2018) also noted a correlation between [NO_3^-], specific conductance (SC), and [Cl^-] in a stream.

2.2.3 Ecological Effects

The effects of high Cl^- on individual species/taxonomic groups have been studied and established (CCME, 2011; Tiwari & Rachlin, 2018), but research on ecosystem level effects due to species and trophic level interactions is ongoing. Many studies have found that elevated salinity has detrimental effects on low trophic level freshwater organisms, especially in certain groups of zooplankton (Waterkeyn et al., 2010; Van Meter et al., 2011; Hintz & Relyea, 2017; Castillo et al., 2018; Venâncio et al., 2019; Perron & Pick, 2020; Haake et al., 2022; Hébert et al., 2022; Huang et al., 2022). Decreased abundance at this trophic level can have critical effects as zooplankton control phytoplankton populations by grazing, and are a food source to higher trophic levels (Van Meter et al., 2011; Hintz et al., 2016). The proliferation of phytoplankton following a decrease in zooplankton has been observed in mesocosm studies (Hintz et al., 2016, 2022).

A recent multi-site mesocosm study found significant declines in crustacean zooplankton abundance and diversity with increasing [Cl^-], with no compensatory increases in other zooplankton groups to fill this void left in the primary consumer trophic level, resulting in a lowered overall abundance of zooplankton (Hébert et al., 2022). A parallel study modeled the response of primary productivity (chlorophyll a) to changes in the abundance of Cladoceran groups of zooplankton and found that decreases in this population would lead to increases in phytoplankton abundance as measured by chlorophyll a at 47% of the study sites (Hintz et al., 2022). Importantly, this study did not include higher trophic level predators which have been seen to exacerbate this issue in previous studies (Hintz et al., 2016). Hintz et al. (2022) also found that 3 of 4 major zooplankton groups had site specific LC50s lower than 120 mg/L at >70% of their study sites, indicating that lakes at, near, or over this guideline may have already experienced trophic shifts.

Though increased water hardness and nutrient levels have been reported as lessening the harmful effects of Cl^- (Elphick et al., 2011; Soucek et al., 2011; Brown & Yan, 2015), research by Hintz et al. (2022) only found these factors to be protective of rotifers. Arnott et al. (2020) found that two Cladocera sp. from soft water oligotrophic lakes showed adverse impacts at just 5 mg/L of Cl^-. A follow up study showed that these two Cladocera sp., when collected from oligotrophic soft water lakes and cultured under similar conditions, had LC50s of 24.3 and 60.3 mg/L. These values are much lower than the 621 mg/L used for this group in the development of the CCME guideline (CCME, 2011; Valleau et al., 2022). In summary these recent studies indicate that zooplankton may be more sensitive to Cl^- than previously thought, and that current water quality guidelines may not be protective of ecosystem health.

Increasing [Cl^-] in freshwater systems may also drive biological community structure towards more salt tolerant species and genotypes (Collins & Russell, 2009; Huang et al., 2022), some of which are of particular concern as they are hazardous or create nuisance conditions. Mosquitos have been found to tolerate high [Cl^-] and thrive when small aquatic invertebrates are excluded due to elevated Cl^- (Petranka & Doyle, 2010). Moreover, the toxin producing cyanobacterial genera *Mycrocystis* and *Planktothirx* have both been observed to tolerate very high [Cl^-] with no decrease in toxin production (Tonk et al., 2007; Vergalli et al., 2020).

2.3 Sources of Chloride to Freshwater Systems

2.3.1 Natural Sources

Chloride is naturally present in surface waters, largely due to atmospheric deposition originating from marine salts. Chloride makes up 55% of the salinity of seawater (CCME, 2011); Cl^- and other salts are entrained into the atmosphere by wind and wave action, where they can be transported inland with moving air masses and deposited with precipitation. The amount of Cl^- that is added to terrestrial and freshwater systems by wet deposition decreases rapidly inland (Feth, 1981). The United States National Atmospheric Deposition project has measured wet deposition rates of up to 12.7 kg/ha annually along the coastlines of states in the North Atlantic region, but these quickly decrease to <2 kg/ha away from the coast (United States Geological Survey [USGS], 2020).

This spatially variable source of Cl^- helps explain the wide range of background concentrations reported for regions with large coastlines. British Columbia, for example, reports background concentrations of 1 to 100 mg/L (Nagpal, 2003). Background concentrations in Atlantic Canada have been estimated to be between 1 and 40 mg/L (Mayer et al., 1999), whereas background [Cl^-] in the middle of the continent is usually <1 mg/L (CCME, 2011). Estimates of background concentrations for freshwater lakes in Nova Scotia (NS) vary. Some report 8.1 mg/L province wide (Underwood et al., 1986), others 3.6 to 5.4 mg/L for inland lakes (Freedman et al., 1989), and 15-20 mg/L for unimpacted lakes near the coast (Thirumurthi and Tan, 1978, as cited in EC & HC, 2001). Additionally, in an unpublished report, the Nova Scotia Department of the Environment (1989) indicated that any lake with a [Cl^-] above 25 mg/L is almost certainly receiving anthropogenic loading (as cited in Evans & Frick, 2001).

Chloride aerosols arising from volcanic emissions can also be deposited on land by wet deposition. Hydrochloric acid (HCl) is a common component of volcanic fumarole, gasses regularly emitted from active volcanoes rather than eruptions (Fischer, 2008). When deposited, HCl dissociates completely and can effect both the [Cl^-] and pH of freshwater systems (Evans & Frick, 2001). However, significant amounts of HCl were not found to be associated with volcanic eruptions compared to other acidic aerosols, and Cl^- contributions from fumarole were found to be very small compared to marine sources in coastal regions (Tang et al., 2018).

Another common source of Cl^- to freshwater systems is geologic deposits of marine evaporite (CCME, 2011). These sedimentary deposits consist of salts left behind from evaporated marine or otherwise saline water, most often in the form of crystalized NaCl or concentrated brine solutions (Hem, 1985). Where they exist, marine evaporites can be the dominant source of Cl^- to freshwater or inland saline water bodies, contributing

Cl^- when runoff flows over exposed deposits, or through groundwater that intercepts buried deposits before discharging to surface water (Albek, 1999).

Rock weathering can also be a source of Cl^- to surface water, especially in regions underlain by limestones and shale (Hem, 1985). However, Cl^- is the least abundant major ion in rocks (excluding marine evaporite) so weathering contributes relatively little to Cl^- budgets (Granato et al., 2015). Seawater intrusion can affect some coastal lakes through the direct addition of marine water. Mechanisms for this direct addition are: groundwater intrusion, large storm events that produce storm surges that reach nearby lakes, and tidal action in lakes that are connected to the ocean (CCME, 2011; Muchebve et al., 2016). Wildfires have also been found to a natural source of Cl^- to freshwaters. Wildfires can mobilize Cl^- ions that are normally biotically cycled within terrestrial ecosystems to groundwater and surface water systems (Bastviken et al., 2006). Though the exact mechanism for release is not well understood, Granath et al. (2021) observed $[Cl^-]$ 1.5-2.5x larger than background concentrations in streams draining catchments that had been burned.

2.3.2 Anthropogenic Sources

2.3.2.1 Atmospheric Deposition

Chloride reaching surface waters via atmospheric deposition of HCl can also originate from anthropogenic emissions. In some cases, anthropogenic sources can constitute nearly half of the HCl deposited on land through precipitation (Shapiro et al., 2007). Burning and/or incinerating coal, biomass, and refuse releases HCl to the atmosphere which can be deposited on landscapes.

Coal used for power production contains 100-300 ppm Cl^- (Yudovich & Ketris, 2006). During incineration this Cl^- is converted to HCl that can be transported with airmasses and deposited with precipitation. Though it was thought that most HCl produced by coal burning was deposited close to the plant, Evans et al. (2011) showed broad transport and deposition of HCl from coal burning in the United Kingdom. However, the authors point out that desulfurization technologies are highly efficient at removing HCl, leading to greatly diminished deposition. In regions employing these technologies or using other means of power production, Cl^- depositions from fossil fuel burning are relatively small.

Burning biomass, for power generation or otherwise, can also produce HCl aerosols. Biomass is typically <1% Cl^- by dry weight. On a per weight basis biomass incineration produces more HCl than coal, but it is less common and technologies/strategies that reduce HCl emissions from these facilities are being developed (Ren et al., 2017). Similarly, waste incineration can contribute significantly to HCl deposition. Municipal waste usually contains 0.2 to 2.5% Cl^- due to the inclusion of some plastics, like polyvinyl chloride. While much of the HCl produced during incineration can be captured by pollution control systems (99%), in their absence waste incineration facilities with capacities of 1000-3000 tons/day could emit 5000-9000 kg Cl/day (Zhang er al., 2019). Researchers Müller and Gächter (2012) found that waste incineration contributed ~3% of the Cl^- deposited annually in the watershed of Lake Constance, which is bordered by Austria, Germany, and Switzerland.

2.3.2.2 Wastewater

Wastewater can be a significant source of Cl^- to freshwater systems. A recent Cl^- mass balance study of Lake Iseo, Italy, found that wastewater treatment plant (WWTP) effluent contributed 23% of the annual Cl^- load to the lake (Nava et al., 2020). Factors that can contribute to WWTP effluent loading include human excreta, cleaning products, drinking water treatment, and water softener residuals (Overbo et al., 2021). Wastewater loading rates in the range of 10 g/capita/week are commonly used. Even so, human waste has been shown to make up relatively little of the Cl^- load to WWTPs (~4%). Similarly, cleaning products, treated drinking water, and wastewater disinfection have been seen to make small contributions to the Cl^- loads of studied WWTPs (Overbo et al., 2021).

The largest contributor of Cl^- to WWTP effluents is by far water softeners in areas where they are commonly used. Common ion-exchange water softeners include NaCl and KCl, where the cations of these salts displace Ca^{2+} and Mg^{2+} ions in water, but result in high Cl^- brines that are discharged to WWTPs or to septic systems. In Minnesota, Overbo et al. (2021) calculated that about 44% of the total Cl^- discharged from WWTPs in the state (114100 t) was contributed by household water softeners. Areas where water softening is not necessary are expected to have much lower wastewater Cl^- loads.

2.3.2.3 Agriculture

In highly agricultural catchments**,** livestock wastes and fertilizer leaching can contribute significant Cl^- loads to surface waters (Müller & Gächter, 2012; Laceby et al., 2019). Potassium chloride is a widely applied fertilizer. Only a small portion of the applied Cl^- is taken up by crops (10-40 kg/ha), while Cl^- leaching rates of ~100-360 kg/ha have been observed (Stites & Kraft, 2001). Animal wastes can also be a significant source of Cl^-. One livestock unit (equivalent to a dairy cow) is estimated to contribute 42 kg Cl^-/yr (Müller & Gächter, 2012). In a study of the North Saskatchewan River, Laceby et al. (2019) found that agriculture contributed 20% of the Cl^- downstream of Edmonton and that a large majority of this came from animal sources.

2.3.2.4 Other Anthropogenic Sources

Dust suppressants are commonly applied to gravel roads once or twice a summer. These brines are typically 30 to 40% $CaCl_2$ or $MgCl_2$ (Overbo et al., 2021). They usually make up small portions of annual Cl^- budgets due to their infrequent application. Landfill leachates [Cl^-] are highly variable and depend on the age and contents of the landfill. Concentrations ranging from 0.13 to 100 g/L have been reported (Granato et al., 2015). More moderate levels were seen by Kennedy and Lentz (2000) who reported [Cl^-] of 1.5 to 3 g/L in a Canadian landfill . However, in a study of an urbanized river system, Thunvist et al. (2004) found that landfills contributed relatively little of the Cl^- load (<1%). Where industrial sources discharge to freshwater bodies, these sources can dominate Cl^- loading. Industries commonly discharging high Cl^- effluent are salt mining, the production of soda ash, the desalination of crude oil, pulp and paper processing, and textile dyeing (Pfromm, 1997; Sonzogni et al., 1983; Ficker et al., 2011; Lu et al., 2015; Islam & Mostafa, 2020).

2.3.3 Deicing Salts

While other sources contribute to Cl^- loading, there is overwhelming evidence that deicing salts are the dominant source of Cl^- in places that apply them. Deicing salts are applied to roads before and during winter storms to increase road safety. Use of deicing salts increased dramatically in the 1980s and has likely remained high since. Road salt has been deemed a toxic substance and placed on the priority substance list as Cl^- levels that commonly arise from broadly applying deicing salts have harmful biological and ecological effects (EC & HC, 2001).

In a review of ten Cl^- mass balance studies from the relevant literature, deicing salts were found to be the dominant source of Cl^- to the studied watershed in 11 of 12 instances (Table 2.1). In these 11 watersheds, deicing salts contributed 33 to 91% of total Cl^- loading. In addition to Cl^- mass balances, there are many indicators that deicing salt contributes significantly to freshwater salinization in regions where they are used. These indicators include geographic and temporal patterns that can be related to road salt application, and the correlation between [Cl^-] and many characteristics associated with de-icing salt application (proxies).

Table 2.1 Sources of Cl^- as found in reviewed mass balance studies, proportion of total Cl^- load allocated to each shown as percentage.

	Percentage of Total Cl- Load								
System	De-icing Salts	WWTP	Agriculture	Dust Suppressants	Deposition (wet and dry)	Industrial	Other	Other:	Study
North Saskatoon River, Alberta	60.3%	17.6%	22.1%						(Laceby et al., 2019)
Lake Constance, DACH Europe	51.6%	23.1%	11.2%		2.7%	2.8%	8.8%	weathering	(Müller & Gächter, 2012)
Sagån River Basin, Sweden	60.8%	4.4%	14.4%	7.4%	12.5%		0.4%	landfill	(Thunqvist, 2004)
Minnesota (statewide)	42.3%	25.5%	29.8%	1.0%		1.5%			(Overbo et al., 2021)
Illinois River Basin, Illinois	27.9%	45.8%	25.1%		1.2%				(Kelly et al., 2010)
Mississippi River downstream of TCMA	72.2%	26.6%	1.1%		0.1%				(Novotny et al., 2009)
Beaver Brook, New Hampshire	87.9%	10.6%			1.5%				(Trowbridge et al., 2010)
Dinsmore Brook, New Hampshire	96.1%	2.9%			1.0%				(Trowbridge et al., 2010)
Policy Brook, New Hampshire	95.5%	3.8%			0.7%				(Trowbridge et al., 2010)
Policy Brook, New Hampshire	99.6%	0.4%			0.1%				(Harte & Trowbridge, 2010)
Lake Iseo, Italy	32.6%	22.3%	14.6%		20.4%		1.0%	error	(Nava et al., 2020)
Wappinger Creek (E. Branch) New York	90.7%	6.7%			1.6%		1.0%	weathering	(Kelly et al., 2008)

2.3.3.1 Geographic Patterns

De-icing salts are broadly accepted as the primary source of Cl^- to freshwater bodies in regions where they are applied. Moore et al. (2008) conducted a study along the Eastern U.S. and showed an increasing trend in [Cl^-] from the south, where little road salt is used, to the north where large amounts of road salt are used. This trend was replicated in specific conductance (SC), which was also correlated with the amount of impervious area in a watershed. The relationship between SC and impervious cover was increasingly steeper from south to north, and the contributions of Cl^- to SC increased along the same gradient, indicating that the per unit loading of Cl^- salts to impervious surface increases moving north, following a general pattern of increasing winter severity. The study concluded that de-icing salts are the primary driver of SC and [Cl^-] where they are used. In addition, the authors noted that SC and [Cl^-] had greater seasonal variability in more northern sites (Moore et al., 2020).

2.3.3.2 Temporal Patterns

A common pattern of winter/early spring Cl^- peaks, followed by summer/fall minimum concentrations is a key indicator of road salt contamination in lotic or moderately-to-frequently flushing systems. This loading pattern generally follows seasonal patterns of road salt application, though in some cases time lags occur, and has been observed in many studies (Trowbridge et al., 2010; Winter et al., 2011; Kelting et al., 2012; Corsi et al., 2015; Scott et al., 2019; Moore et al., 2020; Dugan et al., 2021). The consistently large differences between the winter/spring peaks and summer/fall minima of [Cl^-] cannot be explained by anything other than the large applications of Cl^- based salts to watersheds during winter weather events (Novotny et al., 2008; Sofijanic et al., 2021).

2.3.3.3 Proxies as Explanatory Variables

Studies of North American lakes have found that proxies for road salt loading are highly correlated with [Cl^-]. Road density, impervious cover, urban and suburban development, climate severity, and the distance to the nearest major highway have all been used to predict [Cl^-] in freshwater bodies (Novotny et al., 2008; Trowbridge et al., 2010; Winter et al., 2011; Kelting et al., 2012; Dugan et al., 2017, 2020, 2021; Scott et al., 2019;

Mazumder et al., 2021; Haake et al., 2022). In a recent study of 2773 lakes in the eastern U.S., the most important predictors of [Cl^-] were found to be low/medium intensity development, crop abundance, winter climate severity, and the distance to the nearest interstate (Dugan et al., 2020). Crop abundance was only found to be a significant predictor of lake [Cl^-] in watersheds with low development coverage. The other three predictive variables are indicative of road salt applications, especially when considered together. Low and medium density development often have high proportions of impervious saltable area. Winter severity can dictate the frequency and magnitude of road salt applications (Moore et al., 2020). Interstates have been associated with high road salt application rates (Kelting et al., 2012).

While urban development is consistently a strong predictor of Cl^- levels, Corsi et al. (2015) found that [Cl^-] are increasing faster than watershed urbanization is occurring. Chloride concentrations increased in streams that had any urban landcover in their watersheds, even if the amount of this landcover had not changed. Two possible reasons for this were hypothesized to be an increase in application rates (per unit of saltable area) over time, and/or that the proportion of impervious, saltable, area per unit of developed area had increased over the time of the study.

2.3.4 Road Salt Sources and Pathways to Freshwater Systems

Chloride from road salt can enter freshwater systems through many pathways, beginning at salt storage facilities. Salt storage facilities have been found to be large sources of Cl^- to both groundwater and surface water (Tharp & Allen, 2020). Groundwater concentrations of up to 2.8 g/L have been measured downgradient of salt yards, where Cl^- can originate from uncovered salt or mixed piles, losses during transport/loading, and runoff from equipment wash water (EC & HC, 2001). Chloride concentrations of 130 to 640 mg/L (well above the aesthetic objective of 250 mg/L) were observed in drinking water wells in Bible Hill NS; the source of the contamination was found to be an upgradient salt storage facility (Ruston, 1999, as cited in EC & HC, 2001). Uncovered piles of salt or abrasives mixed with salt can leach large amounts of Cl^-

even if the Cl^- content is low. Runoff concentrations of up to 66 g/L have been observed from mixed piles, and uncovered piles of just road salt have been seen to lose up to 33% of their mass to leaching over a salting season (NB DOE and DOT, 1978; EC & HC, 2001). A study of a New Hampshire stream found that one salt storage facility contributed over 173 Mg to the stream in one year (Trowbridge et al., 2010).

As de-icing salt is transported from storage facilities and applied to roads by trucks and plows many factors can affect the amount of salt applied and subsequently the [Cl^-] of road runoff. Firstly, the number of lanes in a road directly affects the amount of saltable area and therefore amount of salt required per km (Watson, 2000, as cited in EC & HC, 2001). The road type and average traffic volume can also affect the application rate of salt. Streets that carry more traffic are often salted more frequently and at higher rates than those that service only local traffic. In Montreal, Quebec significantly higher [Cl^-] were found in snow cleared from primary streets than secondary streets, and over 10x higher concentrations were found in snow from commercial streets than residential streets (Delisle and Deriger, 2000, as cited in EC & HC, 2001).

Differences in salt application rates can also arise from differences in the authority responsible for winter road maintenance. Kelting et al. (2012) found that interstates, state roads, and 'US routes', all maintained by the State of New York were significant predictors of lake [Cl^-], but local roads (municipally maintained) explained little of the variation in concentrations. The authors also found that a road's proximity to a lake could affect the lake [Cl^-], likely by influencing the transit time of Cl^- to surface water systems. Roads within 10 m of lakes explained 30% of [Cl^-] variation, a number disproportionate to the relative road density in this proximity, and roads within 320 m of lakes explained 76% of [Cl^-] variation (Kelting et al., 2012). This observation could be due to groundwater infiltration and resulting longer travel times, Cl^- traveling through groundwater can take months or years to reach surface water (Kelly et al., 2008).

Several studies have found discrepancies between the mass of Cl^- applied to a watershed and the amounts exiting or stored in surface waters. The storage of this excess input has been attributed to groundwater and soils (Novotny et al., 2009; Müller & Gächter, 2012; Oswald et al., 2019). Several studies have also found elevated groundwater [Cl^-] near or down gradient from surfaces that receive road salt (Briggins & Cross, 1995; Daley et al., 2009; Harte & Trowbridge, 2010; Burgis et al., 2013), especially when these surfaces are not directly connected to surface waters (Briggins & Cross, 1995; Casey et al., 2013).

In areas with larger proportions of impervious surface coverage, stormwater conveyance systems are required to manage surface runoff that cannot infiltrate. Direct connection of these areas to surface water via stormwater pipes can result in large Cl^- spikes in the receiving systems (Trowbridge et al., 2010; Blaszczak et al., 2019). Common stormwater BMPs, such as detention ponds, have not been found to protect receiving waters from high Cl^-. While they may temporarily act as sinks with regards to surface water Cl^-, they are sources to groundwater, which eventually contribute to surface water baseflow and result in elevated [Cl^-] in surface waters year round (Kincaid & Findlay, 2009; Corsi et al., 2015; Robinson et al., 2017; Soper et al., 2021). Snodgrass et al. (2017) found that streams that had stormwater management ponds in their watersheds had consistently and significantly higher [Cl^-] than streams without ponds but with similar levels of development. A study of two Ontario (ON) stormwater ponds found that these ponds extended periods of elevated [Cl^-] in their receiving streams rather than preventing them, with Cl^- pulses occurring more frequently than salting events and continuing after the salting season was over, due to the storage and release of high [Cl^-] water in the ponds. This study also found differences in commercial and residential salt application rates, as one pond drained a largely residential, municipally maintained area, while the other drained a large commercial parking lot. The authors estimated that salt loading at commercial properties was 3 to 7x higher that of residential areas based on the relative differences in the [Cl^-] of the inflow (Lam et al., 2020).

Commercial and residential road salt loading rates are usually difficult to quantify as they are managed by private companies or individuals with no recording or reporting requirements. They have been estimated to make up 5 to 10% of total Cl^- loading, but proportions up to 91% have been reported (Cheminfo, 1999, as cited in EC & HC, 2001; Trowbridge et al., 2010). A loading rate of 58.1 g/m^2/storm event, found from a voluntary survey, is often cited in the literature (Fu et al., 2013). However, Lembcke et al. (2017) monitored two parking lots in the Greater Toronto Area and found application rates of ~65 to 117 g/m^2/storm event, noting variability in salt application within different areas of the same lot, for the same event. Commercial application rates of road salt can be highly variable depending on location, culture, and contractor. It may not be appropriate to use application rates from one region in another. Additionally, rates based on 'saltable commercial area' usually require the hand delineation of parking areas, which can be impractical in large watersheds (Oswald et al., 2019).

2.4 Road Salt Use in Nova Scotia

Estimates for Cl^- loading rates from road salts in NS are relatively high when considered on a per area basis at ~230 kt annually (national range: 18 kt in PEI to 1148 kt in ON). Nova Scotia appears to have one of the highest provincial road densities at 0.001-0.005 m^2 saltable area/m^2 district area, which is comparable to the road density of highly populated areas of southern ON. Though NS has one of the lowest maximum recommended salt application rates (~125 kg salt/2-lane km), the minimum recommended application rate is on par with other provinces (~80 compared to 50-100 kg/2-lane km). This small range of application recommendations, combined with high road density and frequent winter weather events leads to high annual loading rates. Though the last provincial loading and application rates that could be found were collected in 1998, it is unlikely that salting has significantly decreased in the intervening decades. While road salt has been used in Canada since the 1940s, application and loading rates increased dramatically in the 80s, and remained high until at least 2001 when the Priority Substance List Assessment Report on Road Salts was published (EC & HC, 2001). Roads maintained by the HRM, including many in the dense urban and suburban areas, may have seen increases in salt application rates since the report. The municipality reports a standard rate of 95 kg/lane km, which is double the provincial rate reported in 1998 (HRM, 2017). However, both provincial and municipal road maintenance authorities have adopted the use of salt reduction strategies in recent years which can reduce the loading rate of pre-storm salt applications to ~20 kg/lane km (HRM, 2017; NS TIR, n.d.). There is little information available on what proportion of roads these strategies are used, but the reduction in loadings that they achieve are likely balanced by additional road salt application due to urban and suburban expansion in the last 20 years. A large portion of the salt that is applied in NS is directed towards freshwater systems via stormwater drains and pipes (Delisle and Deriger, 2000, as cited in EC & HC, 2001). Direct connection to impervious area combined with high flushing rates may be why NS lakes have responded so strongly to urbanization, with increases in urban cover, and potentially areal salt application rates, quickly translating to increased lake [Cl^-] (Scott et al., 2019). In their study of nine lakes in HRM, Scott et al. (2019) found that when greater than 25% of the lake watershed was developed the mean annual [Cl^-] exceeded the CCME long-term guideline of 120 mg/L, which agrees with thresholds found in other studies (Winter et al., 2011; Lax et al., 2017; Scott et al., 2019).

2.5 Modeling Chloride

Models that have been developed to understand and predict [Cl^-] in freshwater systems generally fall into two categories: (i) statistical models that use large amounts of data to identify relationships between [Cl^-] and potential explanatory variables, and (ii) physically-based models that involve construction of Cl^- mass balances for individual systems. Both techniques can yield valuable information and can be used to predict future lake [Cl^-] or identify potential Cl^- contributors. The following sections provide an overview of these modeling techniques.

2.5.1 Statistical Models

Univariate and multivariate statistical techniques have been applied to explore drivers of lake [Cl^-] across North America and Western Europe. Typically, the application of these models makes use of large spatial or temporal datasets to determine which combination of lake and/or watershed characteristics best explain the variation in [Cl^-]

in a suite of lakes (Novotny et al., 2008; Winter et al., 2011; Kelting et al., 2012; Scott et al., 2019; Doucet et al., 2022). Using this approach, Kelting et al. (2012) found that road densities, separated into classes (interstate, state road, U.S. route, and local), were able to explain 87% of the Cl^- variation in the lakes of Adirondack Park, NY (n=82). In Halifax, NS, Scott et al. (2019) found that urban development (as percent watershed coverage) was the most significant explanatory variable for lake [Cl^-] in a group of urban lakes. Alternatively, these analyses have been performed using more complex algorithms for classification and regression.

Dugan et al. (2017) demonstrated these techniques, grouping 56 lakes into three Cl^- trend classes using hierarchical clustering followed by the application of supervised learning methods to predict the class or a numeric response variable using landscape characteristics. The authors applied both classification and regression tree algorithms (CART) and quantile regression forests (QRF). The use of CART involves the construction of a decision tree that determines the most efficient way to split datasets into groups based on the predictor variables, and QRF iterate this process many times and makes decisions based on the medians of the resulting groups (rather than the means). Predictor variables included road density and impervious surface cover within 100 to 1500 m buffers from the lake, average monthly temperatures, precipitation, combined wet and dry deposition of Cl^-, lake surface area, depth, and type, and ecoregion. The authors found that impervious land cover and road density near a lake were the most important predictor variables, with impervious cover within 100, 500, and 200 m, and road density within 500 m of a lake acting as primary splits in resulting CART and QRF models. Dugan et al. (2020) used a similar technique (QRF) with data from 2773 lakes in the Midwest and Northeast U.S. Their model included 22 predictor variables and showed low and medium density development (as % of watershed coverage) as being the most consistently important predictor variables. The authors noted that lake morphometry variables conventionally thought to be protective did not exhibit a strong influence on lake [Cl^-].

2.5.2 Physically-Based Models

Physically-based models can be used to estimate loading to a lake from different sources (Müller & Gächter, 2012; Nava et al., 2020), and/or to estimate future lake [Cl^-] (Chapra et al., 2009; Novotny & Stefan, 2010; Dugan et al., 2021). While modeling Cl^- is relatively simple because of its conservative nature, it usually requires several years of frequent grab sampling or high frequency conductivity data (calibrated to [Cl^-] in a particular system or region) (Chapra et al., 2009; Novotny & Stefan, 2010; Winter et al., 2011; Nava et al., 2020). Physically-based Cl^- models follow the general mass balance principle of 'In - Out = Accumulation', which can become complicated for lakes with long residence times and nested watersheds (Chapra et al., 2009). To avoid complications arising from stratification, models usually assume complete mixing, or predict volume weighted [Cl^-].

2.5.2.1 Estimating Loading From Concentration

Physical models can be used to calculate net loading from time series lake [Cl^-] data, as was done by Chapra et al. (2009) to reconstruct a 150-year history of Cl^- loading to each of the Great Lakes, and Novotny and Stefan (2010) to determine average seasonal loading to seven lakes in urban Minnesota. In their study of the Great Lakes, Chapra et al. (2009) created mass balance equations for each lake using known historical flows and the change in concentration over each time step. The resulting system of equations was used to solve for loading given the [Cl^-] of each lake which they termed 'inverse loading analysis'. Using this method, the authors were able to distinguish between the direct loading to each lake and Cl^- imported from the upstream lake, concluding that Cl^- loading to most of the lakes had increased since the post industrialization minima of the 1970s, and that the loading rate of Lake Michigan had surpassed that of peak industrial loading.

In a study of urban lakes in Minnesota, Novotny and Stefan (2010) estimated seasonal loading to seven independent lakes using monthly [Cl^-] (volume weighted, five years of data) and known lake volumes. Four remaining parameters (loading rate, hydraulic residence time, and the dates of minimum and maximum [Cl^-]) were estimated during model calibration which aimed to minimize the RMSE between modeled and measured [Cl^-]. By separating the models into loading and flushing phases, both the peak and annual mean [Cl^-] could be predicted for each lake. While this technique is useful for determining total loading rate to each lake, it cannot be used to partition loading into sources (natural, industrial, road salt, WWTP). Novotny and Stefan (2010) avoided this by assuming all loading was in the form of de-icing salts and estimated aerial loading rates by dividing the annual load to each lake by the area of impervious surface in its watershed; however this assumes that all impervious area is salted at the same rate.

2.5.2.2 Source Specific Mass Balance Models

When using a source-based approach, loading rates are calculated for each potential source within a watershed, and estimated loads are then summed and compared to measured net loading. These models require the investigator to carefully select or determine appropriate loading rates for Cl^- sources. In a study of Lake Constance, Müller and Gätcher (2012) developed loading rates for livestock, wastewater, wet deposition, and waste incineration based on data provided by local governing bodies (wastewater, wet deposition, and incineration) or collected themselves (livestock manure). Loading rates for rock weathering were previously established for the area, and total road salt application rates within the sub-catchments were provided by government bodies responsible for winter road maintenance. As the total estimated loading for each sub-catchment was much higher than those calculated from river flux measurements, they concluded that ~35% of Cl^- applied was retained within catchment soils and groundwater.

In a study of Lake Iseo, Italy, Nava et al. (2020) used a similar approach to identify sources of Cl^-. Using some loading rate coefficients previously developed by Müller and Gätcher (for livestock and wastewater), and some that were determined for the region of study (road salt and wet deposition), along with historical [Cl^-] data and land use data, they were able to determine that increases in Cl^- loading were due to increases in urban cover (and associated road salt) and population increase (via wastewater). Even though agricultural land use is prevalent in the area, it was concluded that the increase in chloride concentrations was not attributed to livestock as farming activities decreased over the period that [Cl^-] increased.

2.5.3 Combinations of Physically-Based and Statistical Modeling

A few studies have used a mass balance method to quantify watershed Cl^- loads and then used regression approaches to correlate subcatchment characteristics to the concentrations (Winter et al., 2011; Dugan et al., 2021;). In a study of Cl^- loading to Lake Michigan, Dugan et al. (2021) quantified the Cl^- loadings from 234 of the 239 non-nesting tributaries that drain to the lake using a synoptic sampling survey of summer concentrations to represent annual median [Cl^-]. The authors found that tributaries imported 1.079 Tg/yr of Cl^- into the lake, and estimated that this represented 90-98% of the total loading. They found that 70% of this was contributed by five very large subcatchments that had little (<20%) urban landcover. A QRF was then used to determine watershed characteristics that drive tributary [Cl^-]. Parameters investigated included mean imperviousness (%), population density (n/km^2), urban, agricultural, herbaceous, forest, wetland, shrubland, and barren landcovers (%), road density (km/km^2), watershed area (km^2), and Strahler stream order. Mean imperviousness was found to be the most important predictor. Imperviousness and % percent urban were also found to scale linearly with [Cl^-], whereas % agricultural was only found to contribute significantly when >60%.

Interestingly, a similar but more sampling intensive study found that one small but highly urban subcatchment of Lake Simcoe, ON, contributed the majority (43-59%) of the lake's Cl^- load due to very high [Cl], even though larger catchments contributed much higher flows (Winter et al., 2011). The researchers then correlated watershed characteristics with tributary Cl^- flux and found a strong relationship ($r=0.80$, $p<0.05$) with the percent coverage of road and urban landcover. A threshold was found at 25% urban area and road coverage in a watershed, over which [Cl^-] approached or exceeded guideline levels.

2.6 Mitigation Methods and Prevention of High Chloride Concentrations

The most viable options for reducing [Cl^-] in receiving water bodies focus on reducing the amount of Cl^- that is applied within a watershed, either by reducing application rates, or reducing the amount of saltable area. In areas that are already (sub)urbanized, applying salt brines before a storm event has been shown to reduce Cl^- application rates by ~35%, and post storm runoff [Cl^-] by 45% on average (Haake & Knouft, 2019; Claros et al., 2021). Alternative materials for roads and sidewalks can also reduce the salt required per storm. Permeable interlocking pavement requires much less salt, as water is drained from the surface once ice is melted reducing the risk of refreezing and black ice (Marvin et al., 2021). Anti-icing pavements, with embedded slow-release deicers have been developed, but results of outdoor trials were not found (Zheng et al., 2015; Wang et al., 2017). Deicers that do not contain Cl^- are available, but are rarely used either due to prohibitive costs or detrimental effects to receiving environments (Terry et al., 2020; Abbas et al., 2021; Ullah et al., 2021). Rock salt is often overapplied, but tools have been developed to help determine optimal application rates (Kramberger & Žerovnik, 2008; Trenouth et al., 2015). Techniques promoted to reduce salt use in the *Code of Practice for the Environmental Management of Road Salts* include storing salts on impermeable pads and under roofs, utilizing electronic ground speed controllers for salt application, covering mixed salt and abrasive piles, and pre-wetting road salt to reduce undesirable dispersion. In 2021, Environment and Climate Change Canada (ECCC) reported that most road organizations that submitted results (n=220) had implemented the first two techniques, but fewer were using proper abrasive storage, or pre-wetted salt (ECCC, 2021).

New (sub)urban developments can be designed with Cl^- abatement in mind. These efforts should focus on reducing impervious (saltable) area, as this metric has been conclusively linked with increasing and elevated [Cl^-] (Novotny et al., 2008; Corsi et al., 2015; Dugan et al., 2017, 2020; Moore et al., 2020). Reductions in saltable area can be achieved by narrowing roads and sidewalks, designing smaller cul-de-sacs or replacing them with looped streets or T-turn-arounds, shorter or shared driveways, and smaller parking lots (Schueler, 1995, as cited in Minnesota Pollution Control Agency, n.d.). Low and medium density development (suburbs) has been specifically linked to high Cl^- (Dugan et al., 2020), and so should be avoided in favour of higher density-lower footprint developments that need less road area and leave larger areas in a more natural state (Blaszczak et al., 2019). New and expanding urban areas should aim to develop less than 25% of a watershed to minimize Cl^- risks to freshwater (Winter et al., 2011; Lax et al., 2017; Scott et al., 2019).

Strategies that would protect lakes from high Cl^-, but not necessarily other freshwater systems, involve road placement and stormwater management system design. To reduce or delay Cl^- risks to lakes, roads should be built as far away from lakes as possible, at least 10 m, but ideally more than 320 m away especially those that have high salting rates like expressways and major routes. Alternatively, local roads within a 320 m buffer of a lake should not be salted (Kelting et al., 2012). Disconnecting impervious surface can delay lake salinization, but will not likely provide long term protection as runoff is simply redirected to groundwater (Casey et al., 2013; Snodgrass et al., 2017; Burgis et al., 2020; Lam et al., 2020). Nevertheless, ensuring stormwater systems do not discharge directly to lakes could reduce the risk of mono- or meromixis in heavily developed areas (EC & HC, 2001; Sibert et al., 2015).

Capturing and removing from Cl^- from runoff is challenging as it is inert and conservative. Methods that aim to extract Cl^- from stormwater have not yet been proven at scale in cold climates, but there have been some attempts. Phytodesalinization has been tested in lab and greenhouse settings (Ansari et al., 2016; Xu et al., 2019; Schück & Greger, 2022), anthracite and dolomite have been shown to adsorb large proportions of Cl^- from solution (de Santiago-Martín et al., 2016), and Western Michigan University has announced the trial of a soil based Cl^- capture process for a new a stormwater treatment facility but has not yet released details regarding the removal mechanism (Davis, 2022). One method that has been found to reduce the amount of Cl^- that is available to enter surface waters after road salt application is washing streets after winter storms, which can remove >50% of the Cl^- that is left on the roads after deicing is no longer required (Gronba-Chyła et al., 2022).

CHAPTER 3 Methods

3.1 Study Area

All study lakes and their watersheds are located, at least partially, within the HRM. The study area has both urban and rural land use patterns, with most dense urban development occurring near the Halifax and Dartmouth urban centres (Figure 3.1). The HRM was formed from the amalgamation of several communities in 1996 (Jolly, 1996) and has separate land use by-laws for Community Plan Areas which resemble the pre-amalgamation communities (HRM, 2018). Due to their proximity to the coast, lakes in NS have higher background Cl^- concentrations than other parts of Canada, which are estimated to range from 15 to 20 mg/L (EC & HC, 2001).

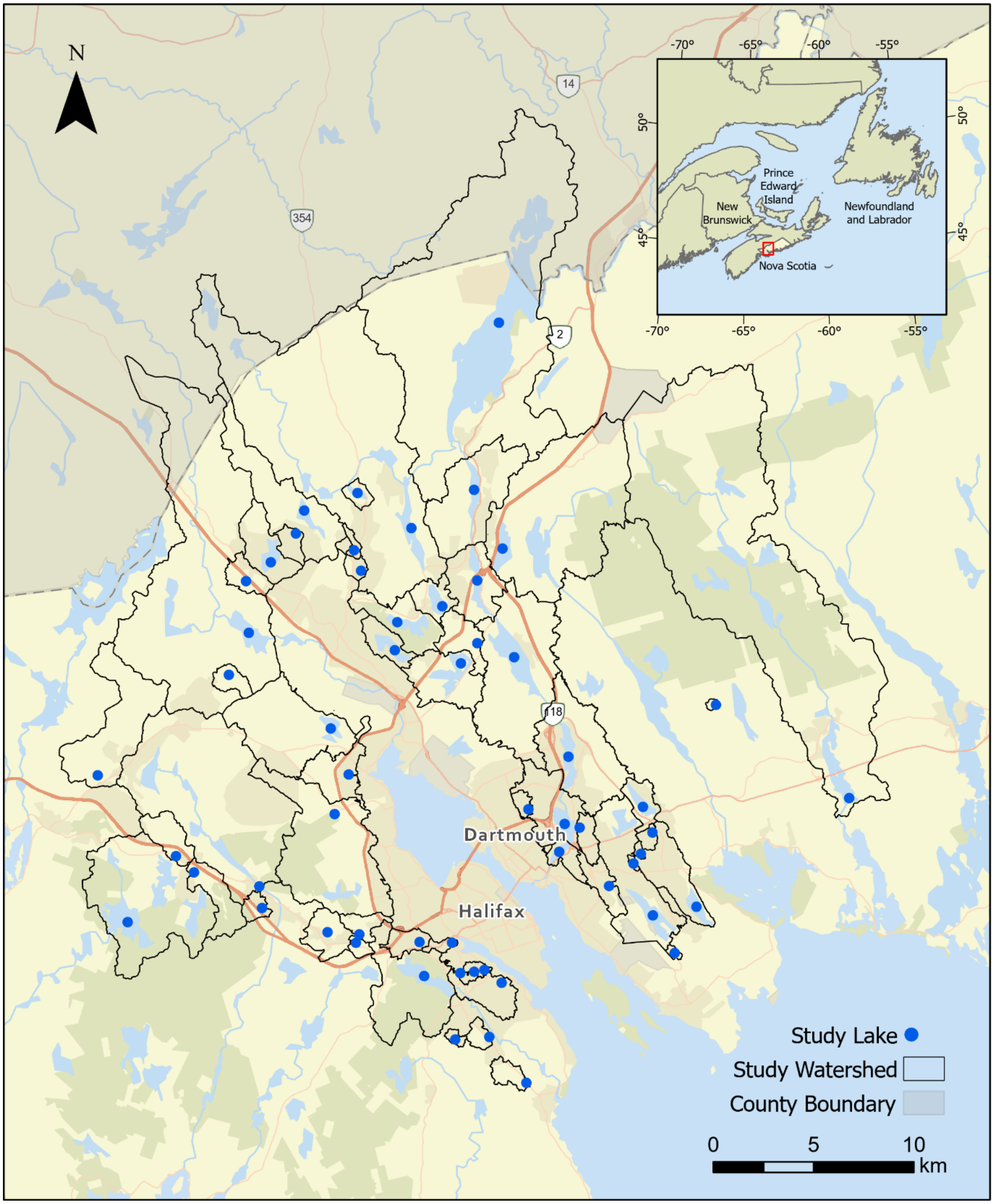


Figure 3.1 Study lakes and their watersheds created and shown in NAD 1983 CSRS UTM Zone 20N. Layers produced based on data from the following sources: lake and stream files (HRM, 2012), (HRM, 2013) 5 m DEM (HRM, 2013), 20 m DEM (NS L&F, 2006), linear flow paths (Natural Resources Canada, 2019), and stormwater infrastructure (Halifax Water, 2019). Basemap, which contains road lines, provided by ESRI.

3.2 Chloride Data

Chloride data was collected by the Energy and Environment division of HRM as part of the Lakes Water Quality Monitoring Program between 2006 and 2011. Water samples were collected from 57 lakes in the municipality three times per year (spring, summer, winter) as surface grab samples. It was indicated that samples were at the location of the deep zone when possible (Stantec, 2012). There were some gaps in the dataset (for example McCabe Lake and Miller Lake were not sampled in Fall 2007), to mitigate this, smooth seasonal variability, and allow for comparison with long term guidelines, mean [Cl^-] over the entire study period were calculated and used for analysis. Summary statistics were calculated in Minitab statistical software to characterize the [Cl^-] data set and spatial autocorrelation was investigated using both global and local Moran's I calculations in ArcGIS Pro. Lakes were then manually categorized into risk levels according to their mean [Cl^-] as compared to the CCME long term guideline for the protection of aquatic life. Risk level groups were used for data visualization and model assessment only.

3.3 Watershed Delineation

Watersheds were delineated from a 5 m LiDAR derived digital elevation model (DEM) where possible (HRM, 2013a), but a provincially available 20 m DEM (NS L&F, 2006) was used for several watersheds. Point-based ArcHydro delineation methods were used (Bajjali, 2018), with the additional step of DEM Reconditioning (Environmental Systems Research Institute [ESRI], 2011) which lowered and created a slight gradient towards roads and streams (HRM, 2012) (lowered by 1 m) which were considered 'preferential flow paths', as well as linear flow paths (Natural Resources Canada, 2019) (lowered by 10 m) which helps maintain hydrological connectivity through lakes. Watersheds were modified manually to account for stormwater infrastructure (Halifax Water, 2019) and neighboring watershed boundaries. All spatial analysis was done in NAD 1983 CSRS UTM Zone 20N projected coordinate system, leading to a small (<0.04%) distortion of length and area within the study area.

3.4 Regression Models

Regressions were carried out in Minitab software using 'mean chloride' as the response variable for all models, and all explanatory variables were input as continuous variables. Accompanying Global Moran's I calculations of model residuals were completed using ArcGIS Pro software with the spatial autocorrelation tool.

3.4.1 Explanatory Variable Processing

All potential explanatory variables were intersected with the watersheds layer and summarized to generate a total length/area/occurrence for each watershed. Explanatory variables that are expressed as a density (road density, stormwater pipe density) or areal percent coverage (surficial geology units, urban area) were normalized to their watershed area (Figure 3.2). Variables investigated included municipally managed infrastructure (roads, sidewalks, salt storage facilities), water infrastructure (stormwater pipes and outfalls, ditches, and wastewater treatment plants) (Halifax Water, 2019), surficial geology units (Stea & Fisher, 2006), distance from the coast (as watershed midpoint to nearest coastline), and urban development. Roads were investigated as total road density, road density by street class type (local (including those with no assigned classification), major collectors, minor collectors, arterials, and expressways), ramp density (in addition to street class), and by maintenance type (ice routes, salt, sand).

All data layers were available, except for an urban land use layer, from within the study period. This layer was created in ArcGIS Pro software using an iso cluster unsupervised classification with 50 classes on a Landsat5 Thematic Mapper image, acquired August 15th, 2009 (USGS, 2009). While 50 classes is likely excessive for this classification, it was necessary to 'reliably' separate bare rock from concrete landcovers. Classes representing urban/developed areas were selected and reclassified into an urban raster. The majority filter tool was applied to remove misclassified cells within patches of urban classified areas. The raster was then converted to a shapefile from which lake polygons were erased to omit rocky shorelines from being included in the developed area estimate. This urban layer was clipped to watersheds before an accuracy assessment was completed using a stratified random sampling as generated by ArcGIS Pro and a confusion matrix.

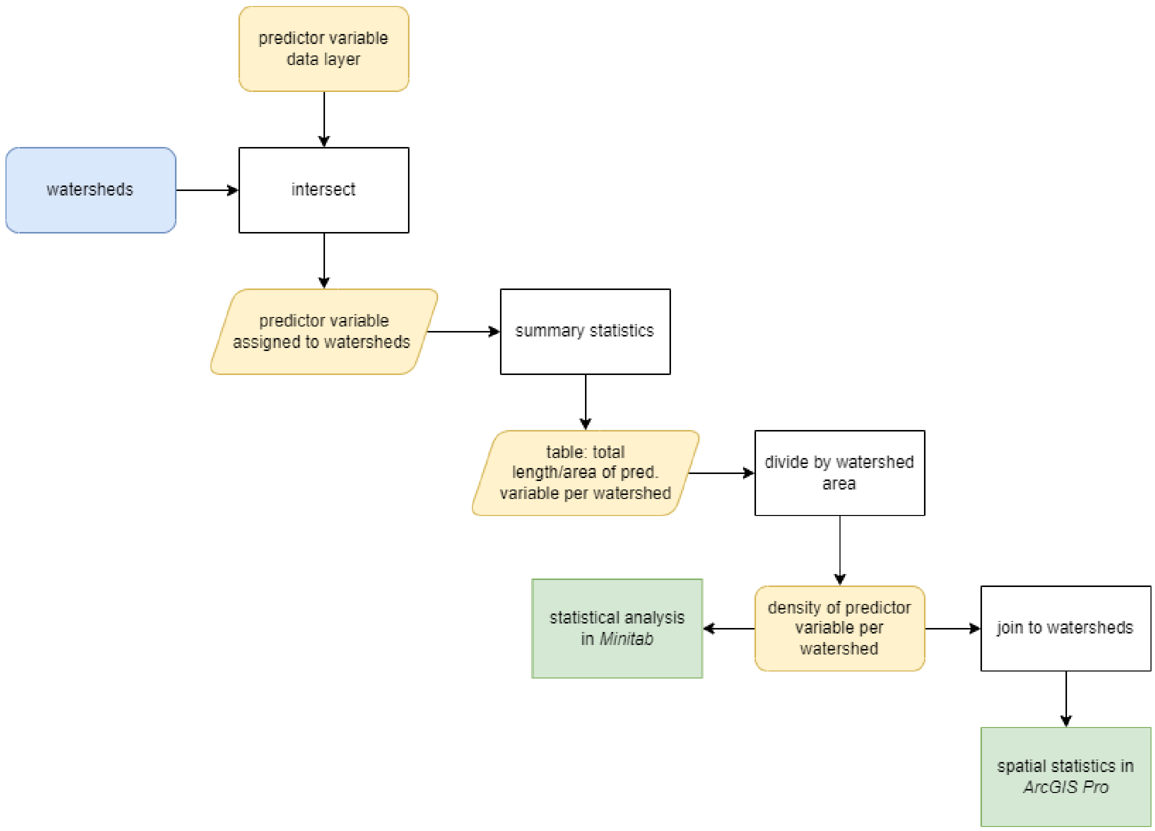


Figure 3.2 Flow chart of variable processing method used for spatially normalized predictor variables in statistical analyses.

3.4.2 Models Explored

Bivariate simple linear regressions (SLR), manually selected models, and models from a best-subsets regression were investigated. A correlation matrix was created in Minitab (Appendix, Figure A2), and variables that had the strongest relationships with [Cl^-] were selected for simple linear regressions. Simple linear regression assumes normal distribution of both the independent and dependent variables, that the true relationship between the two is linear, that all observations are independent, and that errors are randomly distributed. Two models were manually selected for multiple linear regression using variables with known or assumed relationships with road salt. These models were: i) an 'Expressway Model' using an expressway density, the density of all other roads, and ramp density; and ii) a 'Maintenance Model' using the density of salted ice routes, and the density of municipally maintained sidewalks. Best subsets regressions were used to find the most explanatory sets of variables. Models with the highest predictive R^2 values were investigated further; those with constants outside of the background range for chloride, and those that assigned negative coefficients to variables that did not make physical sense, were discarded. Assumptions of this statistical technique are similar to those of SLR, with the additional assumption of no multicollinearity between predictor variables, and unlike SLR, predictor variables are not assumed to be normally distributed.

Models were assessed based on predictive R^2 values between predicted and observed mean [Cl^-]. Variable p-values and variance inflation factors (VIF) were used to assess collinearity (VIF < 5 was considered acceptable) standardized coefficients were used to assess variable contribution, and model constants and corrected Akaike's Information Coefficients (AICs) were used to assess model fit. Anselin's Local Moran's I was carried out in ArcGIS Pro to identify clusters and outliers for [Cl^-] and road density data, to further explore any relationship between total road density and mean lake [Cl^-].

3.5 Mass Balance Models

Land area within each watershed was classified into four land use types: rural, commercial, residential, and roads. Methods based on the concept of 'inverse loading analysis' (Chapra et al., 2009) and adapted from those presented by Novotny and Stefan (2010) were used to calculate a range of loading coefficients for each land use category based on its area within each watershed and the mean [Cl^-] measured in the lake.

3.5.1 Land Use Classification

Zoning data from 2008 was available through HRM. Zoning by-laws differ between community plan areas within the municipality. The by-law document for each plan area was referenced for each zone code (HRM, 2018). Zoned parcels do not include road area, so parcels were classified into three categories (rural, commercial, or residential) based on their by-laws; the landcover of the parcel was only considered where noted. The general method for processing of these variables before they are input to the model is illustrated in Figure 3.3.

The commercial category includes industrial (airport, industrial parks, some portions of mining operations, and some processing facilities), institutional (hospitals, government buildings and schools including greenspaces and sports fields associated with them), and commercial land uses. This category includes some parcels zoned for mixed use where commercial uses were the primary use, however ‘home business’ zones were excluded.

The residential category includes only urban and suburban residential development. This includes any residential zone code listing a minimum lot area of less than 93 m^2 (10000 ft^2). Zones listing separate minimum lot areas for septic and municipally serviced lots were visually estimated for average density using Google Earth imagery from within the study period. Parcels that were zoned for high-low density housing but were undeveloped during the study period were classified as undeveloped. Partially developed parcels were classified as rural residential if the lot size averaged over the parcel area was estimated to be greater than 10000 ft^2.

The rural land use category includes rural residential land uses (residential zones where minimum lot size is 10000 ft^2 or greater, and previously listed exceptions from residential), resource land uses (agriculture, forestry, fisheries, and some of the associated processing facilities), parks and conservation areas (includes parks and some municipally owned recreational facilities including buildings and associated parking lots), nature reserves, wilderness areas, and urban reserves that had yet not been developed during the study period.

Some watersheds extend past HRM municipal boundaries, (Shubenacadie Grand, Kinsac, and McCabe Lakes). For these areas, a land cover database (NS NRR, 2020) was used in lieu of zoning data. These areas were found to be largely resource or rural, except for some more densely developed polygons (classified as urban in the land cover database) which were reclassified to residential land use. A categorized zoning layer was then intersected with the study watersheds, and areas were summarized by land use category to obtain an area per category per watershed.

An approximation of road area was obtained from the 2008 street line file (HRM, 2012), for which line length was summarized by watershed and street class type. These values were then multiplied by their estimated widths from HRM design guidelines (HRM, 2013b) to obtain an area. The guidelines provide a range of road widths for different scenarios in both urban and rural contexts. For this study a value from within the urban range was selected for area calculations because the urban specifications were generally narrower, giving a more conservative estimate, generally these values also fell within the rural range (Table 3.1). Streets with 'null' street class were assumed to be local streets and were assigned the local street width for area calculation. Expressway width was taken to be 12.5 m from the highway design guidelines and was rounded to the nearest half metre, it should be noted highways use a narrower width as each direction is usually represented by a separate street line (NS TIR, 2015).

Table 3.1 Widths used in the calculation of road area for each watershed, as taken from Municipal and Highway Design Guidelines.

Street class	Urban lane width	Rural lane width	Value used
Local	9.0 m	10.5 m	9.0 m
Minor collector	9.0 – 12.0 m	11.0 – 14.0 m	11.5 m
Major collector	15.0 – 20.0 m	12.5 – 20.5 m	16.5 m
Arterial	18.0 – 20.0 m	13.5 – 23.0 m	18.5 m
Expressway			12.5 m

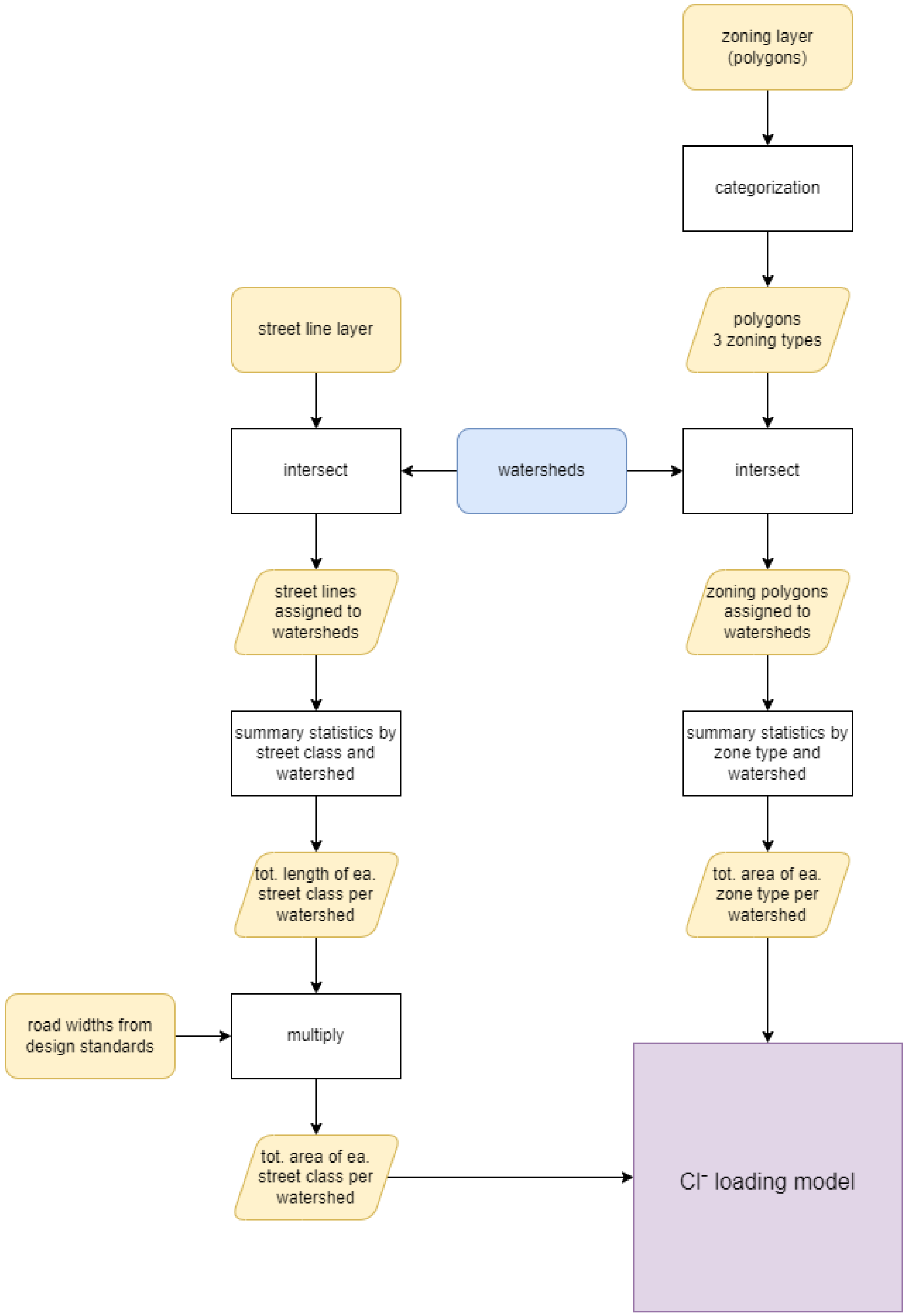


Figure 3.3 Flow chart of variable processing method used to produce land use data input to the mass balance model

3.5.2 Estimation of Chloride Load to Each Lake

An annual Cl^- load (g/yr) was calculated for each study lake using the mean [Cl^-] over the study period and an estimated outflow volume for each lake:

$$\boldsymbol{M_{Cl^-} = \rho_{Cl^-} \times Q_o} \qquad (1)$$

where M_{Cl^-} is the annual Cl^- load to a lake annually (g/yr), ρ_{Cl^-} is the mean lake [Cl^-] (g/m^3), and Q_o is the total hydraulic outflow of the lake (m^3/yr). All lakes were assumed to flush at least once per year. Lake outflows were estimated using the method from the Nova Scotia User's Manual for Phosphorus Prediction (Brylinsky, 2004):

$$\boldsymbol{Q_o = ((A_o \times P_r) + (A_d \times R_u) + Q_i)) - (A_o \times E_v)} \qquad (2)$$

where A_o is the lake surface area (m^2), P_r is the annual unit precipitation (m/yr), A_d is the drainage basin area (excluding lake) (m^2), R_u is the annual unit hydraulic run off (m/yr), Q_i is the upstream hydraulic input (m^3/yr), and E_v is the annual unit lake evaporation (m/yr). Precipitation and evaporation data was obtained from the Environment Canada weather station nearest each lake's centroid (Appendix, Table A6). The long-term average from the Climate Normals for each parameter was used. Runoff values were obtained using a rasterized version of the isorunoff map included in the manual; the runoff value found at the centroid of each watershed was used. In all cases the centroid fell within the watershed. For outflow calculations, all lakes were treated as having no upstream lakes contributing to inflow ($Q_i = 0$), except for those receiving outflow from Lake Charles, which drains in two directions. Outflow from Lake Charles was assumed to drain evenly to Lake Micmac and Lake William (50% each).

3.5.3 Model Set-Up, Calibration, And Performance

Land use loading rates were generated in Excel using the Solver function with the Generalized Reduced Gradient (GRG) Nonlinear solving method. The area of each land use type in a watershed was multiplied by an estimated loading rate for that land use to calculate the contribution of that land use to the lake's Cl^- load. Contributions from each land use were summed across each watershed to obtain an estimated lake chloride load. The model aimed to reduce the root mean squared error (RMSE) between the calculated annual Cl^- loads of the lakes (Equation 1) and those estimated by the model, by changing the loading coefficients over successive iterations. Model performance was judged primarily by the R^2 value between calculated lake Cl^- loads and model estimated Cl^- loads. Because the land use categories used were broad, and salting rates likely vary within them, a range of possible Cl^- loading rates were estimated for each category. The range was obtained by performing a multi-calibration by applying a selection-without-replacement approach. A total of 50 calibration runs were conducted, where in each run 67% of the data was used to fit (calibrate) the loading rates, and the remaining 33% were used to validate the fit. The loading rates for each land use category were then plotted in a box and whisker plot using R Statistical Software (R Core Team, 2022), and the interquartile range was taken as the lower and upper limits of the loading rate range. The upper limit of the range was used to assess model performance based on estimated Cl^- concentration and classification of lakes based on risk level.

CHAPTER 4 Results

4.1 Lake and Watershed Characterization

The dataset represents a range of lake and watershed characteristics (Appendix, Table A1). The smallest lake in the dataset is Dent's Punch Bowl (0.97 ha) while the largest is Shubenacadie Grand Lake (1883 ha); their watersheds cover 9.8 ha and 36 917 ha, respectively. The watersheds studied range widely in their degree of development. This range is seen in the distribution of key indicators like road density (0 – 11.8 km/km^2), expressway density (0 – 2.1 km/km^2), the percent of urban landcover (6.6 – 85.1 %), and the percent coverage of rural (0 – 99.4 %), commercial (0 – 89.7 %), and urban/suburban residential (0 – 76.3 %) land uses. Some lakes receive drainage almost exclusively by piped stormwater conveyance systems (Red Bridge Pond, Cranberry Lake, Settle Lake, and Bissett Lake), while some contain no stormwater piping at all, apart from culverts (Bell, Desaid). Several watersheds extend beyond, or exist outside of, the municipal stormwater service boundary and do not have complete coverage of drainage infrastructure data (Lake Echo, Hubley Big Lake, Black Point Lake, Sheldrake Lake, McCabe Lake, Kinsac Lake, Shubenacadie Grand Lake), but would presumably be serviced by roadside ditches and culverts.

The study area is underlain mostly by ground moraine/streamline drift and rock, with smaller patches of alluvial, colluvial, and organic deposits (Appendix, Table A1). The lakes belong to 12 secondary watersheds and 21 of the lakes in the dataset are headwater lakes.

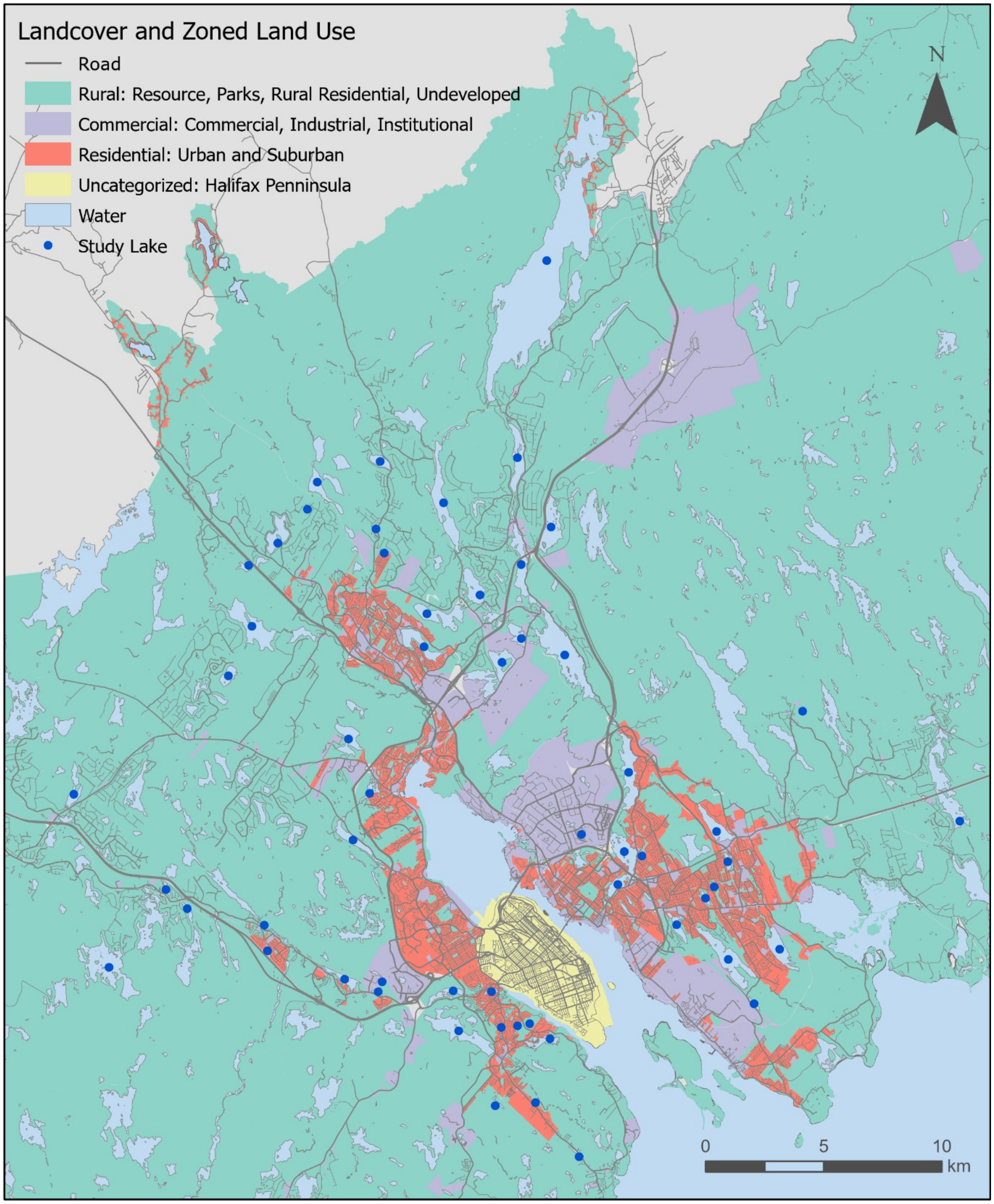


Figure 4.1 Land use in HRM, derived from 2008 zoning data (HRM, 2012), FORNON land cover data (NS NRR, 2020) used in areas where zoning data unavailable.

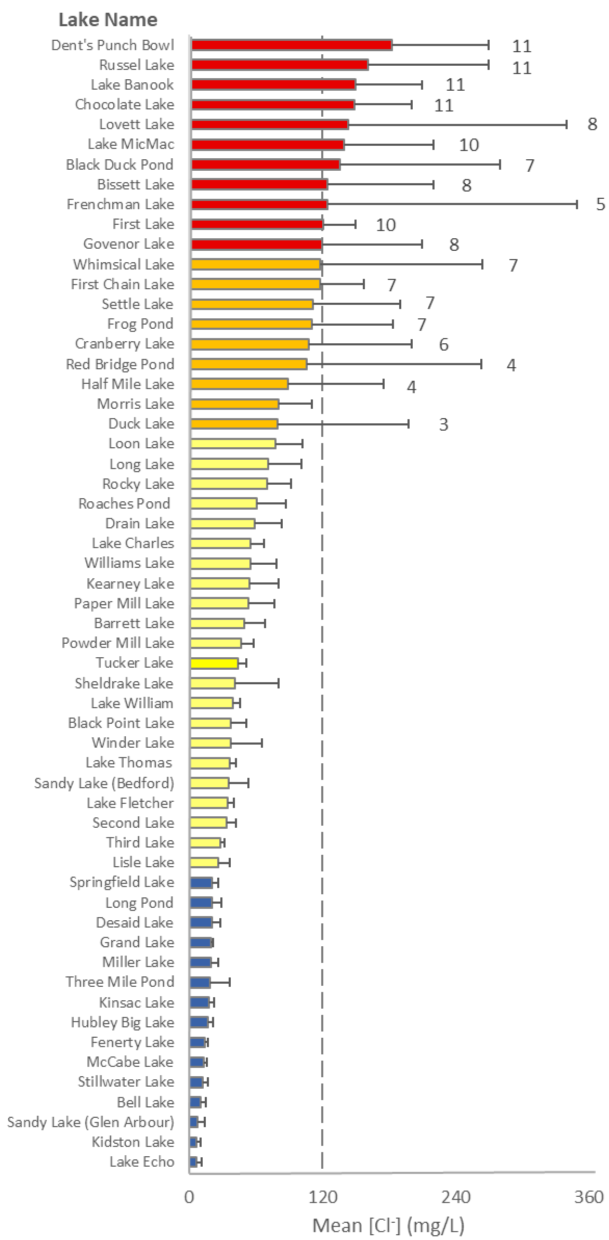
4.2 Chloride Data and Groups

The distribution of mean [Cl^-] among study lakes is positively skewed (skewness = 0.62), with most of the mean concentrations falling below 53 mg/L and a tail containing a few extremely high values. The data are also spatially clustered, with a global Moran's I of 0.47 (z-score = 6.24, $p < 0.001$), and local Moran's I analysis showing with 2 significant high-high clusters occurring in the south of the study area, on either side of the Halifax Harbour (Appendix, Figure A3).

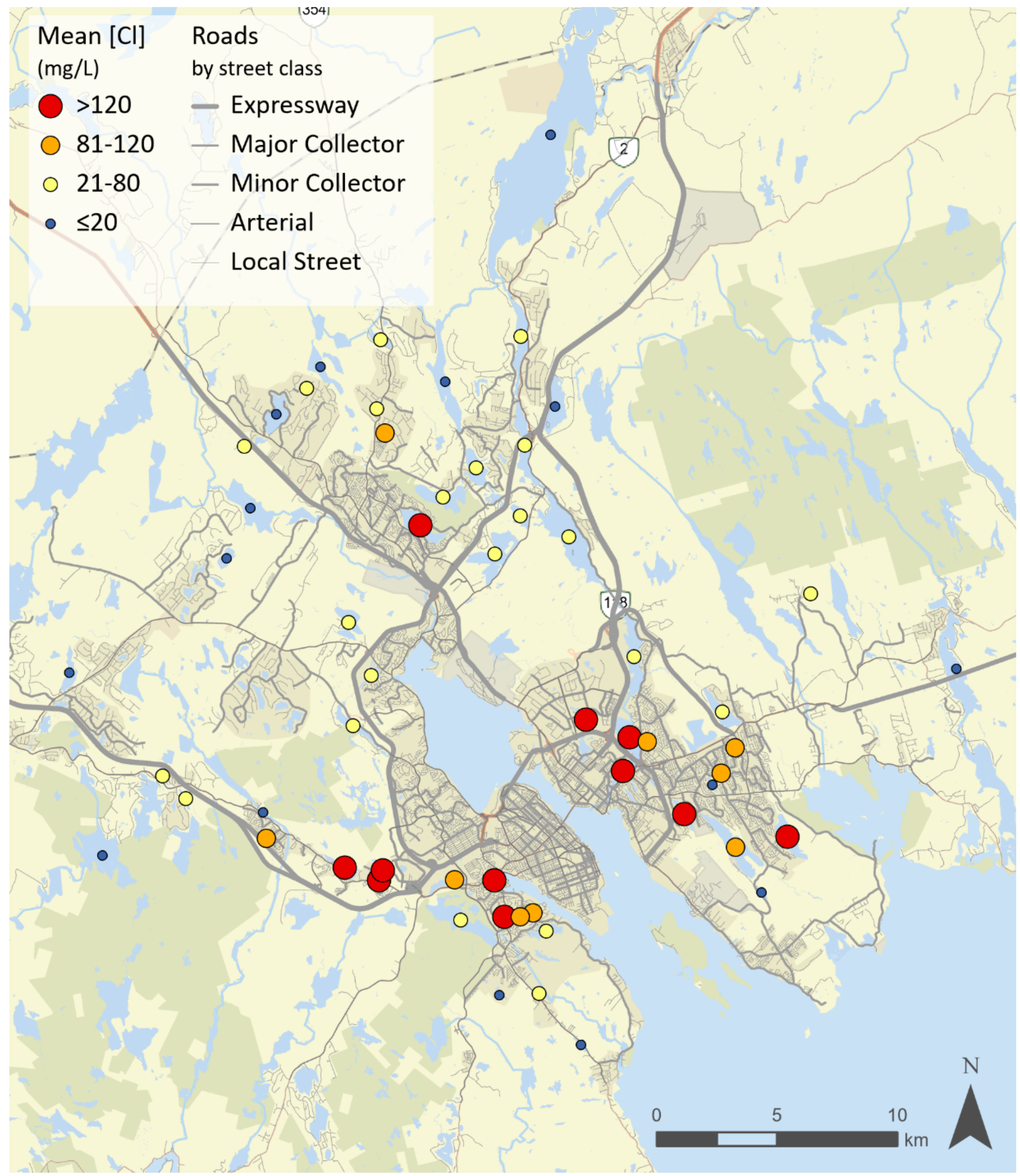
The lakes were divided into 4 groups based on mean chloride concentrations (Figure 4.2). These groups are: impacted lakes (11, red), high risk lakes (9, orange), moderate to low risk lakes (22, yellow), and background level lakes (15, blue). The impacted (red) lakes had mean [Cl^-] that exceed the CCME chronic guideline level of 120 mg/L. Of the 11 lakes in this group, eight had [Cl^-] exceeding guideline levels for three or more consecutive sampling events, indicating that these eight lakes did not comply with the chronic guideline for chloride concentration. Though, according to CCME definitions, these exceedances do not necessarily indicate negative impacts, the United States Environmental Protection Agency (US EPA) states that systems that exceed chronic guidelines more frequently than once in any three year period are in a constant state of recovery and so are likely to be impacted (US EPA, 2017). High-risk lakes (orange) had mean [Cl^-] that fell between 80 mg/L and 119 mg/L. Of the nine lakes in this category, eight had at least one [Cl^-] measurement that exceeded the chronic guideline. Though sporadic exceedances of the guideline do not indicate chronic impairment of the ecosystem due to Cl^-, recent evidence suggests food-webs may be impacted at levels below the chronic guideline (Hintz et al., 2022). Due to these recent findings, and their proximity to the chronic guideline, these lakes were categorized as high risk. Moderate/low risk (yellow) lakes experienced mean [Cl^-] between 21 and 79 mg/L. None of the 22 lakes in this category possessed [Cl^-] that exceeded the chronic guideline during sampling events, but all of these lakes had mean [Cl^-] that exceeded background levels. For these reasons, these lakes were considered to be at low or moderate risk of the impacts of Cl^-. Background lakes (blue) had mean [Cl^-] that were less than 20 mg/L.

Chloride concentrations in these lakes fell within the range of background chloride concentrations for NS freshwaters of 10-20 mg/L (EC & HC, 2001). Though some individual lakes in this category experienced seasonal Cl^- patterns consistent with de-icing salt impacts, the mean concentrations are assumed to be low enough to avoid negative impacts on ecosystems associated with Cl^-.

Figure 4.2 Right: study lake locations and chloride levels basemap provided by ESRI. Left: mean lake [Cl^-] (2006-2011), error bars showing maximum observed chloride concentration, number of exceedances over study period shown beside error bar (0s not shown), CCME chronic guideline for the protection of aquatic life shown by dashed line. Marker and bar colours correspond with chloride-impact risk categories.



Lake Name
Dent's Punch Bowl
Russel Lake
Lake Banook
Chocolate Lake
Lovett Lake
Lake MicMac
Black Duck Pond
Bissett Lake
Frenchman Lake
First Lake
Govenor Lake
Whimsical Lake
First Chain Lake
Settle Lake
Frog Pond
Cranberry Lake
Red Bridge Pond
Half Mile Lake
Morris Lake
Duck Lake
Loon Lake
Long Lake
Rocky Lake
Roaches Pond
Drain Lake
Lake Charles
Williams Lake
Kearney Lake
Paper Mill Lake
Barrett Lake
Powder Mill Lake
Tucker Lake
Sheldrake Lake
Lake William
Black Point Lake
Winder Lake
Lake Thomas
Sandy Lake (Bedford)
Lake Fletcher
Second Lake
Third Lake
Lisle Lake
Springfield Lake
Long Pond
Desaid Lake
Grand Lake
Miller Lake
Three Mile Pond
Kinsac Lake
Hubley Big Lake
Fenerty Lake
McCabe Lake
Stillwater Lake
Bell Lake
Sandy Lake (Glen Arbour)
Kidston Lake
Lake Echo
11
11
11
11
8
10
7
8
5
10
8
7
7
7
7
6
4
4
3
0
120
240
360
Mean [Cl-] (mg/L)



Mean [Cl]
(mg/L)
>120
81-120
21-80
≤20
Roads
by street class
Expressway
Major Collector
Minor Collector
Arterial
Local Street
0
5
10
km
N

4.3 Correlations and Regressions

Road density, stormwater pipe density, and percent urban coverage (overall accuracy = 78%, K_{hat} = 0.56, confusion matrix Appendix, Table A2) of a watershed had the strongest relationships with mean lake [Cl^-] of the variables explored (Figure 4.3). Road density alone was able to explain 48% of the variability in mean [Cl^-] in the study lakes (Table 4.1). Additionally, Cluster Analysis (Anselin Local Moran's I) showed some consistency between clusters formed on the basis of [Cl^-] and those formed based on road density. Lakes/watersheds that appeared in high-high clusters in both analyses were: Russel Lake, Red Bridge Pond, Cranberry Lake, Loon Lake, Frenchman Lake, Whimsical Lake, and Frog Pond. Bell Lake appeared as an outlier (high-low) in both analyses (Figure 4.4). Stormwater pipe density was able to explain 55%, and urban landcover was able to explain 64% ,of the variability in study lakes, these predictor variables are highly colinear (Appendix, Table A3) and so could not be combined in MLR.

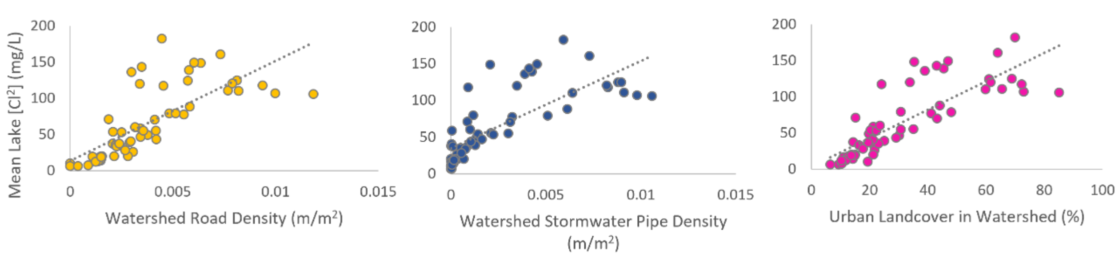


Figure 4.3 Correlations between [Cl^-] and the explanatory variables: road density (left, yellow, R^2-pred = 0.48), stormwater pipe density (middle, blue, R^2-pred = 0.55), and urban landcover (right, pink, R^2-pred = 0.64).

Two roads-based models that incorporated more variables had reasonable coefficients for all variables and performed better than the 'roads only' model but not better than the 'urban landcover' model. An 'expressway' model that separated expressways from other road types and added a 'ramp density' variable explained 56% of chloride variability with ramp density having the highest coefficient, followed by expressway density, and then the density of other road types. While the VIF for each variable was acceptable, the p-value of most of the variables in the model did not meet significance criteria. A ‘municipal maintenance’ model that considered only the densities of roads that are maintained by the municipality where salt is used (sanded roads excluded), and sidewalks maintained by the municipality performed only very slightly better than the ‘roads only’ model and explained 48% of chloride variability. This model also produced a constant that was higher than background levels. The ‘urban landcover’ model performed better than both other bivariate models and ‘hand-picked’ models in terms of both predictive R^2 and corrected Akaike’s Information Coefficient (AIC_c).

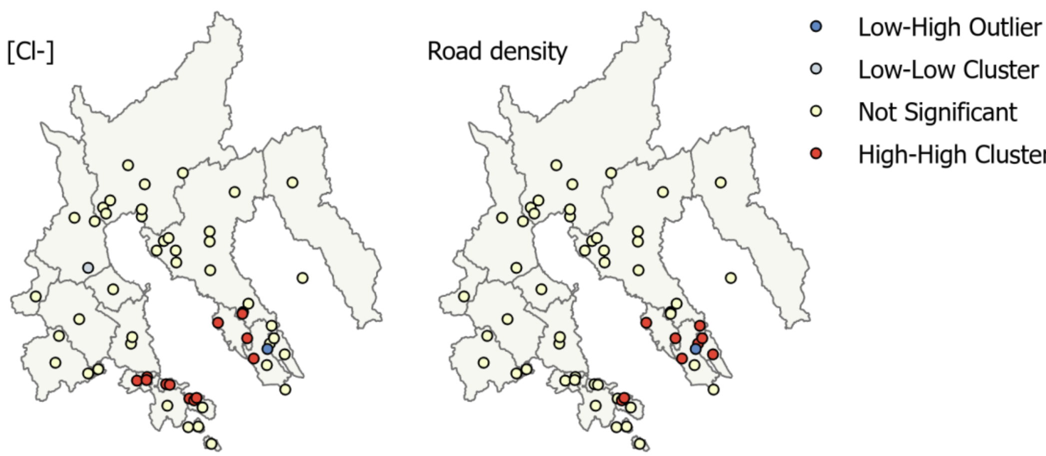


Figure 4.4 Results of cluster analyses, on mean lake chloride concentration (left) and road density (right). High-high shown in red, low-high shown in blue.

The best performing model found (‘best subsets 2’) included ramp density, stormwater pipe density, the percent of the watershed where the surficial geology was listed as ‘rock’, and the distance from the midpoint of the watershed to the nearest coast as predictor variables. The model was able to explain 71% of variability in mean [Cl^-], with all variables having acceptable variance inflation factors. The standardized coefficients in this model show that the density of stormwater pipes has the greatest impact on the model estimated mean [Cl^-]; it also generated a low variance inflation factor (VIF) and p-value. Though the ‘distance to coast’ variable produced a large p-value (0.22), and the constant of the model was higher than the background range of lake [Cl^-], the AICc of this model was the lowest of all models explored (Table 4.1) and it was the only model that produced spatially random residuals (Appendix, Table A4). Residual plots show near normal histogram distribution, and a slight fan-shaped distribution when compared to fitted value (Appendix, Figures A5 & A6). Therefore, 'best subsets 2' represents the best overall model performance.

Though 'best subsets 1', which uses the same predictor variables as 'best subsets 2' but excludes 'distance to coast', produced a higher AICc and slightly spatially clustered residuals, it performed reasonably well. The model was able to explain 71% of variability in mean [Cl^-], with all variables having acceptable p-values and variance inflation factors. The constant generated by the model was slightly higher than background concentrations (20.93), but lower than that of 'best subsets 2'. Residual plots show near normal histogram distribution, but the distribution of residuals vs. fitted values (Appendix, Figures A7 & A8) is slightly more fan-shaped than that of 'best subsets 2'.

Table 4.1 Regression analysis of lake chloride concentrations as explained by watershed variables. Analysis performed using Minitab and ArcGIS Pro software.

Model	Units	Coeff.	B-Coeff.	p-value	VIF	R^2_{Adj}	R^2_{Pred}	AIC
Roads Only						0.54	0.51	563
Constant	mg/L	14		0.088				
Road Density	m/m^2	13793	0.74	0	1.00			
Stormwater Only						0.58	0.55	559
Constant	mg/L	35		0				
Stormwater Pipe Density	m/m^2	11886	0.76	0	1.00			
Urban Cover Only						0.67	0.64	545
Constant	mg/L	3		0.617				
Urban Land Cover	%	2	0.82	0	1.00			
Expressway Model						0.60	0.57	559
Constant	mg/L	11		0.162				
Expressway Density	m/m^2	17523	0.19	0.151	2.29			
Density of Other Roads	m/m^2	12812	0.68	0	1.01			
Ramp Density	m/m^2	41954	0.22	0.089	2.29			
HRM Maintenance Model						0.53	0.48	566
Constant	mg/L	32		0				
Density of Salted Ice Routes	m/m^2	9675	0.56	0.003	3.78			
Density of Salted Sidewalks	m/m^2	7653	0.20	0.261	3.78			
Best Subsets 1						0.75	0.71	531
Constant	mg/L	21		0				
Ramp Density	m/m^2	62066	0.33	0	1.09			
Stormwater Pipe Density	m/m^2	11861	0.76	0	1.00			
Percent Rock	%	0.30	0.19	0.009	1.09			
Best Subsets 2						0.76	0.71	512
Constant	mg/L	24		0				
Ramp Density	m/m^2	62124	0.36	0	1.09			
Stormwater Pipe Density	m/m^2	11512	0.74	0	1.08			
Percent Rock	%	0.25	0.12	0.013	1.10			
Distance to Coast	km	-7E-05	-0.05	0.219	1.09			

4.4 Mass Balance Model

The multi-calibration process resulted in R^2 values that ranged from 0.88 to 0.99 for training data sets and 0.69 to 0.99 for testing datasets (Appendix, Table A7). In all calibrations, the land use type with the highest Cl^- loading coefficient was roads, followed by urban/suburban residential (Figure 4.5). The ranking of commercial/industrial and rural land use types was more variable, with 21 calibrations generating a higher loading coefficient for commercial/industrial, 23 generating the higher coefficient for rural, and 6 calibrations attributing no Cl^- loading to either of these land use types (coefficient = 0.00).

4.4.1 Coefficients

4.4.1.1 Roads

Roads were found to contribute considerably to lake Cl^- loading in every calibration set, with a minimum estimated loading coefficient of 579 $g/m^2/yr$. This land use had the highest estimated loading rate, with over 15% of calibrations generating a 'roads' loading rate above 1000 $g/m^2/yr$, an order of magnitude higher than other land uses. The range of estimated loading rates as taken from the interquartile range of the multiple-calibration is 840-965 $g/m^2/yr$ (Table 4.2).

4.4.1.2 Urban/suburban residential

Urban and suburban residential land use contributed to lake Cl^- loading in every calibration iteration, with the lowest estimated residential loading rate being 57 $g/m^2/yr$. This land use produced the highest loading rate of the zoned land use types (which does not include road area), with an estimated loading rate range of 95-160 $g/m^2/yr$.

4.4.1.3 Commercial/Industrial/Institutional

The commercial land use type was found to have minimal contributions to lake Cl^- loading in more than half of the calibration iterations (26 calibrations, loading rate = 0). The maximum loading rate estimated was 42 $g/m^2/yr$, but this was considered an outlier. The reported range of estimated loading rates is 0-10 $g/m^2/yr$. This land use category was arguably the most variable of those explored, with parcels that range from large shopping centres to rural lots with large, yet undeveloped portions.

4.4.1.4 Rural: Resource, Parks, Rural Residential, and Undeveloped

The rural land use type generally had low loading rates in all calibrations. This land use was found to have negligible contributions to Cl^- loading in just over 15% of calibration iterations (8 calibrations with loading rate = 0). The maximum observed loading rate was 6 $g/m^2/yr$ which was not considered an outlier and was included in Q4. The interquartile range of loading rates generated for rural land use was found to be 1-3 $g/m^2/yr$.

4.4.2 Statistics: Coefficients

The distribution of coefficients found in the multi-calibration process was normally distributed for road and urban/suburban land use types and not normally distributed for rural and commercial land use types ($p < 0.05$ in normality test). The median estimated loading rate of each land use type was found to be significantly different from that of all other types via a Mann-Whitney test, except for those of rural and commercial land uses. These two land use types may not contribute significantly different Cl^- loading to lakes when considered as a whole, however a finer resolution within these categories may be required to distinguish them.

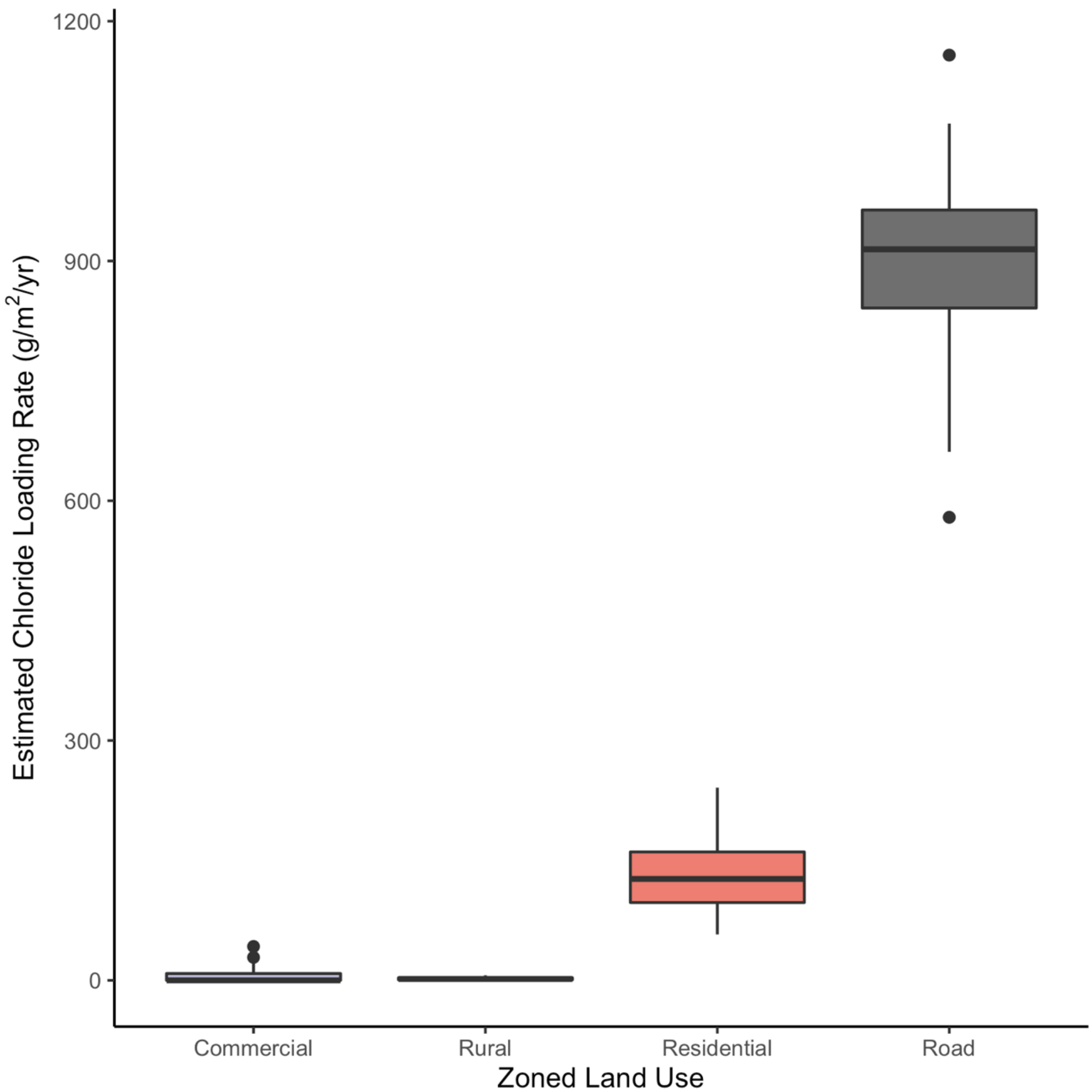


Figure 4.5 Boxplot of Cl^- loading rates for each land use type as generated in multiple calibration.

Table 4.2 Estimated loading rate ranges of four land use types. Values shown are interquartile range of estimated loading rates from 50 calibration datasets.

Land Cover or Zoned Land Use	Estimated Cl^- Loading Rate ($g/m^2/yr$)
Commercial, Industrial, Institutional	0 - 9
Rural: Resource, Parks, Rural Residential, and Undeveloped	1 - 3
Residential: Urban and Suburban	97 - 162
Road	840 - 964

4.4.3 Q3 Test/Validation

Using the top of the interquartile loading rate range (Q3 values) for each land use to model Cl^- loading to all lakes/watersheds produced an R^2 value of 0.97 when comparing calculated (~observed) and estimated loading rates (Appendix, Table A8). When these Cl^- loads are used to back-calculate an estimated lake [Cl^-], and this is compared with the observed mean [Cl^-], a lower R^2 value is achieved (0.57). The three most overpredicted lakes were Cranberry Lake, Red Bridge Pond, and Roaches Pond. Two of these (Red Bridge Pond and Cranberry Lake) have very high road coverage (>14%), but less extreme mean [Cl^-] (~100 mg/L). The three most underpredicted lakes were Dent's Punch Bowl, Black Duck Ponds, and Lovett Lake, all three have average road coverage (~5% of watershed) but very high [Cl^-] compared to the rest of the dataset, and two of the three have very high proportions of commercially zoned area (>50%). Additionally, three of these worst predicted lakes are small lakes with highly urbanized watersheds (>70% urban; Cranberry Lake, Dent's Punch Bowl, and Red Bridge Pond). Using the predicted [Cl^-], ~58% of lakes would be put in their correct risk category (red, orange, yellow, blue), but only ~19% would be placed in a risk category lower than their observed [Cl^-] would place them in. Of the 11 lakes that were estimated to belong to a lower risk category, nine have >10% or their watershed area zoned as commercial.

CHAPTER 5 Discussion

Chloride is threatening the ecological health of lakes throughout the HRM, with most lakes possessing [Cl^-] that exceed background levels, and 11 of the 57 study lakes exceeding the CCME [Cl^-] guideline of 120 mg/L during the study period (2006-2011). Recent studies have also questioned whether this guideline value is low enough to be protective of lake ecosystem function (Arnott et al., 2020; Hintz et al., 2022). With Cl^- gaining attention as a constituent that adversely effects lake food chains, pushing them towards greater primary productivity and potentially promoting phytoplankton blooms, further study and greater focus on the reduction of lake [Cl^-] is needed (Hébert et al., 2022; Hintz et al., 2016). The intent of this study was to determine the primary drivers of high [Cl^-] in HRM lakes and develop generalized loading rates that can be used for land use planning.

5.1 Regression Models

The strongest single variable predictor of lake [Cl^-] was the percent of urban landcover in a watershed (R^2 = 0.64). This result is consistent with the findings from Dugan et al. (2020) and Scott et al. (2019). This relationship indicates that concentrations of Cl^- in HRM lakes are affected by anthropogenic activities associated with urban land use, such as road salting. However, the threshold of 25% urban development found by both Scott et al. (2019), and Winter et al. (2011), was not evident in this dataset (~60% urban development). Nevertheless, the relationship drawn between urban development and [Cl^-] from HRM lakes was statistically significant and supports a strong connection between these characteristics. Though highly colinear with % urban development (as they are all likely acting as proxies for road salt application), road density(s) and stormwater pipe density were also strong predictors of [Cl^-], consistent with many other studies (Blaszczak et al., 2019; Dugan et al., 2017; Kelting et al., 2012; Snodgrass et al., 2017). It should be noted that several of the assumptions of SLR were not met in this analysis. Firstly, neither variable was normally distributed in any of the relationships explored, all distributions were positively skewed (Appendix, Figure A2). As well, it could be argued that the observations used to quantify these relationships may not be independant as many of the study watersheds are nested. This violation of model assumptions could possibly have been mitigated by using ‘snap shots’ from the [Cl^-] time series data rather than mean [Cl^-] over the study period. The similarities between results of cluster analyses of [Cl^-] and watershed road density, may also indicate a relationship between these variables, especially in the positive ends of thier distributions, towards which both variables were skewed. However, the results of one cluster analysis were not perfectly mirrored in the other, indicating that other variables also influence lake [Cl^-].

The model that best predicted lake [Cl^-] ($R^2 = 0.71$) included ramp density, stormwater pipe density, % rock (surficial geology), and distance to the coast. The first two factors are associated with urban development and road salt application, while % rock may represent a lack of storage in soils, or regional differences in salt application rates. Though the addition of ‘distance to coast’ to model predictors improved model performance by decreasing AIC_c and reducing clustering of residuals, caution should be taken in interpreting this as a predictor variable. While it may represent higher background concentrations nearer to the coast due to atmospheric deposition, many of the heavily developed urban centres are coastal and very few rural study watersheds were as near coastlines. Thus, this model may be including ‘distance to coast’ as a proxy for ‘ruralness’, as all proxies for ‘urban-ness’ are highly correlated with stormwater pipe density (Appendix, Figure A4). Point sources of Cl^- (salt storage facilities and wastewater treatment effluent) were not found to be among the strong predictors of [Cl^-], though they have been significant sources of Cl^- in other studies (Nava et al., 2020; Oswald et al., 2019; Tharp & Allen, 2020).

5.2 Mass Balance Model

During the calibration of the mass balance model, road area was consistently assigned the highest loading rate, which converges with the results of the regression analysis, as well as previous studies, and gives credibility to this estimation technique (V. R. Kelly et al., 2008; Novotny et al., 2009; Harte & Trowbridge, 2010; Trowbridge et al., 2010; Laceby et al., 2019). The Cl^- loading rates generated for road area and residential area

(964 and 162 g/m^2/year, respectively) are much higher than other reported values for roads and residential impervious area (45-220 and 157 g/m^2/year, respectively) (Novotny & Stefan, 2010 & Lam et al., 2020). While the estimate for road area may seem comparatively high, it is equivalent to ~1 L of rock salt/m^2/year, which is not unreasonable given the high provincial and municipal application rates, and winter storm frequency (EC & HC, 2001; HRM, 2017). The loading rate for residential area is also significantly higher than those reported elsewhere as it is normalized by total zoned area and excludes roads.

Conversely, the loading rate generated for commercial areas (9 g/m^2/year) appears to be much lower than those reported elsewhere (662 g/m^2 impervious/year) (Lam et al., 2020). This rate is normalized by total zoned area, much of which remained undeveloped during the study period. The range of values generated for commercial land use in the multi-calibration process (0-42 g/m^2/year) highlights the variability of land cover in areas zoned as commercial in this dataset, and the limitations of this method. Further evidence that the commercial loading rate was either poorly estimated or is highly variable is given by the high proportion of severely underestimated lakes that were largely zoned as commercial (9 of 11). However, difficulty estimating commercial application rates is not unique to this study, as they can vary widely between commercial properties even when normalized by impervious area (Lembcke et al., 2017).

The lowest loading rate generated was for rural land use (3 g/m^2/year), which cannot be directly compared to published activity-based rates (Müller & Gächter, 2012). This rate may be underestimated due to the assumption of 'no net Cl^- retention' in watersheds. Studies indicate that large proportions of Cl^- can be stored in soils, groundwater, and surface water, which can be Cl^- sinks before these systems achieve steady state (Novotny et al., 2009; Müller & Gächter, 2012; Dugan et al., 2021). However, the assessment of subsurface flow paths, and if individual watersheds had come to steady-state with respect to Cl^-, was outside the scope of this study. More data could be collected in future investigations to confirm or refute this assumption and make necessary modifications to the loading rates.

Other assumptions that may affect the validity of this model include:

1) Study lakes flushed at least once per year, and therefore mean [Cl^-], rather than Δ[Cl^-], was appropriate for determining annual loading;

2) Study lakes were completely mixed with respect to Cl^- during each sampling event, making surface [Cl^-] representative of volume weighted [Cl^-]; and

3) All land uses are salted evenly (within their category), and therefore the use of consistent areal rates was appropriate.

While there is evidence to suggest that these assumptions may not be valid, they were necessary to carry out the analysis with the available data (Smoll et al., 1983; EC & HC, 2001; Kelting et al., 2012).

Combining physically-based and statistical modeling approaches allowed Cl^- loads to be estimated and apportioned to different sources. This methodology was especially useful in this context due to the non-point source nature of the contaminant, and the fact that Cl^- is largely conservative in the environment. Furthermore, the areal loading rates generated in this study are based on land use data that are usually readily available (zoning and road networks), potentially making it a more practical method in jurisdictions with few resources to track salting rates.

It is understood that the assumption of lakes being flushed at least once per year might not be valid in other regions. In these cases, the apportionment step could be combined with more complex physically-based modeling techniques to determine loads, like those presented in Chapra et al. (2009) or Novotny and Stefan (2010). Locally appropriate loading rates that are generated could then be used in models that predict future [Cl^-], like those developed by Trowbridge et al (2010). Obtaining regional, and land use specific loading rates, is important for predicting Cl^- levels in receiving water bodies as they can vary with location, operator preference, and policy. This has proven to be a challenge in areas with mixed land use catchments (Kelting et al., 2012; Lembcke et al., 2017; Moore et al., 2020), but this study presents a possible solution to this issue.

The HRM continues to experience ongoing urban sprawl. Lakes are abundant in these areas and therefore more lakes are being impacted by this development (Doucet et al., *in review*).The Cl^- loading rates generated in this study could be used to estimate the impacts of land use change on HRM lakes that are currently unmonitored, or to assess the effect of salt management strategies (e.g. reduced salting rates) on lakes that currently have elevated concentrations of Cl^-. Use of Q3 values generated from the calibration of the mass balance model are recommended as a conservative approach when conducting impact assessments. As Cl^- is increasingly recognized as a contaminant of concern, urban planning that considers salting impacts early in the planning process should become standard practice, while Cl^- reduction measures on existing urban landscapes should be implemented in parallel.

CHAPTER 6 Conclusions and Recommendations for Future Research

Many lakes in the HRM with highly developed watersheds are approaching or exceeding the chronic freshwater aquatic life guideline for Cl^-. These elevated levels are largely due to road salt use within watersheds. These exceedances represent an ecological risk that requires mitigation for lakes currently experiencing high Cl^-, and preventative steps should be taken to protect lakes that will be impacted by future development. In order to effectively mitigate and manage this risk, the sources of Cl^- to lakes must be well understood. This thesis showed that roads in HRM contribute the highest Cl^- loadings to lakes compared to other types of land use, and that these rates are higher than those estimated for other cities. Thus, urban planning that minimizes road area might help protect lakes from high [Cl^-]. In areas that are already developed, technologies that reduce the amount of salt applied, while maintaining road safety may help mitigate the existing risks.

The loading coefficients for roads, residential, commercial, and rural land uses presented here could be used to estimate Cl^- loading to unmonitored lakes in the municipality. From a policy perspective, this subsequent analysis can be used to identify lakes that are vulnerable to impacts from road salt and help fulfill this objective in the Road Salt Code of Practice. Alternatively, the model could also be used to predict the Cl^- loading the new development would add to lakes, to assess the risks that a development poses to aquatic systems.

With regards to future work the model calibration could be improved with more data, and more current data may be required to reflect any changes in salting practices that may have occurred since the study period. Additionally, the coefficient for rural land use may require an assessment of Cl^- potentially stored in soils and groundwater to confirm or improve its acceptability. The commercial loading rate could also be improved with finer resolution data (more categories) and/or the omission of yet undeveloped but commercially zoned polygons to improve the accuracy of the estimated loading rate.

Estimates of event-based average loading rates could be produced from those presented here by considering the average number of annual winter weather events during the study period. These per-event estimates could be compared with those of other cities and NS/HRM's own published loading rates more easily. The potential for the reduction of salt use could then be more accurately assessed.

The monitoring program that provided the data used for this thesis concluded in 2011. The reintroduction of a similar surveillance monitoring program would help assess and inform the management of the current risks to local lakes posed by Cl^- and other stressors.

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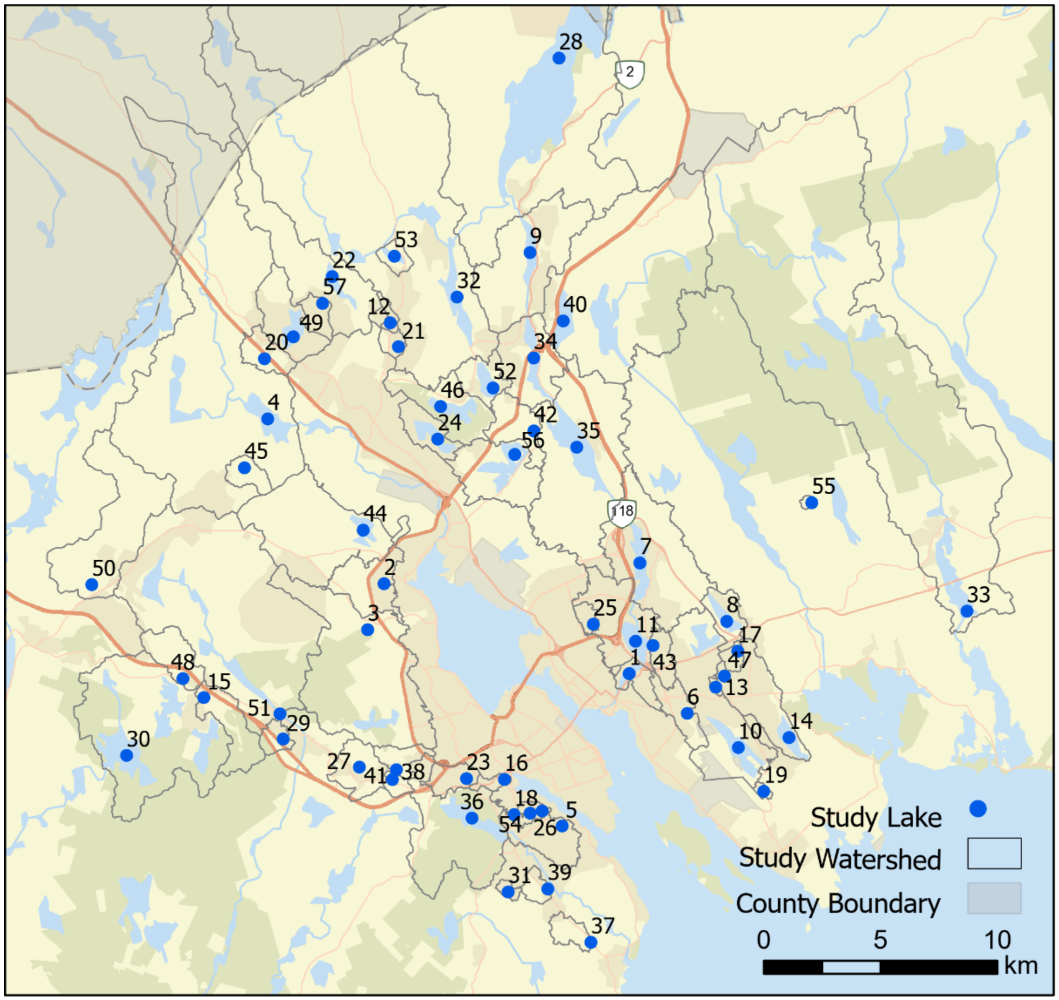
APPENDIX: Supplemental Figures and Tables



Lake Banook	1	Hubley Big Lake	30
Paper Mill Lake	2	Kidston Lake	31
Kearney Lake	3	Kinsac Lake	32
McCabe Lake	4	Lake Echo	33
Williams Lake	5	Lake Thomas	34
Russell Lake	6	Lake William	35
Lake Charles	7	Long Lake	36
Loon Lake	8	Long Pond	37
Lake Fletcher	9	Lovett Lake	38
Morris Lake	10	Roachs Pond	39
Lake Micmac	11	Miller Lake	40
Barrett Lake	12	Black Duck Ponds	41
Bell Lake	13	Powder Mill Lake	42
Bissett Lake	14	Red Bridge Pond	43
Black Point Lake	15	Sandy Lake (Bedord)	44
Chocolate Lake	16	Sandy Lake (Glen Arbour)	45
Cranberry Lake	17	Second Lake	46
Dent's Punch Bowl	18	Settle Lake	47
Desaid Lake	19	Sheldrake Lake	48
Drain Lake	20	Springfield Lake	49
Duck Lake	21	Stillwater Lake	50
Fenerty Lake	22	The Mill (Three Mile Pond)	51
First Chain Lake	23	Third Lake	52
First Lake	24	Tucker Lake	53
Frenchman Lake	25	Whimsical Lake	54
Frog Lake	26	Winder Lake	55
Governor Lake	27	Rocky Lake	56
Grand Lake	28	Lisle Lake	57
Half Mile Lake	29		

Figure A1 Labeled map of study lakes.

Table A1 Lake and watershed variables used in analysis.

Lake Name	Lake Area (ha)	Watershed Area (ha)	Road Density (m/m^2)	Expressway Density (m/m^2)	Ramp Density (m/m^2)	Road Density excluding expressways (m/m^2)	Salted Ice Route Density (m/m^2)	Sidewalk (maintained) density (m/m^2)	Stormwater Pipe Density (m/m^2)	Surficial Geology Ground Moraine (%)	Surficial Geology Rock (%)	Urban Landcover (%)	Rural Land Use (%)	Commercial Land Use (%)	(Sub)urban Residential Land Use (%)	Estimated Landcover Roads
Barrett Lake	9.00	71.34	3.78E-03	0	0	3.78E-03	3.43E-03	1.18E-03	1.34E-03	100.0	0.0	19.8	94.6	0.0	0.0	5.4
Bell Lake	9.70	25.18	0	0	0	0	0	7.19E-05	0	100.0	0.0	19.3	69.3	0.0	30.7	0.0
Bissett Lake	89.54	792.75	8.12E-03	0	0	8.12E-03	8.09E-03	4.33E-03	8.80E-03	100.0	0.0	68.9	13.9	14.4	60.1	11.5
Black Duck Pond	4.55	206.16	2.98E-03	5.54E-04	0	2.43E-03	3.80E-03	0	3.90E-03	5.8	94.2	38.9	15.1	70.7	9.1	5.1
Black Point Lake	15.20	517.26	2.06E-03	8.47E-04	3.37E-04	1.21E-03	0	0	2.11E-05	32.3	67.7	14.3	96.8	0.2	0.1	2.9
Chocolate Lake	8.72	298.81	6.36E-03	1.61E-03	9.61E-04	4.75E-03	3.55E-03	1.44E-03	2.07E-03	3.5	96.5	35.3	60.1	14.1	14.3	11.5
Cranberry Lake	11.23	90.89	9.97E-03	0	0	9.97E-03	9.79E-03	3.46E-03	9.80E-03	100.0	0.0	72.9	14.9	2.7	68.1	14.3
Dent's Punch Bowl	0.97	9.82	4.46E-03	0	0	4.46E-03	4.47E-03	7.21E-04	5.92E-03	61.9	38.1	70.0	19.0	11.8	63.9	5.3
Desaid Lake	7.42	35.39	2.15E-03	0	0	2.15E-03	1.92E-03	0	0	100.0	0.0	13.7	58.6	36.7	0.0	4.7
Drain Lake	17.68	282.24	3.40E-03	1.81E-03	0	1.60E-03	1.90E-05	0	5.68E-05	100.0	0.0	21.2	94.9	0.0	0.0	5.1
Duck Lake	9.47	146.23	4.83E-03	0	0	4.83E-03	2.36E-03	1.16E-03	1.18E-03	100.0	0.0	30.8	68.7	12.9	13.0	5.4
Fenerty Lake	64.68	2300.54	1.50E-03	0	0	1.50E-03	7.05E-05	1.78E-05	1.38E-04	100.0	0.0	14.2	97.9	0.2	0.0	1.9
First Chain Lake	19.56	222.89	4.53E-03	2.16E-03	1.29E-03	2.38E-03	1.54E-03	7.32E-04	8.96E-04	0.0	100.0	24.1	71.5	18.3	0.8	9.4
First Lake	82.70	338.57	7.89E-03	0	0	7.89E-03	7.55E-03	2.71E-03	8.21E-03	100.0	0.0	61.5	18.9	12.6	55.3	13.2
Frenchman Lake	6.75	95.08	5.73E-03	0	0	5.73E-03	5.76E-03	3.67E-03	8.97E-03	100.0	0.0	61.0	0.0	89.7	0.0	10.3
Frog Lake	5.07	106.76	8.19E-03	0	0	8.19E-03	8.06E-03	2.00E-03	6.40E-03	17.2	82.8	59.8	35.9	1.2	53.1	9.8
Governor Lake	37.72	711.65	3.38E-03	2.59E-04	0	3.12E-03	3.18E-03	1.02E-03	3.47E-03	45.3	54.7	33.8	56.7	28.3	9.9	5.1

Lake Name	Lake Area (ha)	Watershed Area (ha)	Road Density (m/m^2)	Expressway Density (m/m^2)	Ramp Density (m/m^2)	Road Density excluding expressways (m/m^2)	Salted Ice Route Density (m/m^2)	Sidewalk (maintained) density (m/m^2)	Stormwater Pipe Density (m/m^2)	Surficial Geology Ground Moraine (%)	Surficial Geology Rock (%)	Urban Landcover (%)	Rural Land Use (%)	Commercial Land Use (%)	(Sub)urban Residential Land Use (%)	Estimated Landcover Roads (%)
Grand Lake	1882.76	36916.87	1.48E-03	1.88E-04	4.90E-05	1.29E-03	4.94E-04	1.14E-04	3.30E-04	80.5	12.1	13.5	87.3	7.6	3.0	2.0
Half Mile Lake	6.31	146.17	5.82E-03	1.57E-03	0	4.24E-03	4.24E-03	3.71E-04	6.13E-03	88.3	11.7	44.2	61.2	2.6	29.2	7.1
Hubley Big Lake	257.98	3799.22	1.51E-03	3.04E-04	6.34E-05	1.21E-03	0	0	1.01E-05	69.1	27.4	11.0	97.5	0.4	0.0	2.0
Kearney Lake	59.82	2741.96	2.09E-03	4.54E-04	1.75E-04	1.64E-03	1.28E-03	7.84E-04	1.42E-03	11.6	88.4	20.3	83.1	3.0	10.9	3.0
Kidston Lake	9.85	56.83	0	0	0	0	0	0	3.54E-05	76.4	23.6	6.6	99.4	0.0	0.6	0.0
Kinsac Lake	168.14	11598.5	1.19E-03	0	0	1.19E-03	3.03E-04	4.42E-05	1.11E-04	94.7	1.5	12.4	96.5	1.0	1.0	1.4
Lake Banook	41.47	3454.50	6.02E-03	1.07E-03	6.31E-04	4.95E-03	4.08E-03	1.95E-03	4.53E-03	96.2	3.8	47.0	36.7	22.5	31.9	8.9
Lake Charles	141.43	2084.05	4.17E-03	9.62E-04	4.67E-04	3.20E-03	2.54E-03	7.30E-04	2.11E-03	98.1	1.9	35.1	52.1	9.9	32.0	6.0
Lake Echo	212.20	12096.50	3.85E-04	2.14E-05	2.42E-06	3.64E-04	0	0	0	73.7	22.5	9.4	94.6	5.0	0.0	0.4
Lake Fletcher	100.62	14137.36	2.43E-03	4.92E-04	1.28E-04	1.93E-03	1.04E-03	2.62E-04	7.66E-04	78.1	20.1	21.2	71.4	19.0	6.2	3.4
Lake MicMac	104.08	3244.12	5.79E-03	1.13E-03	6.32E-04	4.66E-03	3.78E-03	1.76E-03	4.25E-03	96.0	4.0	45.5	38.1	23.5	30.0	8.5
Lake Thomas	112.89	12549.35	2.40E-03	5.55E-04	1.44E-04	1.84E-03	1.07E-03	2.52E-04	8.27E-04	80.1	17.8	21.6	68.0	21.5	7.1	3.4
Lake William	302.19	7420.88	2.89E-03	5.60E-04	1.68E-04	2.33E-03	1.68E-03	3.82E-04	1.29E-03	85.1	13.4	25.1	62.5	20.8	12.3	4.4
Lisle Lake	5.38	803.85	3.06E-03	0	0	3.06E-03	8.77E-05	5.09E-05	3.02E-04	100.0	0.0	21.7	95.6	0.3	0.0	4.1
Long Lake	166.17	1570.21	1.87E-03	2.59E-04	1.30E-04	1.61E-03	1.20E-03	1.94E-04	8.51E-04	66.4	23.6	15.1	82.9	12.4	1.4	3.3
Long Pond	6.31	184.70	1.54E-03	0	0	1.54E-03	1.43E-03	0	6.65E-04	0.0	100.0	15.0	83.9	0.8	12.9	2.4
Loon Lake	76.63	377.80	5.52E-03	5.87E-04	0	4.94E-03	4.49E-03	1.62E-03	3.23E-03	100.0	0.0	41.2	17.4	9.3	63.7	9.6
Lovett Lake	8.66	295.67	3.49E-03	3.86E-04	0	3.10E-03	4.06E-03	1.46E-03	4.13E-03	7.2	92.8	43.0	23.0	55.6	15.6	5.8
McCabe Lake	166.78	7909.59	1.35E-03	1.76E-04	2.39E-05	1.17E-03	1.34E-04	0	6.99E-05	95.4	3.1	11.9	95.1	0.2	3.1	1.6
Miller Lake	125.83	4318.04	1.09E-03	4.32E-04	5.41E-05	6.57E-04	0	0	1.43E-04	67.8	28.7	14.9	74.6	24.2	0.0	1.3
Morris Lake	162.52	1857.05	5.13E-03	1.51E-04	1.20E-04	4.98E-03	4.60E-03	2.81E-03	5.10E-03	91.0	0.0	48.0	41.0	20.2	31.4	7.4
Paper Mill Lake	21.93	3361.17	2.50E-03	5.48E-04	2.15E-04	1.95E-03	1.84E-03	8.10E-04	2.23E-03	13.9	86.1	22.3	80.3	4.8	11.4	3.5

Lake Name	Lake Area (ha)	Watershed Area (ha)	Road Density (m/m^2)	Expressway Density (m/m^2)	Ramp Density (m/m^2)	Road Density excluding expressways (m/m^2)	Salted Ice Route Density (m/m^2)	Sidewalk (maintained) density (m/m^2)	Stormwater Pipe Density (m/m^2)	Surficial Geology Ground Moraine (%)	Surficial Geology Rock (%)	Urban Landcover (%)	Rural Land Use (%)	Commercial Land Use (%)	(Sub)urban Residential Land Use (%)	Estimated Landcover Roads (%)
Powder Mill Lake	43.10	2655.69	3.43E-03	3.97E-04	8.97E-05	3.03E-03	2.08E-03	4.32E-04	1.62E-03	85.4	10.3	30.2	51.0	33.8	9.4	5.8
Red Bridge Pond	8.20	205.09	1.18E-02	6.12E-04	1.12E-04	1.12E-02	1.09E-02	4.30E-03	1.06E-02	100.0	0.0	85.1	5.2	12.3	68.0	14.5
Roaches Pond	1.04	129.37	3.15E-03	0	0	3.15E-03	3.34E-03	2.69E-04	9.98E-04	20.8	79.2	23.5	17.5	2.2	76.3	4.0
Rocky Lake	145.77	1197.05	4.11E-03	4.87E-04	1.99E-04	3.62E-03	3.02E-03	8.32E-04	3.11E-03	80.9	19.1	43.1	24.3	52.5	16.2	7.0
Russel Lake	34.82	317.57	7.31E-03	8.81E-04	7.04E-04	6.43E-03	5.37E-03	4.10E-03	7.28E-03	100.0	0.0	64.0	46.4	16.0	26.6	11.0
Sandy Lake (Bedford)	78.46	1802.05	2.19E-03	4.69E-05	6.74E-05	2.14E-03	1.16E-03	3.66E-06	5.05E-04	93.8	5.0	23.0	86.6	8.1	2.3	3.0
Sandy Lake (Glen Arbour)	35.69	199.72	8.70E-04	0	0	8.70E-04	0	0	2.72E-05	100.0	0.0	10.4	98.5	0.0	0.0	1.5
Second Lake	112.67	706.67	2.25E-03	0	0	2.25E-03	1.78E-03	2.13E-04	7.52E-04	94.1	0.0	16.4	82.3	5.7	8.2	3.8
Settle Lake	5.45	33.14	7.68E-03	0	0	7.68E-03	7.68E-03	3.93E-03	9.11E-03	100.0	0.0	65.5	20.8	0.0	68.0	11.1
Sheldrake Lake	14.12	401.98	2.95E-03	9.06E-04	1.66E-04	2.04E-03	0	0	3.99E-05	91.6	8.4	21.1	94.4	1.6	0.0	4.0
Springfield Lake	81.28	622.00	2.81E-03	0	0	2.81E-03	1.13E-04	1.82E-05	2.25E-04	100.0	0.0	21.3	95.5	0.4	0.0	4.1
Stillwater Lake	49.43	1648.07	1.23E-03	0	0	1.23E-03	2.85E-04	0	6.32E-05	98.3	0.0	10.3	98.5	0.0	0.0	1.5
Third Lake	84.80	1029.61	2.67E-03	0	0	2.67E-03	1.51E-03	1.46E-04	5.44E-04	88.9	0.0	17.8	84.9	4.6	5.9	4.6
Three Mile Pond	1.73	4822.19	1.51E-03	4.92E-05	6.71E-07	1.46E-03	4.17E-04	8.83E-06	1.58E-04	91.5	7.7	13.6	96.8	1.1	0.1	2.0
Tucker Lake	32.65	133.85	4.19E-03	0	0	4.19E-03	2.27E-03	0	1.06E-03	100.0	0.0	29.1	92.9	0.3	0.0	6.9
Whimsical Lake	2.20	40.36	9.37E-03	0	0	9.37E-03	8.85E-03	1.31E-03	8.27E-03	43.5	56.5	72.2	18.7	2.9	67.9	10.5
Williams Lake	39.82	467.17	3.55E-03	0	0	3.55E-03	2.59E-03	1.02E-03	3.02E-03	28.4	71.3	30.7	72.1	2.6	20.2	5.1
Winder Lake	3.09	27.11	2.40E-03	0	0	2.40E-03	9.37E-05	0	9.76E-05	100.0	0.0	19.5	94.1	3.7	0.0	2.3

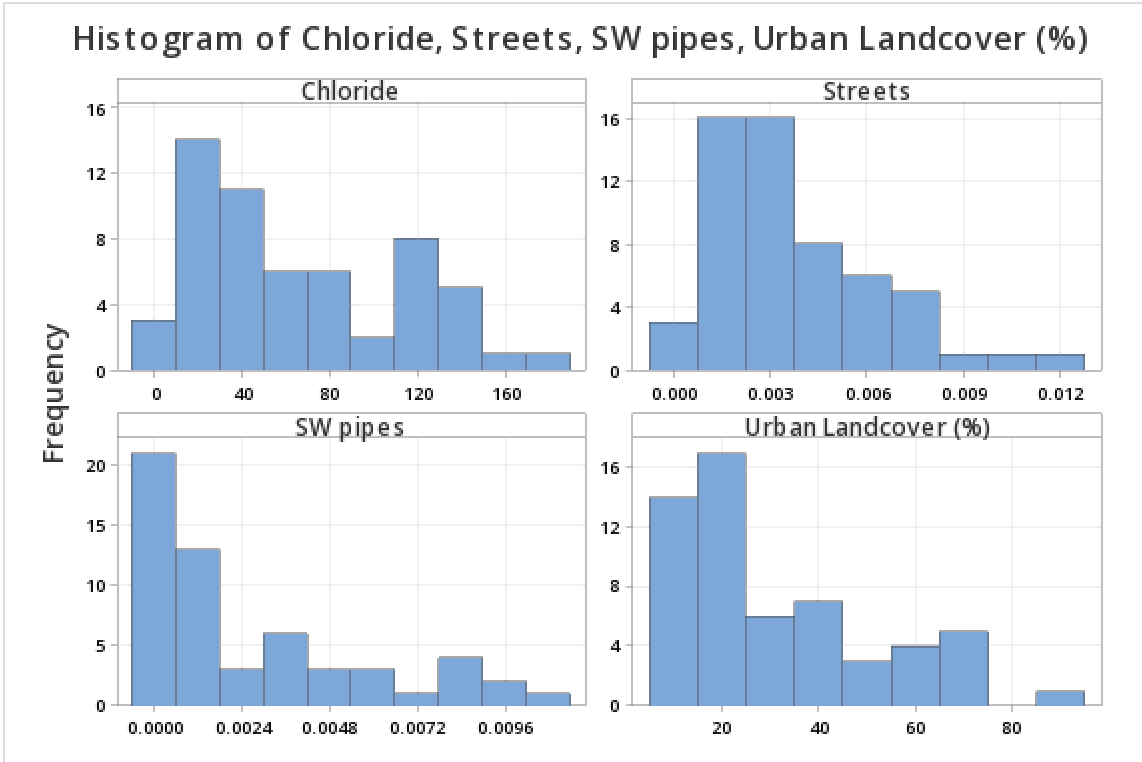


Figure A2 Histograms of mean lake [Cl^-] (Anderson Darling (AD) = 1.88) (top left) and watershed road density (AD= 1.50) (top right), watershed stormwater pipe density (AD= 4.44) (bottom left), and percent watershed coverage by urban land use (AD = 2.56) (bottom right), generated by Minitab.

Table A2 Confusion matrix produced in accuracy assessment of ‘urban landcover’ layer used to obtain ‘% urban’ for regression analysis.

		Reference		
		urban	not urban	
Classification	urban	85	38	123
	not urban	6	71	77
		91	109	200

Urban land class:

Producer accuracy: 93.4%
User accuracy: 69.1%

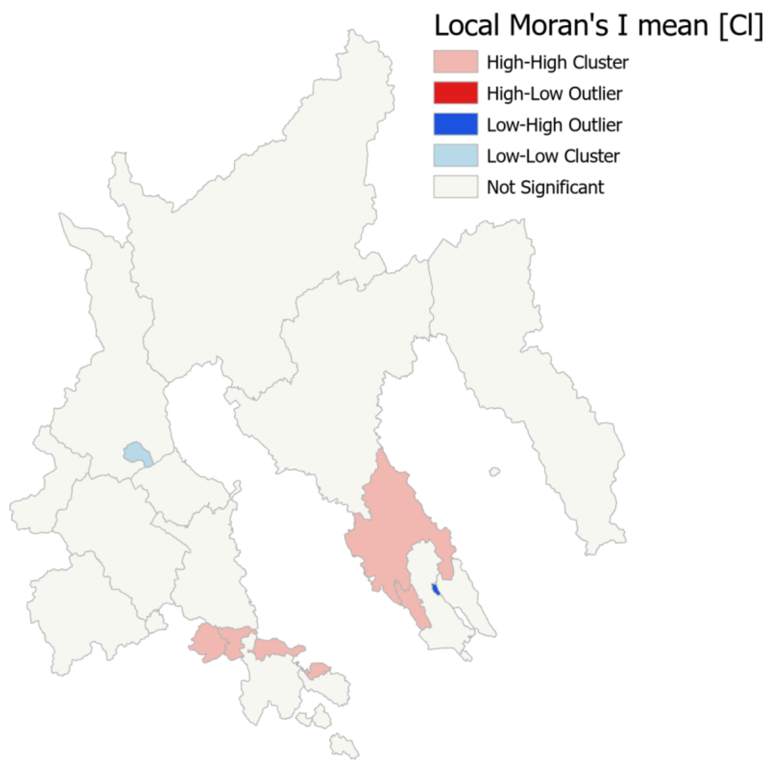


Figure A3 Results of Local Moran's I (Anselin) in ArcGIS Pro, permutations set to zero, p-value used to determine cluster (<0.05) use traditional calculation procedure. Significant high-low cluster (dark blue) represents the watershed of Bell Lake.

Table A3 Correlation matrix of select variables used in regression models, all units are (m/m^2) unless otherwise indicated.

	Roads	Expressways	Ice Routes	Ramps	SW pipes	SW outfalls (#)	Sidewalks	SW ditches	Dist. To coast (km)	WWTP (#)	Salt Yards (#)	Ground Moraine (%)	Rock (%)	Urban (%)
Roads	x													
Expressways	0.177	x												
Ice Routes	0.938	-0.032	x											
Ramps	0.199	0.748	0.017	x										
SW pipes	0.896	0.001	0.953	0.019	x									
SW outfalls (#)	0.421	-0.127	0.459	-0.078	0.476	x								
Sidewalks	0.843	0.02	0.858	0.165	0.894	0.454	x							
SW ditches	-0.06	-0.233	-0.043	-0.214	-0.056	0.023	-0.066	x						
Dist. To coast (km)	-0.3	-0.041	-0.283	-0.034	-0.258	-0.102	-0.221	-0.25	x					
WWTP (#)	-0.033	0.088	-0.041	0.085	-0.029	0.301	-0.02	-0.255	0.635	x				
Salt Yards (#)	-0.098	-0.026	-0.072	0.005	-0.075	-0.033	-0.078	-0.072	-0.026	-0.036	x			
Ground Moraine (%)	0.058	-0.228	-0.017	-0.28	0.043	0.244	0.142	0.26	0.075	0.102	-0.034	x		
Rock (%)	-0.035	0.24	0.035	0.286	-0.24	-0.215	-0.126	-0.236	-.108	-0.122	0	-0.997	x	
Urban (%)	0.927	0.059	0.941	0.087	0.964	0.418	0.854	-0.023	-0.293	-0.017	-0.11	0.046	-0.022	x
Chloride (mg/L)	0.744	0.338	0.735	0.397	0.765	0.256	0.683	-0.084	-0.308	-0.067	0.016	-0.245	0.265	0.82

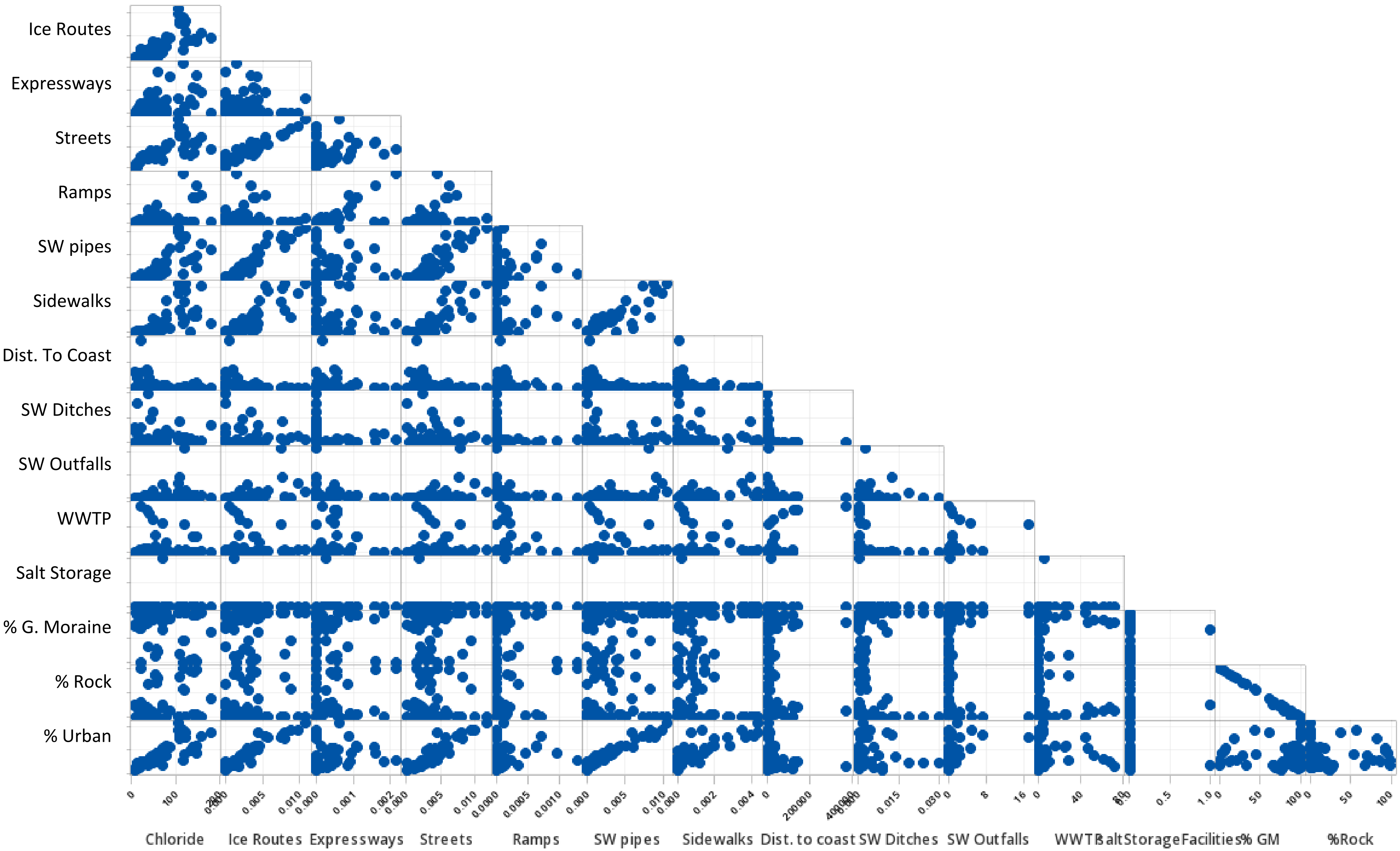


Figure A4 Scatter plot matrix of select variables used in regression analysis, as generated by Minitab.

Table A4 Results of Global Moran's I analysis performed in ArcGIS Pro on the residuals of regression models. Most models were found to have residuals that are spatially clustered, the 'Best subsets 1' model was found to have nearly spatially random residuals, and 'Best subsets 2' which added a 'distance to the coast' variable produced residuals that were spatially randomly distributed, the rest were severely clustered indicating poor model performance.

Model	Moran's Index	z-score	p-value	
'Best subsets 1'	0.108	1.663	0.096	Clustered
'Maintenance model'	0.354	4.883	<0.001	Clustered
'Expressway model'	0.313	4.495	<0.001	Clustered
'Roads only'	0.391	5.408	<0.001	Clustered
'SW pipe only'	0.368	5.054	<0.001	Clustered
'% urban only'	0.454	6.17	<0.001	Clustered
'Best subsets 2'	0.065	1.093	0.274	Random

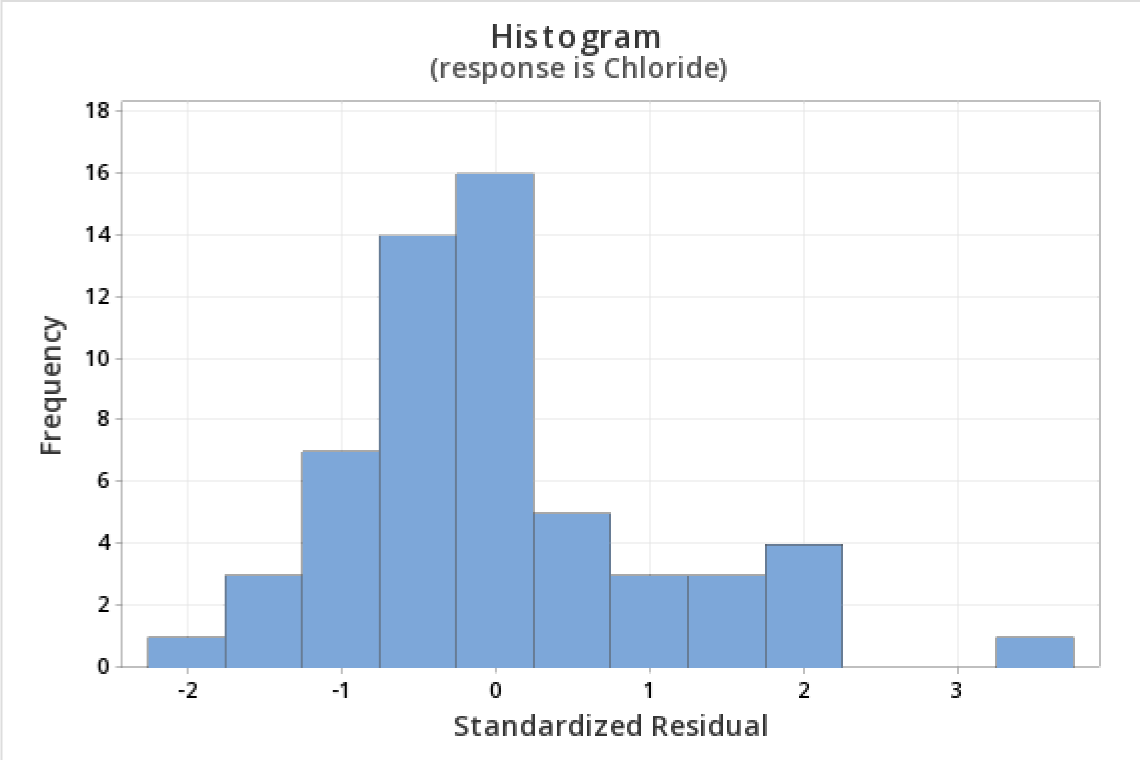


Figure A5 Histogram of standardized residuals from 'Best Subsets 2' regression, generated by Minitab.

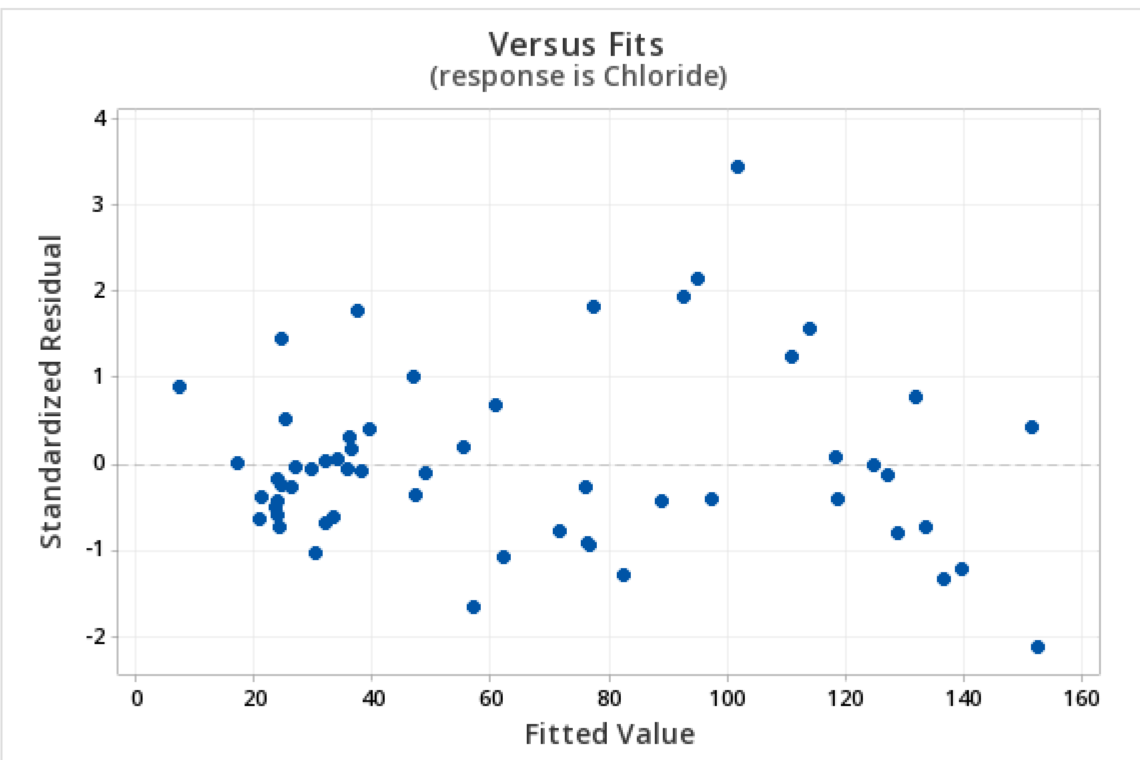


Figure A6 Residuals vs fit plot from 'Best Subsets 2' regression, generated by Minitab.

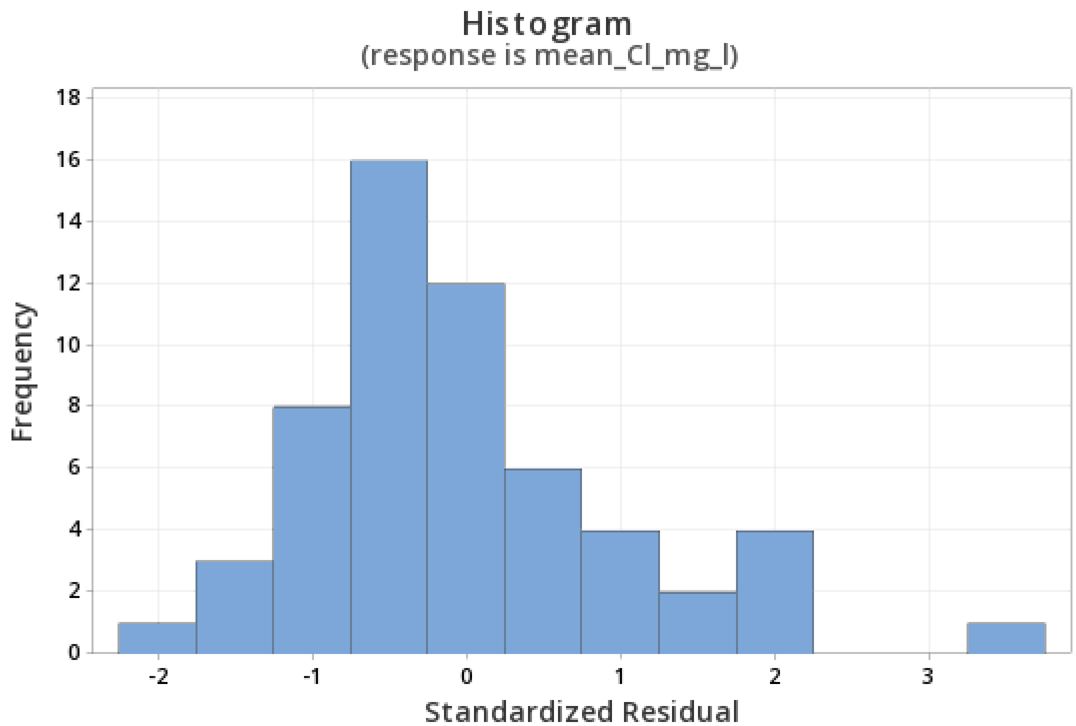


Figure A7 Histogram of standardized residuals from 'Best Subsets 1' regression, generated by Minitab.

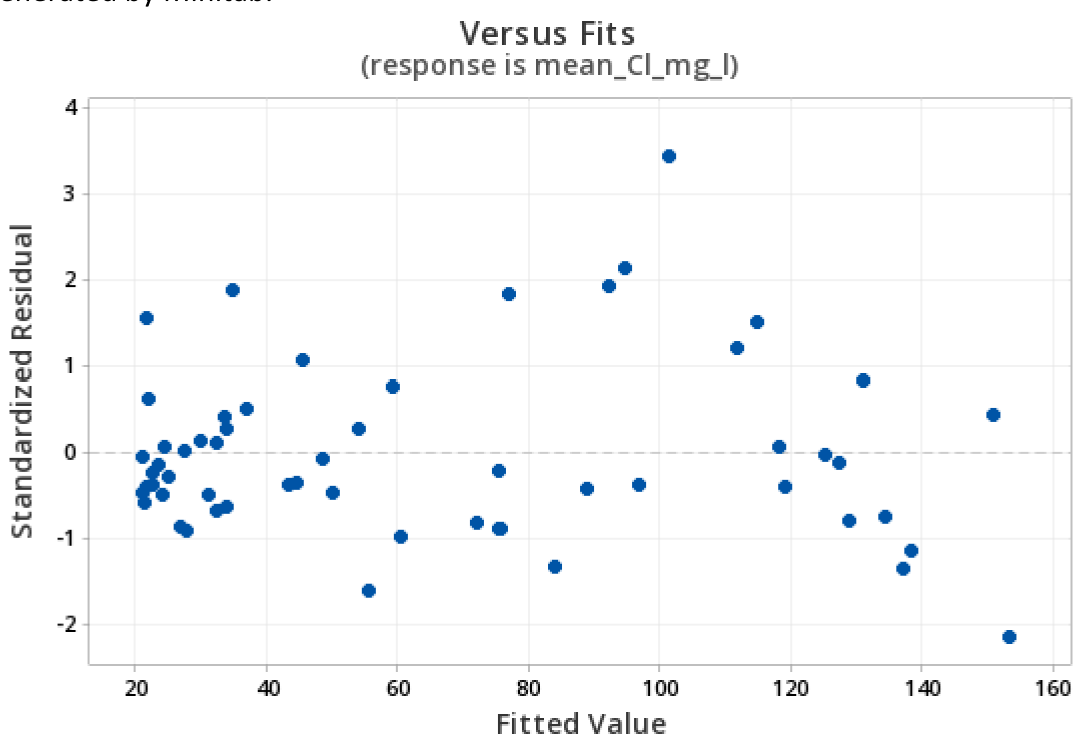


Figure A8 Residuals vs fit plot from 'Best Subsets 1' regression, generated by Minitab.

Table A6 Chloride Loading Calculations.

Lake	Weather Station	Q_o (m^3/yr) Total Outflow	Q_t (m^3/yr) Total Inflow	E_o (m3/yr) Evap. Loss	A_o (m^2) Lake Surface Area	A_d (m2) Drainage Basin Area*	P_{pti} (m^3/yr) Total Precip. Input	Q_1 (m^3/yr) Total Hydraulic Surface Runoff	Q_i (m3/yr) Upstream Hydraulic Input	P_r (m/yr) Annual Precip.	E_v (m/yr) Annual Unit Lake Evap.	R_u (m/yr) Annual Unit Runoff
Barrett Lake	Pockwock Lake	708278	759605	51327	90048	623345	136260	623345	-	1.51	0.57	1.00
Bell Lake	Westphal	274111	329398	55287	96995	154757	143339	186059	-	1.48	0.57	1.20
Bissett Lake	Shearwater A	9344440	9854841	510401	895440	7032032	1273853	8580988	-	1.42	0.57	1.22
Black Duck Pond	Halifax Citadel	2357281	2383236	25955	45536	2016048	66851	2316386	-	1.47	0.57	1.15
Black Point Lake	St Margarets Bay	5690159	5776817	86658	152032	5020531	210047	5566771	-	1.38	0.57	1.11
Chocolate Lake	Halifax Citadel	3482688	3532412	49724	87235	2905409	128070	3404342	-	1.47	0.57	1.17
Cranberry Lake	Westphal	1053512	1117516	64005	112289	796601	165941	951575	-	1.48	0.57	1.19
Dent's Punch Bowl	Halifax Citadel	114689	120237	5547	9732	88500	14288	105949	-	1.47	0.57	1.20
Desaid Lake	Shearwater A	413152	455462	42311	74229	279694	105599	349864	-	1.42	0.57	1.25
Drain Lake	Pockwock Lake	2812735	2913494	100758	176769	2646007	267487	2646007	-	1.51	0.57	1.00
Duck Lake	Pockwock Lake	1456962	1510914	53951	94652	1367687	143227	1367687	-	1.51	0.57	1.00
Fenerty Lake	Pockwock Lake	22968654	23337310	368655	646764	22358627	978683	22358627	-	1.51	0.57	1.00
First Chain Lake	Halifax Citadel	2559933	2671419	111486	195589	2039948	287145	2384274	-	1.47	0.57	1.17
First Lake	Pockwock Lake	3382570	3431907	471390	826999	2558683	1251415	2562975	-	1.51	0.57	1.00
Frenchman Lake	Westphal	1069609	1108075	38467	67485	883331	99729	1008346	-	1.48	0.57	1.14
Frog Lake	Halifax Citadel	1266984	1295874	28891	50686	1016889	74411	1221463	-	1.47	0.57	1.20
Governor Lake	Halifax Citadel	8070311	8285288	214977	377152	6739365	553698	7731590	-	1.47	0.57	1.15
Grand Lake	Halifax Stanfield	365896502	376628224	10731722	18827583	350341153	26287072	350341153	-	1.40	0.57	1.00
Half Mile Lake	Halifax Citadel	1622635	1658574	35939	63050	1398657	92564	1566010	-	1.47	0.57	1.12
Hubley Big Lake	St Margarets Bay	40712469	42182972	1470503	2579830	35412419	3564294	38618678	-	1.38	0.57	1.09
Kearney Lake	Halifax Citadel	30259195	30600190	340995	598238	26821410	878273	29721917	-	1.47	0.57	1.11
Kidston Lake	Halifax Citadel	662341	718463	56122	98460	469804	144549	573914	-	1.47	0.57	1.22

Lake	Weather Station	Q_o (m^3/yr) Total Outflow	Q_t (m^3/yr) Total Inflow	E_o (m3/yr) Evap. Loss	A_o (m^2) Lake Surface Area	A_d (m2) Drainage Basin Area*	P_{pti} (m^3/yr) Total Precip. Input	Q_l (m^3/yr) Total Hydraulic Surface Runoff	Q_i (m3/yr) Upstream Hydraulic Input	P_r (m/yr) Annual Precip.	E_v (m/yr) Annual Unit Lake Evap.	R_u (m/yr) Annual Unit Runoff
Kinsac Lake	Halifax Stanfield	115693376	116651792	958416	1681432	114304177	2347615	114304177	-	1.40	0.57	1.00
Lake Banook	Westphal	27088935	27325328	236392	414724	1689069	612879	1939100	24773349	1.48	0.57	1.15
Lake Charles	Westphal	23454242	24260408	806165	1414325	19426159	2090090	22170318	-	1.48	0.57	1.14
Lake Echo	Westphal	132539686	133749226	1209540	2122000	118842976	3135892	130613334	-	1.48	0.57	1.10
Lake Fletcher	Halifax Stanfield	137990161	138563678	573517	1006170	14873897	1404815	15364517	121794346	1.40	0.57	1.03
Lake MicMac	Westphal	24773349	25366619	593270	1040825	10559883	1538131	12101368	11727121	1.48	0.57	1.15
Lake Thomas	Halifax Stanfield	121794346	122437798	643452	1128863	50155848	1576118	52236239	68625441	1.40	0.57	1.04
Lake William	Westphal	68625441	70347928	1722487	3021907	50346400	4465774	54155032	11727121	1.48	0.57	1.08
Lisle Lake	Pockwock Lake	8035449	8066112	30663	53795	7984709	81403	7984709	-	1.51	0.57	1.00
Long Lake	Halifax Citadel	18188384	19135557	947172	1661706	14040418	2439551	16696006	-	1.47	0.57	1.19
Long Pond	Halifax Citadel	2285350	2321321	35971	63107	1783916	92648	2228674		1.47	0.57	1.25
Loon Lake	Westphal	4248617	4685405	436788	766295	3011755	1132430	3552974	-	1.48	0.57	1.18
Lovett Lake	Halifax Citadel	3382570	3431907	49337	86557	2870136	127074	3304833	-	1.47	0.57	1.15
McCabe Lake	Pockwock Lake	79001213	79951873	950660	1667824	77428121	2523751	77428121	-	1.51	0.57	1.00
Miller Lake	Halifax Stanfield	43087281	43804491	717210	1258263	41822229	1756787	42047705	-	1.40	0.57	1.01
Morris Lake	Shearwater A	21860701	22787093	926392	1625249	16945225	2312079	20475014	-	1.42	0.57	1.21
Paper Mill Lake	Halifax Citadel	37005193	37130221	125028	219347	33392335	322024	36808198	-	1.47	0.57	1.10
Powder Mill Lake	Westphal	27003115	27248772	245657	430977	26125884	636897	26611874	-	1.48	0.57	1.02
Red Bridge Pond	Westphal	2391229	2437943	46715	81955	1968991	121114	2316830	-	1.48	0.57	1.18
Roaches Pond	Halifax Citadel	1584291	1590191	5900	10351	1283353	15196	1574995	-	1.47	0.57	1.23
Rocky Lake	Westphal	12209281	13040146	830865	1457658	10512861	2154127	10886019	-	1.48	0.57	1.04
Russel Lake	Westphal	3717100	3915593	198493	348234	2827464	514620	3400973	-	1.48	0.57	1.20
Sandy Lake (Bedford)	Pockwock Lake	18003034	18450243	447210	784578	17235950	1187224	17263019	-	1.51	0.57	1.00

Lake	Weather Station	Q_o (m^3/yr) Total Outflow	Q_t (m^3/yr) Total Inflow	E_o (m3/yr) Evap. Loss	A_o (m^2) Lake Surface Area	A_d (m2) Drainage Basin Area*	P_{pti} (m^3/yr) Total Precip. Input	Q_l (m^3/yr) Total Hydraulic Surface Runoff	Q_i (m3/yr) Upstream Hydraulic Input	P_r (m/yr) Annual Precip.	E_v (m/yr) Annual Unit Lake Evap.	R_u (m/yr) Annual Unit Runoff
Sandy Lake (Glen Arbour)	Pockwock Lake	1976903	2180362	203459	356945	1640233	540129	1640233	-	1.51	0.57	1.00
Second Lake	Pockwock Lake	7002725	7644918	642193	1126655	5940064	1704854	5940064	-	1.51	0.57	1.00
Settle Lake	Westphal	381066	412129	31062	54496	276855	80534	331595	-	1.48	0.57	1.20
Sheldrake Lake	St Margarets Bay	4153712	4234224	80511	141248	3878578	195148	4039076	-	1.38	0.57	1.04
Springfield Lake	Pockwock Lake	6173875	6637188	463313	812830	5407213	1229975	5407213	-	1.51	0.57	1.00
Stillwater Lake	St Margarets Bay	16387579	16669309	281730	494262	15986436	682873	15986436	-	1.38	0.57	1.00
Third Lake	Halifax Stanfield	10148688	10632023	483335	847956	9448107	1183917	9448107	-	1.40	0.57	1.00
Three Mile Pond	Halifax Citadel	49264933	49274776	9843	17269	48205303	25353	49249423	-	1.47	0.57	1.02
Tucker Lake	Pockwock Lake	1319959	1506052	186093	326478	1012025	494027	1012025	-	1.51	0.57	1.00
Whimsical Lake	Halifax Citadel	478462	490984	12522	21969	381633	32252	458732	-	1.47	0.57	1.20
Williams Lake	Halifax Citadel	3343000	3814390	226957	398170	4275298	584553	5192408	-	1.47	0.57	1.21
Winder Lake	Westphal	304178	321786	17608	30892	240182	45652	276134	-	1.48	0.57	1.15

Table A7 Loading rates and model statistics from multiple-calibration analysis

Calibration Set	Calibrated Land Use Loading Coefficient				Train		Test	
	Rural	Comm.	Residential	Roads	RMSE	R^2	RMSE	R^2
1	3.51	13.69	234.30	579.31	2.44E+08	0.9371	3.34E+08	0.9741
2	0	0	125.89	977.72	2.55E+08	0.9661	2.73E+08	0.9682
3	5.23	0	179.86	800.87	2.7E+08	0.9449	3.2E+08	0.9859
4	5.42	28.74	139.36	895.15	1.8E+08	0.9423	6.43E+08	0.9697
5	6.35	0	241.12	661.73	1.72E+08	0.9823	4.7E+08	0.9586
6	2.66	9.66	96.16	960.84	2.67E+08	0.8883	2.89E+08	0.9837
7	0	0	150.60	964.00	2.59E+08	0.9766	2.8E+08	0.9279
8	2.33	7.13	111.12	914.71	2.37E+08	0.9520	3.15E+08	0.9767
9	1.59	18.34	79.50	932.27	2.4E+08	0.9769	3.42E+08	0.9368
10	1.53	0	98.71	1013.93	2.64E+08	0.9577	2.65E+08	0.9786
11	2.84	0	116.54	953.20	2.99E+08	0.9259	1.91E+08	0.9926
12	0	0	108.63	1033.06	2.34E+08	0.9593	3.07E+08	0.9724
13	0	0	96.53	1027.65	2.65E+08	0.9694	2.57E+08	0.9650
14	3.21	0	167.72	803.96	2.38E+08	0.9790	3.06E+08	0.8777
15	4.05	0.88	207.01	793.89	2.3E+08	0.9602	3.93E+08	0.9777
16	1.58	8.76	163.08	840.27	2.04E+08	0.9843	3.52E+08	0.8111
17	1.58	0	127.90	928.19	2.75E+08	0.9668	2.27E+08	0.9709
18	1.09	0	137.97	926.18	2.81E+08	0.9727	2.11E+08	0.9028
19	0	0	72.62	1157.54	2.35E+08	0.9381	3.43E+08	0.9765
20	2.59	0	168.68	799.65	2.41E+08	0.9747	3.05E+08	0.9491
21	0	0	114.15	992.52	2.55E+08	0.9747	2.74E+08	0.9265
22	1.39	0.11	155.80	874.86	2.99E+08	0.9624	1.62E+08	0.9823
23	1.38	7.68	96.82	955.29	2.72E+08	0.9673	2.54E+08	0.9690
24	0	6.29	134.54	925.23	2.98E+08	0.9624	1.78E+08	0.9824
25	3.16	18.07	95.44	825.75	2.19E+08	0.9777	3.93E+08	0.9555
26	4.98	0	140.39	870.02	2.88E+08	0.9474	2.76E+08	0.9860
27	1.80	11.05	82.23	931.35	2.74E+08	0.9635	2.9E+08	0.9801
28	1.04	20.99	90.23	896.65	1.97E+08	0.9812	3.94E+08	0.9479
29	0.62	0	139.49	926.48	2.08E+08	0.9841	3.44E+08	0.6992
30	3.93	7.91	113.38	808.33	2.02E+08	0.9808	4.03E+08	0.9399
31	2.60	0	156.36	840.00	2.46E+08	0.9757	2.89E+08	0.9254
32	3.03	10.82	216.90	661.24	2.47E+08	0.9776	3.14E+08	0.7289
33	1.63	14.10	57.39	963.28	2.14E+08	0.9775	4.07E+08	0.9488
34	1.94	6.94	70.93	1041.41	2.74E+08	0.9263	2.85E+08	0.9812
35	1.30	12.63	65.80	1026.53	2.5E+08	0.8930	3.23E+08	0.9789
36	1.13	6.30	162.04	882.67	2.6E+08	0.9730	2.76E+08	0.9544
37	0	2.46	125.16	965.64	2.72E+08	0.9676	2.4E+08	0.9666
38	4.90	0	115.66	909.17	2.66E+08	0.9386	3.24E+08	0.9762
39	0	42.37	79.31	1071.90	2.01E+08	0.9177	5.55E+08	0.9705
40	2.37	0	71.43	1028.56	2.55E+08	0.9405	3.16E+08	0.9765

Calibration Set	Calibrated Land Use Loading Coefficient				Train		Test	
	Rural	Comm.	Residential	Roads	RMSE	R2	RMSE	R2
42	4.74	0	146.08	887.46	2.18E+08	0.9667	3.96E+08	0.9683
43	4.39	0	100.15	942.62	2.42E+08	0.9478	3.55E+08	0.9689
44	0.59	0	199.70	843.80	1.73E+08	0.9863	4.14E+08	0.9100
45	0.97	0.78	176.72	836.38	2.7E+08	0.9727	2.61E+08	0.8333
46	0.02	0	131.54	968.44	2.85E+08	0.9665	2.07E+08	0.9649
47	0	1.48	190.09	877.07	2.46E+08	0.9788	3.34E+08	0.8874
48	3.63	0	210.64	709.13	2.46E+08	0.9787	3.09E+08	0.8679
49	4.63	0	116.93	912.90	2.71E+08	0.9504	3.06E+08	0.9756
50	2.70	0	140.34	884.92	2.76E+08	0.9489	2.31E+08	0.9836
Loading Rate Statistics								
Q1	0.62	0	96.75	840.20				
Q2	1.70	0	126.90	914.62				
Q3	3.29	8.98	162.30	964.41				
Q4	6.36	42.37	241.12	1157.54				

Table A8 Results of Q3-validation of reported loading rate ranges.

	Model Inputs						Model Outputs					
			Total Area of Land Use Type (m^2)				Estimated Chloride Contribution from Land Use Type (g/yr)					
Lake	Mean [Cl^-] (mg/L)	Calculated Chloride Loading (g/yr)	Rural	Commercial/ Industrial	(Sub)urban Residential	Roads	Rural	Commercial/ Industrial	(Sub)urban Residential	Roads	Estimated Chloride Loading (g/yr)	Estimated [Cl^-] (mg/L)
Barrett Lake	49.2	3.48E+07	5.64E+05	0	0	3.24E+04	1.81E+06	0	0	3.13E+07	3.31E+07	46.7
Bell Lake	10.5	2.88E+06	1.07E+05	0	4.74E+04	0	3.42E+05	0	7.61E+06	0	7.96E+06	29.0
Bissett Lake	125.0	1.17E+09	9.11E+05	9.45E+05	3.93E+06	7.54E+05	2.92E+06	8.08E+06	6.32E+08	7.27E+08	1.37E+09	146.5
Black Duck Pond	136.2	3.21E+08	2.93E+05	1.37E+06	1.78E+05	9.94E+04	9.39E+05	1.17E+07	2.85E+07	9.58E+07	1.37E+08	58.1
Black Point Lake	37.7	2.14E+08	4.22E+06	7.89E+03	5.75E+03	1.28E+05	1.35E+07	6.75E+04	9.24E+05	1.23E+08	1.38E+08	24.2
Chocolate Lake	148.8	5.18E+08	1.36E+06	3.20E+05	3.24E+05	2.61E+05	4.36E+06	2.74E+06	5.20E+07	2.51E+08	3.11E+08	89.2
Cranberry Lake	107.2	1.13E+08	1.12E+05	2.01E+04	5.14E+05	1.08E+05	3.59E+05	1.71E+05	8.25E+07	1.04E+08	1.87E+08	177.5
Dent's Punch Bowl	182.4	2.09E+07	1.58E+04	9.84E+03	5.31E+04	4.37E+03	5.05E+04	8.41E+04	8.53E+06	4.21E+06	1.29E+07	112.2
Desaid Lake	20.2	8.35E+06	1.65E+05	1.03E+05	0	1.34E+04	5.28E+05	8.84E+05	0	1.29E+07	1.43E+07	34.6
Drain Lake	58.8	1.65E+08	2.23E+06	6.04E+02	0	1.19E+05	7.12E+06	5.16E+03	0	1.14E+08	1.21E+08	43.2
Duck Lake	79.6	1.16E+08	9.15E+05	1.72E+05	1.74E+05	7.26E+04	2.93E+06	1.47E+06	2.79E+07	7.00E+07	1.02E+08	70.2
Fenerty Lake	14.1	3.24E+08	1.98E+07	3.17E+04	0	3.84E+05	6.35E+07	2.71E+05	0	3.70E+08	4.34E+08	18.9
First Chain Lake	117.6	3.01E+08	1.16E+06	2.96E+05	1.33E+04	1.52E+05	3.70E+06	2.53E+06	2.13E+06	1.46E+08	1.55E+08	60.4
First Lake	120.4	4.07E+08	4.56E+05	3.03E+05	1.33E+06	3.17E+05	1.46E+06	2.59E+06	2.14E+08	3.05E+08	5.23E+08	154.7
Frenchman Lake	124.7	1.33E+08	0	7.31E+05	0	8.35E+04	0	6.25E+06	0	8.05E+07	8.67E+07	81.1
Frog Lake	110.3	1.40E+08	3.27E+05	1.12E+04	4.83E+05	8.91E+04	1.05E+06	9.57E+04	7.76E+07	8.58E+07	1.65E+08	129.9
Governor Lake	120.2	9.70E+08	3.59E+06	1.79E+06	6.24E+05	3.24E+05	1.15E+07	1.53E+07	1.00E+08	3.12E+08	4.39E+08	54.4
Grand Lake	19.7	7.22E+09	2.73E+08	2.39E+07	9.54E+06	6.22E+06	8.75E+08	2.04E+08	1.53E+09	5.99E+09	8.61E+09	23.5

	Model Inputs						Model Outputs					
			Total Area of Land Use Type (m^2)				Estimated Chloride Contribution from Land Use Type (g/yr)					
Lake	Mean [Cl^-] (mg/L)	Calculated Chloride Loading (g/yr)	Rural	Commercial/ Industrial	(Sub)urban Residential	Roads	Rural	Commercial/ Industrial	(Sub)urban Residential	Roads	Estimated Chloride Loading (g/yr)	Estimated [Cl^-] (mg/L)
Half Mile Lake	88.5	1.44E+08	7.79E+05	3.27E+04	3.72E+05	8.99E+04	2.49E+06	2.79E+05	5.97E+07	8.67E+07	1.49E+08	91.9
Hubley Big Lake	16.4	6.69E+08	3.00E+07	1.38E+05	5.75E+03	6.29E+05	9.59E+07	1.18E+06	9.24E+05	6.06E+08	7.04E+08	17.3
Kearney Lake	53.9	1.63E+09	1.96E+07	7.00E+05	2.58E+06	7.16E+05	6.29E+07	5.98E+06	4.15E+08	6.91E+08	1.17E+09	38.8
Kidston Lake	6.7	4.45E+06	4.67E+05	0	2.86E+03	0	1.49E+06	0	4.59E+05	0	1.95E+06	3.0
Kinsac Lake	17.6	2.03E+09	1.03E+08	1.08E+06	1.08E+06	1.52E+06	3.28E+08	9.23E+06	1.74E+08	1.47E+09	1.98E+09	17.1
Lake Banook	149.5	4.05E+09	1.02E+07	6.25E+06	8.87E+06	2.47E+06	3.26E+07	5.35E+07	1.42E+09	2.38E+09	3.89E+09	143.7
Lake Charles	55.4	1.30E+09	8.78E+06	1.66E+06	5.40E+06	1.01E+06	2.81E+07	1.42E+07	8.67E+08	9.74E+08	1.88E+09	80.3
Lake Echo	6.7	8.86E+08	1.07E+08	5.70E+06	0	4.44E+05	3.44E+08	4.87E+07	0	4.28E+08	8.21E+08	6.2
Lake Fletcher	34.8	4.80E+09	8.38E+07	2.23E+07	7.30E+06	4.05E+06	2.68E+08	1.90E+08	1.17E+09	3.90E+09	5.53E+09	40.1
Lake MicMac	139.3	3.45E+09	1.00E+07	6.16E+06	7.87E+06	2.24E+06	3.20E+07	5.27E+07	1.26E+09	2.16E+09	3.50E+09	141.4
Lake Thomas	36.3	4.42E+09	7.01E+07	2.21E+07	7.30E+06	3.55E+06	2.24E+08	1.89E+08	1.17E+09	3.42E+09	5.01E+09	41.1
Lake William	39.2	2.69E+09	3.70E+07	1.23E+07	7.29E+06	2.60E+06	1.19E+08	1.06E+08	1.17E+09	2.51E+09	3.90E+09	56.9
Lisle Lake	26.3	2.11E+08	6.72E+06	2.06E+04	0	2.89E+05	2.15E+07	1.76E+05	0	2.79E+08	3.00E+08	37.4
Long Lake	71.2	1.30E+09	1.10E+07	1.64E+06	1.92E+05	4.39E+05	3.52E+07	1.40E+07	3.08E+07	4.24E+08	5.04E+08	27.7
Long Pond	20.3	4.64E+07	1.42E+06	1.30E+04	2.19E+05	4.12E+04	4.55E+06	1.11E+05	3.51E+07	3.97E+07	7.95E+07	34.8
Loon Lake	77.5	3.29E+08	4.63E+05	2.47E+05	1.69E+06	2.54E+05	1.48E+06	2.11E+06	2.72E+08	2.45E+08	5.21E+08	122.6
Lovett Lake	143.3	4.85E+08	6.20E+05	1.50E+06	4.21E+05	1.55E+05	1.98E+06	1.28E+07	6.76E+07	1.49E+08	2.32E+08	68.5
McCabe Lake	12.8	1.01E+09	6.91E+07	1.35E+05	2.26E+06	1.16E+06	2.21E+08	1.16E+06	3.63E+08	1.12E+09	1.70E+09	21.6
Miller Lake	19.4	8.36E+08	2.80E+07	9.08E+06	0	4.80E+05	8.97E+07	7.76E+07	0	4.63E+08	6.30E+08	14.6
Morris Lake	79.2	1.73E+09	6.18E+06	3.05E+06	4.73E+06	1.11E+06	1.98E+07	2.60E+07	7.59E+08	1.07E+09	1.88E+09	86.0
Paper Mill Lake	53.4	1.98E+09	2.34E+07	1.39E+06	3.33E+06	1.01E+06	7.49E+07	1.19E+07	5.35E+08	9.78E+08	1.60E+09	43.2
Powder Mill Lake	46.8	1.26E+09	1.01E+07	6.69E+06	1.87E+06	1.15E+06	3.23E+07	5.72E+07	3.00E+08	1.10E+09	1.49E+09	55.3

			Model Inputs				Model Outputs					
			Total Area of Land Use Type (m^2)				Estimated Chloride Contribution from Land Use Type (g/yr)					
Lake	Mean $[Cl^-]$ (mg/L)	Calculated Chloride Loading (g/yr)	Rural	Commercial/ Industrial	(Sub)urban Residential	Roads	Rural	Commercial/ Industrial	(Sub)urban Residential	Roads	Estimated Chloride Loading (g/yr)	Estimated $[Cl^-]$ (mg/L)
Roaches Pond	60.5	9.59E+07	2.19E+05	2.76E+04	9.56E+05	5.06E+04	7.00E+05	2.36E+05	1.54E+08	4.88E+07	2.03E+08	128.3
Rocky Lake	70.5	8.60E+08	2.11E+06	4.55E+06	1.41E+06	6.09E+05	6.75E+06	3.89E+07	2.26E+08	5.87E+08	8.58E+08	70.3
Russel Lake	160.9	5.98E+08	1.18E+06	4.06E+05	6.76E+05	2.80E+05	3.78E+06	3.47E+06	1.09E+08	2.70E+08	3.85E+08	103.7
Sandy Lake (Bedford)	35.5	6.39E+08	1.47E+07	1.37E+06	3.93E+05	5.10E+05	4.72E+07	1.17E+07	6.31E+07	4.92E+08	6.14E+08	34.1
Sandy Lake (Glen Arbour)	7.7	1.52E+07	1.60E+06	0	0	2.47E+04	5.12E+06	0	0	2.38E+07	2.89E+07	14.6
Second Lake	33.5	2.35E+08	4.62E+06	3.20E+05	4.59E+05	2.14E+05	1.48E+07	2.73E+06	7.36E+07	2.06E+08	2.98E+08	42.5
Settle Lake	111.0	4.23E+07	5.49E+04	0	1.79E+05	2.94E+04	1.76E+05	0	2.88E+07	2.83E+07	5.73E+07	150.3
Sheldrake Lake	40.7	1.69E+08	3.29E+06	5.61E+04	0	1.41E+05	1.05E+07	4.79E+05	0	1.36E+08	1.47E+08	35.3
Springfield Lake	20.3	1.25E+08	5.06E+06	2.06E+04	0	2.16E+05	1.62E+07	1.76E+05	0	2.08E+08	2.24E+08	36.3
Stillwater Lake	12.4	2.03E+08	1.48E+07	2.93E+02	0	2.23E+05	4.72E+07	2.50E+03	0	2.15E+08	2.62E+08	16.0
Third Lake	28.3	2.87E+08	6.65E+06	3.63E+05	4.60E+05	3.60E+05	2.13E+07	3.10E+06	7.39E+07	3.47E+08	4.45E+08	43.9
Three Mile Pond	18.9	9.33E+08	4.13E+07	4.89E+05	2.39E+04	8.61E+05	1.32E+08	4.18E+06	3.84E+06	8.30E+08	9.70E+08	19.7
Tucker Lake	43.7	5.77E+07	8.90E+05	2.64E+03	0	6.57E+04	2.85E+06	2.26E+04	0	6.33E+07	6.62E+07	50.1
Whimsical Lake	117.7	5.63E+07	6.60E+04	1.02E+04	2.39E+05	3.70E+04	2.11E+05	8.68E+04	3.85E+07	3.56E+07	7.44E+07	155.4
Williams Lake	55.3	1.85E+08	2.86E+06	1.03E+05	7.99E+05	2.03E+05	9.15E+06	8.84E+05	1.28E+08	1.96E+08	3.34E+08	100.0
Winder Lake	37.4	1.14E+07	2.41E+05	9.40E+03	0	5.85E+03	7.73E+05	8.04E+04	0	5.64E+06	6.49E+06	21.3

Model Performance		
RMSE	3.04E+08	
R^2	*0.9821*	*0.5709*