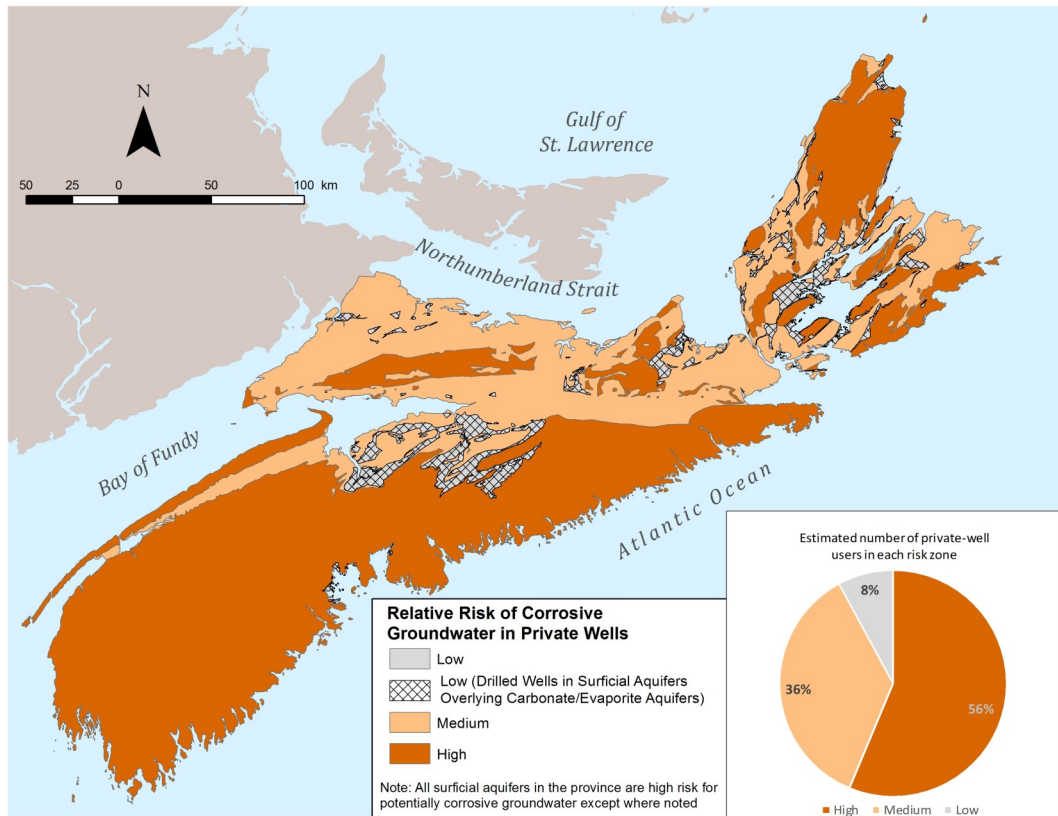


# Potential Corrosivity of Groundwater in Nova Scotia and its Association with Lead in Private Well Water

G. W. Kennedy

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# Potential Corrosivity of Groundwater in Nova Scotia and its Association with Lead in Private Well Water

*G. W. Kennedy*

## Abstract

Lead in drinking water is associated with a range of adverse health effects, including impaired neurological function. The content of lead in plumbing materials has been restricted over the last 30+ years, but issues with respect to lead exposure from drinking water supplies persist, mainly due to the presence of lead in older plumbing materials. Although municipal drinking water systems in Nova Scotia are risk-managed for lead exposure, private well water supplies account for 42% of Nova Scotia's domestic water supplies and are not regulated, and therefore lead exposure in private wells is a significant public health concern.

The potential corrosivity of groundwater was characterized for seven major aquifer types across Nova Scotia using available groundwater chemistry data, and the chloride-to-sulphate mass ratio and Langelier Saturation indices, to identify areas where there may be a greater likelihood of lead in private well water supplies. Crystalline rock type bedrock aquifers, especially plutonic aquifers, and most of the province's surficial aquifers, showed a high potential for corrosive groundwater, whereas water well chemistry data from carbonate/evaporite aquifers and surficial aquifers in contact with these aquifers showed a lower potential for corrosive groundwater. It is estimated that approximately 56%, or 111,100, of the province's private well users rely on aquifers associated with a high potential for corrosive groundwater.

Analysis of lead in well water data showed that the percentage of lead exceedances of the Health Canada maximum acceptable concentration (MAC) in well water for each of the seven major aquifer types assessed in the study followed a similar trend as the percentage of sample results with a very high combined index for potential corrosivity for each of the aquifer types. In general, aquifer types associated with a higher potential for corrosive groundwater were associated with a higher likelihood of lead concentrations in well water exceeding acceptable levels. Lead exceedances were predominantly attributed to leaching of lead from plumbing materials rather than from naturally occurring sources.

A relative risk map of the potential corrosivity of groundwater was produced to communicate risk to private well owners and to highlight the importance of routine water testing to assess the risk of lead in private well water supplies.

## Introduction

Various private well surveys have attributed elevated levels of lead in drinking water to corrosive groundwater that leaches lead and other metals from plumbing materials (Maessen et al., 1985; Swistock et al., 2009; Pieper et al., 2015a; Sweeney et al., 2017; Pieper et al., 2018a). Waterborne lead is associated with adverse health effects, such as impaired neurological function, and Health Canada has recently lowered the maximum acceptable concentration of lead from 10 µg/L to 5 µg/L in drinking water (Health Canada, 2019). Although municipal water systems in Nova Scotia are required to implement corrosion control programs if their water is corrosive, private well water supplies are not regulated with respect to water quality and treatment, and therefore these water users pose a challenge for public health protection. Increased awareness about the corrosivity of groundwater is viewed as one approach to manage the risk of lead in private well drinking water supplies (Belitz et al., 2016). A cost-benefit analysis reported by the World Health Organization (2010) estimates that for every US \$1 spent to reduce lead hazards in the United States, there is a benefit of US \$17-220.

## Historic Uses of Lead

Historically, lead was used in a variety of applications due to its low melting point, malleability and resistance to corrosion. Lead was commonly used as a pigment (lead chromate) and drying agent in paints, and as an additive (tetraethyl lead) in gasolines to boost engine performance and fuel economy. Lead was also commonly present in water service line pipes and brass and bronze fittings until the National Plumbing Code of Canada (NPC) was amended in 1975 to limit the amount of lead in these components (Health Canada, 2019). Lead, however, continued to be present in solder for new plumbing and repairs of public water systems until the NPC was amended again in 1986 to require that “lead-free” materials (<8% lead for pipes and pipe fittings and <0.2% lead for flux and solder) be used in public systems. The ban was extended to any new drinking water system plumbing or repair work under the code in 1990. The NPC was amended again in November of 2013 to limit the average amount of lead in plumbing materials and components to less than 0.25% (National Research Council of Canada, 2013).

Following reductions in the amount of lead in paints and gasolines, drinking water became the primary source of exposure of lead in human populations. Although the amount of lead is restricted in new plumbing materials, older materials continue to be a source of lead exposure as homeowners are not required to update their plumbing to the current code.

## Health Effects and Exposure

Lead is a heavy metal that is persistent and bioaccumulative in humans, and has been associated with various adverse neurological impacts. Lead has also been classed by the International Agency for Research on Cancer as a probable human carcinogen (International Agency for Research on Cancer, 2006). Common pathways of lead exposure include air, water, soil and food, but the most prevalent pathway for lead exposure in humans is food and drinking water (Health Canada, 2019). Lead in drinking water does not have a smell or taste but can be associated with other parameters (e.g. pH) or effects that are noticeable in drinking water supply systems, such as a metallic taste, blue-green stains on plumbing fixtures or obvious signs of corrosion of plumbing components (Pieper et al., 2015a). In addition to the presence of lead in water system components, a person’s exposure to lead from private well drinking water can depend on such factors as water usage patterns, the configuration of the plumbing system (e.g. length of distribution line from well to house), contact time of the water with the plumbing, and water temperature and chemistry (Cardew, 2003; Lytle and Schock, 1996; Ngueta et al., 2014; Deshommes et al., 2016). Groundwater chemistry can be affected by factors such as seasonality (e.g. water temperature), groundwater travel time, groundwater interactions with minerals in the host aquifer, and anthropogenic influences on groundwater chemistry, such as inputs of salts from road de-icing (e.g. Sun et al., 2015; Pieper et al., 2018b).

The maximum acceptable concentration (MAC) for lead in drinking water was lowered from 50 µg/L to 10 µg/L in 1992 (Health Canada, 1992), and more recently from 10 µg/L to 5 µg/L in March of 2019 (Health Canada, 2019). Health Canada (2019) recommends, however, that lead in drinking water be reduced to levels as low as reasonably achievable because there is no minimum threshold below which no adverse neurodevelopment effects are observed. Even at low blood lead levels (e.g. <5 µg/dL), exposure to lead has been associated with neurodevelopmental, neurodegenerative, cardiovascular, renal, reproductive, and developmental impacts in both children and adults (National Toxicology Program, 2012). Vulnerable populations, such as fetuses and infants, are especially susceptible to the toxic effects of low levels of lead, which include a decrease in cognitive performance and an increase in attention-related behavioural issues (National Toxicology Program, 2012). There is also evidence that chronic, low-dose exposure to lead in older adults can contribute to cognitive decline (Weuve et al., 2009). Health Canada (2019), therefore, recommends that exposure reduction efforts be targeted at these vulnerable populations.

## Lead in Private Well Drinking Water Supplies in Nova Scotia

Lead exposures from municipal water systems have gained significant attention in recent years (e.g. Washington, DC, and Flint, Michigan). Various studies have been conducted in Nova Scotia on lead in distribution systems, including the impact of service line replacement on lead levels (e.g. Camara et al., 2013) and the effectiveness of corrosion control practices (e.g. Gagnon and Doubrough, 2014; Trueman et al., 2018). In response to proposed guidance from Health Canada regarding sampling protocols for lead in distribution systems (Health Canada, 2016), the Province of Nova Scotia recently commissioned research on the effect of sampling protocols on lead levels measured at the tap in municipal water systems (Dalhousie Centre for Water Resource Studies, personal communication).

Lead exposure from private wells, however, is not well understood and remains a significant public health concern. A 1985 study on leaching of heavy metals from plumbing materials in Nova Scotia indicated that 20% of private well water supplies (n=55) sampled as a 'first draw' exceeded 50 µg/L (Maessen et al., 1985), and more recently Sweeney et al. (2017) found 2.7% of private well users across Nova Scotia had concentrations of lead exceeding 10 µg/L in 'flushed' (allowing water to run for 10 minutes in this study) water samples. While efforts are on-going to mitigate lead exposure associated with the municipal water systems in Nova Scotia through lead service line replacement and corrosion control programs; private well users, which account for about 42% of the province's domestic water supplies (Kennedy and Polegato, 2017), are not regulated when it comes to drinking water quality, and few of these well users (i.e. <15%) are following the province's recommended guidance for private well water quality monitoring (Chappells et al., 2014).

A recent study published by the Atlantic Path research group (a long-term cohort study of chronic disease in Atlantic Canada), found that the highest rates of lead exceedances in Nova Scotia drinking water were associated with water samples collected from private wells (Sweeney et al., 2017). Sweeney et al. (2017) reported that the geometric mean for lead concentrations in all flushed drinking water samples collected as part of the project was 0.35 µg/L. Overall, 2.2% (n=59) of all water samples had lead levels exceeding the previous Health Canada MAC (i.e. 10 µg/L). A comparison of sources of drinking water noted significant differences in lead concentrations ( $p < 0.001$ ). Dug wells had the highest rate of lead exceedances (4.4%) compared to drilled wells (1.8%) and municipal treated water (0.7%). A significant correlation ( $p < 0.001$ ) between lead body burden (as measured in toenails) and lead in drinking water in Nova Scotia was also identified during the study (Sweeney et al., 2017).

Although the amount of lead contained in National Science Foundation (NSF) plumbing components has been reduced over the years, there is a significant legacy issue with respect to the release of lead from older household plumbing materials. Records of distribution line material are seldom available or known to homeowners. In private wells, galvanized iron and brass components in submersible pumps, pitless adaptors and piping, and galvanized iron well casings can also be a source of lead in drinking water (Environmental Protection Agency, 1994; Pieper et al., 2015a; Pieper et al., 2015b; Pieper et al., 2018a). Pieper et al. (2015b) identified three main patterns of lead occurrence in private wells: (1) lead (mostly dissolved phase) elevated in only the 'first draw' sample due to dissolution of lead from plumbing during water stagnation, (2) erratic concentration spikes of lead (mainly particulate phase with lead often adsorbed to iron oxides and oxyhydroxides) mobilized at semi-random times due to scouring of leaded scales associated with high flows or hydraulic disturbances (e.g. pump engagement to fill pressure tank), and (3) sustained detectable concentrations of lead (mainly dissolved phase) with continued water use due to corrosion of well components.

Numerous factors influence the corrosion of plumbing materials that may release lead in private wells, including the corrosivity of groundwater supplying the water system (Belitz et al., 2016). The

relationship between groundwater chemistry and corrosion processes is complex but has been associated with groundwater travel times and the mineralogy of geologic materials encountered (Pieper et al., 2015a; Belitz et al., 2016). For example, the more geochemically evolved water of deeper aquifer systems (e.g. bedrock aquifers) with longer groundwater travel times tends to have higher pH and alkalinity, which can decrease the corrosivity of groundwater to plumbing components (US Environmental Protection Agency, 2016). Groundwater in contact with carbonate rocks is also associated with higher pH and alkalinity compared to other aquifer types in Nova Scotia (Kennedy and Drage, 2009).

In low pH water, more  $H^+$  ions are available (removal of  $OH^-$  from solution) increasing the concentration of electrolytes which promotes the corrosion of metals. Lower pH water can also increase the solubility of various carbonate films (e.g.  $PbCO_3$ ), preventing them from forming a protective barrier (passivating film) on the inside of pipes (Schock, 1989). Alkalinity is mainly produced by bicarbonate and carbonate ions, and in low alkalinity water, there is limited capacity to buffer changes in pH or form passivating films that can inhibit lead release. At pH above 7, however, moderate to high alkalinities (as dissolved inorganic carbon) in groundwater may be associated with increased lead solubility due to the formation of lead-carbonate complexes (Schock, 1980). At pH above 7, lead solubility is also sensitive to orthophosphate levels (Schock, 1989); however, orthophosphate concentrations are typically low in groundwater.

While pH and alkalinity can decrease the corrosivity of groundwater, more geochemically evolved groundwater (or groundwater in contact with carbonate rocks) tends to have higher total dissolved solids (TDS), which can increase the corrosivity of groundwater (Sterrett, 2007) especially where concentrations of calcium are low. Dissolved minerals (especially chlorides and sulphates) influence the corrosion process because the greater the concentration of dissolved minerals in the water supply, the greater the electrical conductivity, which tends to accelerate corrosion processes (Sterrett, 2007). At a galvanic lead-copper couple, chloride and sulphate can become concentrated at the anode, which affects lead solubility because lead tends to form soluble complexes with chloride and insoluble precipitates with sulphate (Nguyen et al., 2011b).

## **Geogenic Sources and Geochemical Controls of Lead in Nova Scotia Groundwater**

The province can be divided into five dominant bedrock groundwater regions or aquifer types (Fig 1., Kennedy and Drage, 2009). Although naturally occurring sources of lead (i.e. lead-bearing minerals) have been identified in rocks in Nova Scotia (e.g. McCulloch, 1984; Canadian Environmental Technologies Inc., 2005), lead is not highly mobile in groundwater flow systems and, therefore, the presence of lead in well water from geogenic sources is not considered a significant concern.

Mineral occurrences of lead have been identified in association with the base of the Windsor Group, which corresponds to the carbonate/evaporite groundwater region (Fig. 1), and in the Cumberland Basin (Ryan and Boehner, 1991). The most well-known environments in Nova Scotia for lead deposits include carbonate-hosted Pb-Zn deposits, which may be related to faulting, and unconformity related, sandstone-hosted Pb-Zn-Ag deposits (Ryan and Boehner, 1991). The Gays River mine is a well-known example of a carbonate-hosted Pb-Zn deposit, whereas the Yava deposit near Loch Lomond (Fig. 1), Cape Breton Island, is an example of a Pb-Zn-Ag deposit (Ryan and Boehner, 1991). Pb-Zn anomalies have also been mapped in the Cumberland Basin along the southeast margin of the Cobequid Highlands between Mount Thom and Scotsburn (Fig. 1), particularly in the Boss Point conglomerates (Ryan, 1993).

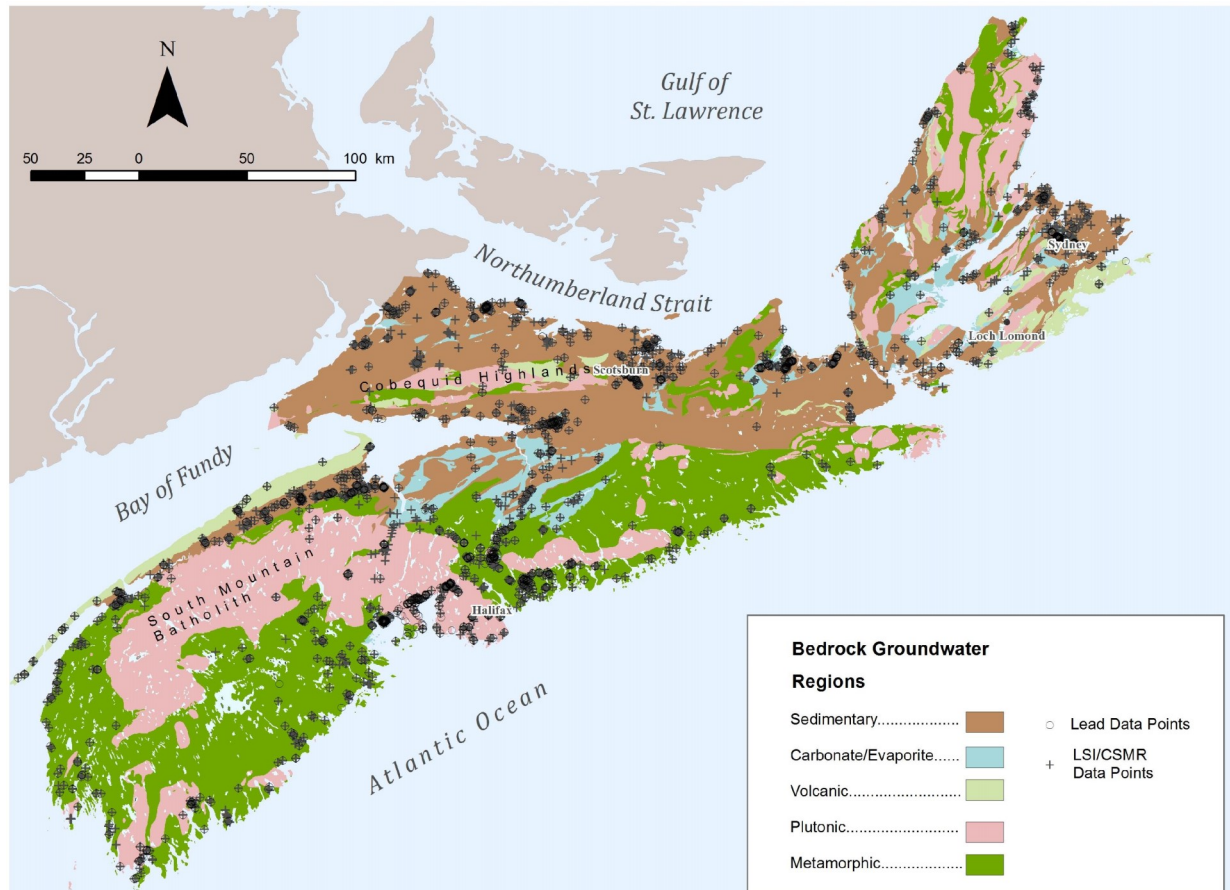
The dominant forms of lead in the environment include the  $Pb^{2+}$  and  $Pb^{4+}$  species (Health Canada, 2019). Lead is insoluble in its elemental state but can be sparingly soluble in water as lead salts, forming complexes with chloride, hydroxyl, and organics, which can increase its mobility and result in elevated concentrations of lead in well water (Woosely and Millero, 2013; Sun et al., 2015). Lead-chloride species are the most common form of lead in groundwater flow systems, although diverse species of lead (e.g.  $PbCO_3$ ,  $PbOH_2$ ) can form from the dissolution of lead oxides, carbonate, phosphate and

sulphate minerals, and desorption of lead species from organic matter and clay particles, and iron and manganese oxyhydroxides (Woosely and Millero, 2013; Sun et al., 2015). Because lead forms mobile complexes with chloride, road de-icing salt applications, salt structures (e.g. diapirs), and seawater intrusion can all mobilize naturally occurring lead. These sources of chloride (in addition to other salts such as sulphate) can also make well water more corrosive to lead contained in plumbing fixtures by influencing the corrosion process, as discussed previously. In general, many of the chemical characteristics of water that tend to be associated with corrosion of plumbing materials also tend to mobilize geogenic lead (low pH, high chloride, etc.) (Sun et al., 2015).

There are no known documented private well surveys in Nova Scotia related to concerns about naturally occurring lead in well water, although there have been several investigations that have attributed elevated lead in groundwater to geogenic sources. For example, the two background groundwater samples collected at the Yava mine site (Canadian Environmental Technologies Inc., 2005) both had lead levels exceeding 10 µg/L (up to 27 µg/L). A study on elevated lead concentrations in a school well in Scotsburn, Pictou County (Fig. 1), an area with known lead mineralization (Ryan, 1993), was suspected to be attributed to elevated levels of lead (up to 40 µg/L) in particulate that was dissolved during the sample analysis (CBCL Limited, personal communication). There are few examples of private wells contaminated by geogenic sources of lead in Canada, although a cluster of elevated lead levels in rural private wells in the City of Hamilton, Ontario, was attributed to bedrock sources of lead, which resulted in the city issuing a drinking water advisory (Richardson, 2006).

**Objectives**

The objectives of the current work are (1) to investigate spatial patterns of the potential corrosivity of groundwater and lead in well water compared to Nova Scotia’s major aquifer types, and (2) to produce a



**Figure 1.** Groundwater regions of Nova Scotia (Kennedy and Drage, 2009), and distribution of lead and LSI/CSMR datasets.

map to communicate to private well users the relative risk of having potentially corrosive well water, and hence the relative risk of having elevated levels of lead and other metals (e.g. copper) that may be leached from well and plumbing components, in their drinking water. The map will be used to help communicate to private well users the importance of testing for lead in their private well water supplies.

## Methods

The approach used to characterize the corrosivity of groundwater across Nova Scotia follows closely the approach recently published by the United States Geological Survey (USGS) for mapping the potential corrosivity of groundwater across the United States (Belitz et al., 2016). The USGS approach involved the use of two indicators of potential corrosivity, the Langelier Saturation Index (LSI) (Langelier, 1936; Larson et al., 1942) and the Potential to Promote Galvanic Corrosion (PPGC) (Nguyen et al., 2011a). The two indicators were also combined into an overall rank to characterize corrosion potential for various geographic distributions in the United States (Belitz et al., 2016).

The LSI is used to indicate whether a calcium-carbonate scale may be deposited inside pipes and other components of a distribution system, although it should be noted that there is limited evidence that calcium-carbonate coatings occur on pipes in practice (Schock, 1989). The LSI is calculated as the difference between the measured pH of the water and the theoretical pH at calcite saturation ( $\text{pH}_s$ ). Where a protective scale (or passivating film, often carbonate) is not present, lead may be more readily dissolved from pipes and plumbing components (Langelier, 1936; United States Environmental Protection Agency, 2016; Hu et al., 2012). If deposition of a carbonate scale does occur, lead may be sequestered in the scale as lead carbonates (e.g. hydrocerussite). Therefore, LSI should not be used as a direct indicator of corrosion, but rather as an indicator of the tendency for scaling to occur, which can help indicate the likelihood of corrosion of lead from water system components (Belitz et al., 2016). The LSI is not considered a reliable predictor of corrosion of plumbing components (e.g. Schock, 1989), however, the index does incorporate variables that affect the corrosivity of groundwater such as pH, calcium, TDS and alkalinity.

The Potential to Promote Galvanic Corrosion (PPGC) indicator of groundwater corrosivity is based on a three-tier classification system developed by Nguyen et al. (2011a) to assess the likelihood of galvanic corrosion of lead in water distribution systems. Galvanic corrosion is an electrochemical process that can occur when two or more dissimilar metals are in electrical contact with each other in the presence of an electrolyte. Galvanic corrosion of lead can occur when lead pipe or solder contacts other metals, such as copper or tin (Edwards and Triantafyllidou, 2007). Chloride forms soluble complexes with lead, whereas sulphate forms precipitates; therefore, groundwater having a higher chloride-to-sulphate mass ratio has a greater potential for galvanic corrosion to release lead to drinking water (Edwards and Triantafyllidou, 2007; Hu et al., 2012), especially in water with low alkalinity (Nguyen et al., 2011b). In alkaline waters, bicarbonate can compete with chloride and sulphate transport, and can also buffer changes in pH at the anode surface (Nguyen et al., 2011b).

A dataset of relevant general chemistry parameters, including alkalinity, chloride, sulphate, TDS, electrical conductivity (specific electrical conductance), pH, and calcium was extracted from the existing Nova Scotia Groundwater Chemistry Database to calculate the LSI and PPGC (n=2633). The Nova Scotia Groundwater Chemistry Database is maintained by the Nova Scotia Department of Energy and Mines, and consists mostly of non-domestic well-water sample results collected between 1954 and the present that are assumed to reflect ambient groundwater chemistry (i.e. treated well water samples and data from contaminated site investigations are excluded). This database was compiled from various federal, provincial, and municipal groundwater chemistry data sources, including water-quality monitoring data from government buildings with well water supplies, community well water surveys, and Nova Scotia Environment groundwater chemistry data from registered public drinking-water supplies, pumping tests, municipal groundwater systems, and provincial observation wells. Most of



these data are publicly available (Nova Scotia Department of Energy and Mines, 2018); however, some data (including some data used for this study; 2018 to current) are not public due to privacy considerations.

The general chemistry dataset was filtered for the LSI and PPGC input parameters to retain only the latest sampling event for each well in the dataset, and to include only data where the spatial resolution of the sample location was adequate (i.e. included only water sample locations accurate to at least the land parcel-scale), and there was adequate confidence in the aquifer type and sample type (i.e. include only raw or suspected raw water samples).

The LSI was calculated according to Equation 1, where pH was measured as part of the sample laboratory analysis and  $\text{pH}_s$  was calculated from data on alkalinity, calcium, TDS, and water temperature. Because field measurements of water temperature at the time of sampling were not available, a uniform temperature of 10°C was used in the calculation, which reflects typical ambient temperatures of Nova Scotia groundwater (Nova Scotia Environment, 2015). In practice, however, groundwater temperatures would vary seasonally and shallow wells in surficial aquifers would exhibit greater seasonal ranges with higher peak water temperatures in summer compared to drilled wells in bedrock aquifers. A regression was performed of electrical conductivity and TDS for the seven major aquifer types assessed during the study so that TDS could be estimated where only electrical conductivity data were available (n=315). Water samples were then classified as potentially corrosive ( $\text{LSI} < -0.5$ ), indeterminate ( $\text{LSI} \geq -0.5$  and  $\text{LSI} \leq 0.5$ ) or scale-forming ( $\text{LSI} > 0.5$ ) (after Belitz et al., 2016).

**Equation 1** (after Roberge, 2007):

$$\text{LSI} = \text{pH} - \text{pH}_s$$

$$\text{Where } \text{pH}_s = (9.3 + A + B) - (C + D)$$

$$A = (\log_{10} [\text{TDS}] - 1) / 10,$$

$$B = -13.12 \times \log_{10} (10^\circ\text{C} + 273) + 34.55,$$

$$C = \log_{10} [\text{Ca}^{2+} \text{ as CaCO}_3] - 0.4, \text{ and}$$

$$D = \log_{10} [\text{alkalinity as CaCO}_3].$$

The chloride-to-sulphate mass ratio (CSMR) was calculated according to Equation 2, using available chloride and sulphate concentration data. Water samples were classified as being low ( $\text{CSMR} < 0.2$ ), moderate ( $\text{CSMR} \geq 0.2$  and  $\text{CSMR} \leq 0.5$  or  $\text{CSMR} > 0.5$  and alkalinity  $\geq 50$ ), or high ( $\text{CSMR} > 0.5$  and alkalinity  $< 50$ ) according to the classification system proposed by Nyguen et al. (2011).

**Equation 2**

$$\text{CSMR} = \text{chloride (mg/L)} / \text{sulphate (mg/L)}$$

A combined ranking of the potential corrosivity of groundwater was estimated by combining the two indicators into a matrix consisting of four different categories (Fig. 2, after Belitz et al., 2016). Because lead is not always measured during water sampling for general chemistry parameters, or it is measured individually, separate extractions of the Nova Scotia Groundwater Chemistry Database were conducted for lead and the LSI/PPGC parameters to maximize the size of the two datasets. The lead data were filtered to include only lead in well water results from the past 20 years, and where the laboratory method detection limit for sample analyses was  $\leq 2$   $\mu\text{g/L}$ . Where time series data were available for a given well, only the maximum lead result was retained in the dataset to provide a conservative assessment of risk (n=1975). Similar to the LSI/PPGC dataset, lead data were used where there was adequate resolution of the sample location, and the aquifer type and sample type were known.

The locations of all water chemistry samples were plotted, and one of the province's five bedrock groundwater regions (Fig. 1) was assigned to each water sample collected from a bedrock aquifer, based on its location relative to the regions. The groundwater chemistry of surficial aquifers in contact with

carbonate/evaporite rocks tends to be strongly influenced by the underlying bedrock aquifer, and therefore water samples collected from surficial aquifers were divided into two categories: (1) water samples from drilled wells installed in surficial aquifers in contact with carbonate/evaporite rocks, and (2) all other water samples collected from wells installed in surficial aquifers. Thus, seven different major aquifer types were assigned to the water samples used in the spatial analysis.

To investigate whether certain aquifer types were associated with more corrosive groundwater, statistics and boxplots were produced for each of the seven major aquifer types with respect to LSI, CSMR, the combined index, and other parameters such as pH, alkalinity and lead. Statistical analyses were conducted using statistical software STATA® (StataCorp, LP) and ProUCL 5.0, which was developed by the United States Environmental Protection Agency (US Environmental Protection Agency, 2013). ProUCL 5.0 is designed for statistical analyses of environmental data and is adapted to handle left censored datasets with a skewed distribution, such as water chemistry (i.e. analyte concentrations may be below laboratory detection limits). Non-parametric statistical analyses were employed due to the non-normal distribution of the data (Shapiro-Wilk,  $p < 0.05$ ), with an assumed alpha of 0.05 as an indication of significance.

To compare two or more groupings of data, the non-parametric rank sum Kruskal-Wallis test, a one-way ANOVA test on ranks that extends the rank sum test to more than two groups, was applied to the data. Rank-sum tests require no assumptions about the population distribution and are resistant to outliers. The rejection of the null hypothesis (i.e. all samples originate from the same distribution) indicates that at least one sample stochastically dominates at least one other sample, meaning that a randomly drawn sample from at least one group is more likely to be larger than a randomly drawn sample from a different group. The post-hoc Dunn’s pair-wise comparison test (Benjamini-Hochberg correction) was applied following the Kruskal-Wallis test to evaluate stochastic dominance within the groupings (Dinno, 2018). Based on the combined index results for the seven major aquifer types, the potential for corrosive groundwater across Nova Scotia was characterized using a simplified risk framework, and the spatial distribution of lead in well water was compared to the interpreted risk zones. The percentage of private well users especially susceptible to corrosive groundwater was estimated by comparing the distribution of private wells in Nova Scotia (Kennedy and Polegato, 2017) to the extent of the risk zones.

It should be noted that concentrations of lead are highly sensitive to sampling technique and the Nova Scotia Groundwater Chemistry Database includes data where the recommended sampling protocols for assessing potential lead exposure were not followed (e.g. see Health Canada, 2019). Factors affecting lead concentration in tap water include stagnation time, presence of particulates, laboratory sample preparation method (filtration, acidification), temperature, volume collected, and flow rate (Schock, 1990; Schock and Lemieux, 2010; Pieper et al., 2015a). Because the data from the Nova Scotia

		PPGC Class		
		High	Moderate	Low
LSI Class	Potentially corrosive	Very High (1)	High (2)	Moderate (3)
	Indeterminate	High (2)	Moderate (3)	Low (4)
	Scale-forming	NA	Low (4)	Low (4)

**Figure 2.** Classification system for ranking the potential corrosivity of groundwater based on the Langelier Saturation Index (LSI) and potential to promote galvanic corrosion (PPGC).

Groundwater Chemistry Database were compiled from various secondary sources, sampling locations and protocols varied widely. For example, the dataset contains water samples collected directly from wells equipped with a test pump, from pressure tank sediment valves, or from locations within a distribution system, with variable flushing times prior to sampling. It is likely that most samples were collected following flushing, which tends to be associated with lower concentrations of lead in well water (e.g. Pieper et al., 2015a), and analyzed for total lead (dissolved + particulate). Assuming the age of private well distribution components and sample biases are not spatially distributed relative to the distribution of the major aquifer types, however, it is expected that the dataset will capture the dominant spatial trends with respect to the distribution of lead in Nova Scotia’s principal aquifers.

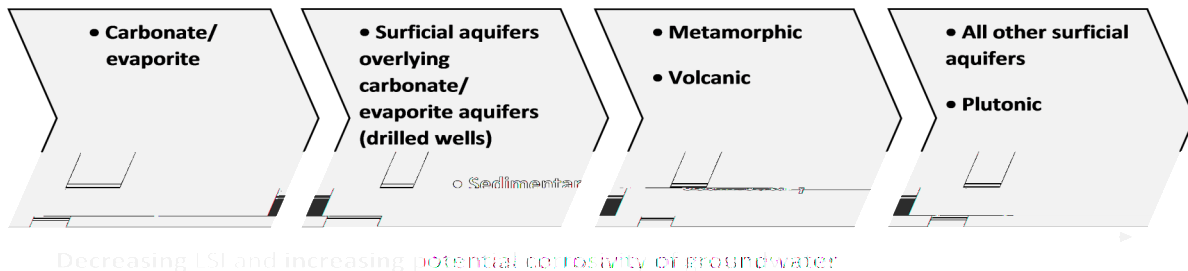
## Results & Discussion

### Total Dissolved Solids/Conductivity

The slope of the regression between electrical conductivity and TDS ranged from 0.51 to 0.69 for the seven major aquifer types assessed during the study (Table 1). Generally, higher concentrations of TDS relative to electrical conductivity are observed in carbonate/evaporite aquifers and surficial aquifers in contact with these rocks, compared to crystalline bedrock aquifers and other surficial aquifers in the province. Hem (1985) notes that the typical range of the TDS to electrical conductivity ratio in natural waters is from 0.55 to 0.75, although higher ratios are often associated with waters high in sulphate and/or chloride.

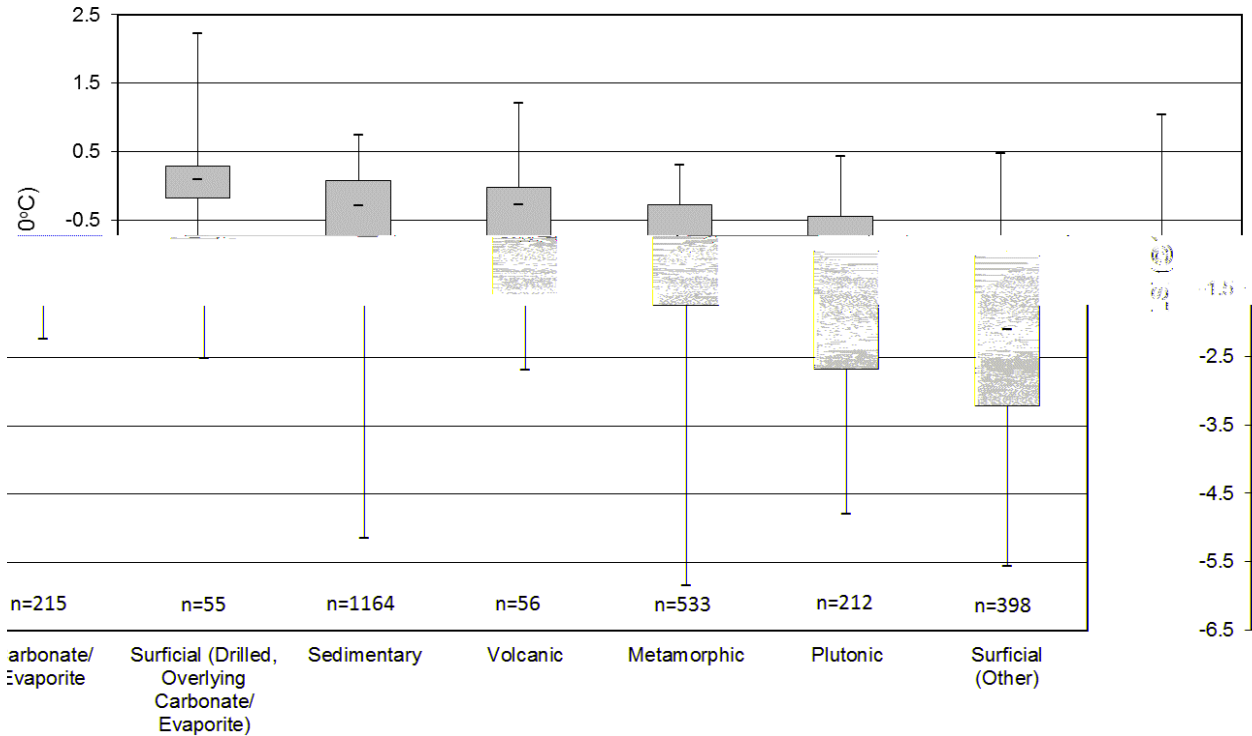
### Langelier Saturation Index

Statistical summary of the LSI shows that carbonate/evaporite (including drilled wells in overlying surficial aquifers) and sedimentary aquifers had a median LSI in the indeterminate category (>-0.5 and <0.5) (Fig. 3, Table 2), whereas bedrock aquifers in crystalline rocks and all other surficial aquifers in the province had median LSI values below -0.5 (potentially corrosive). Kruskal-Wallis analysis of the distribution of these data shows that there are statistically different populations amongst the groupings ( $p < 0.0001$ ), and implementation of the Dunn test shows that the following four groupings, arranged from highest to lowest LSI, are statistically different in terms of their medians ( $p < 0.05$ ):



**Table 1.** Summary of linear regressions between total dissolved solids and conductivity for the seven major aquifer types assessed.

Aquifer Type	Slope	Coefficient of determination ( $r^2$ )	Count (n)
Carbonate/Evaporite	0.64	0.92	288
Surficial (Drilled, Overlying Carbonate/Evaporite Aquifers)	0.69	0.90	216
Sedimentary	0.59	0.97	2295
Volcanic	0.50	0.98	102
Metamorphic	0.54	0.97	922
Plutonic	0.52	0.97	346
Surficial (other)	0.51	0.96	478
	Average: 0.57		Total: 4647

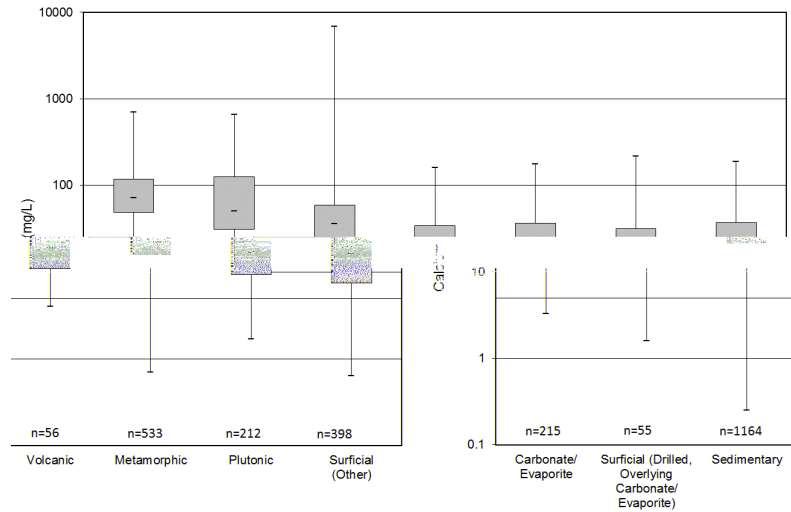


**Figure 3.** Boxplot of Langelier Saturation Index for the seven major aquifer types assessed.

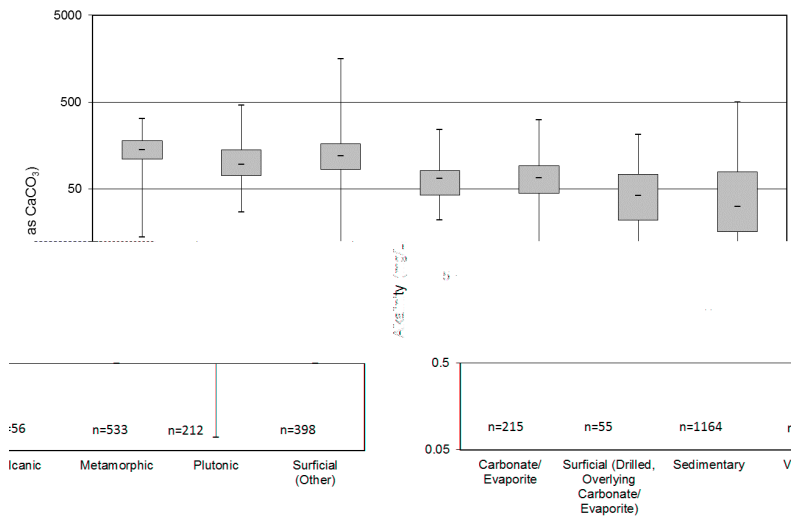
**Table 2.** Summary of medians for LSI, CSMR, alkalinity, calcium and pH for the seven major aquifer types assessed. The combined index for potential groundwater corrosivity is interpreted for each of the seven aquifer types based on the reported medians.

Aquifer Type	Medians					Combined Index
	LSI	CSMR	Alkalinity	Ca	pH	
Carbonate/Evaporite	0.09	1.93	140.00	71.70	7.90	Moderate
Surficial (Drilled, CO <sub>3</sub> )	-0.29	0.30	96.00	50.40	7.60	Moderate
Sedimentary	-0.28	2.00	120.00	35.65	7.88	Moderate
Volcanic	-0.81	2.70	65.50	19.00	7.76	High
Metamorphic	-0.98	1.73	67.00	24.00	7.60	High
Plutonic	-1.85	4.26	41.90	17.15	7.10	Very High
Surficial (Other)	-2.09	2.33	31.55	16.55	6.90	Very High

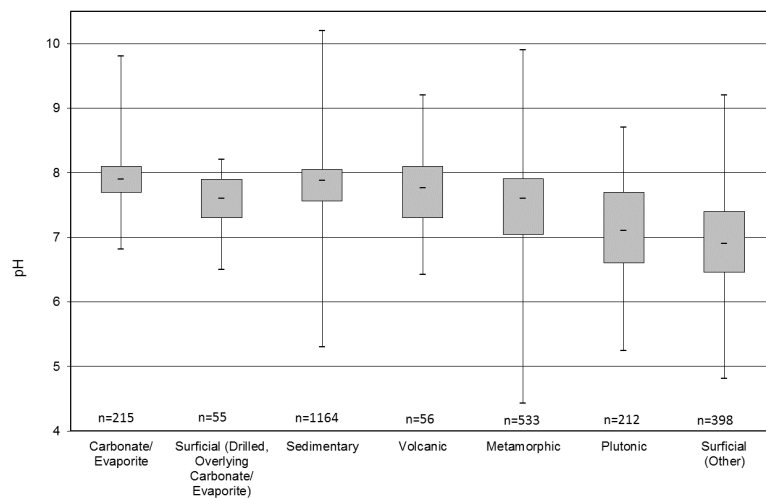
Carbonate/evaporite aquifers (e.g. limestone and gypsum rock aquifers) and drilled wells in overlying surficial aquifers tend to be associated with higher pH, calcium, sulphates, TDS, and alkalinity. Although increasing TDS can result in lower LSI values up to 500 mg/L, the LSI is more sensitive to pH, and to calcium and alkalinity concentrations (Zhang et al., 2014). Calcium, alkalinity and pH follow a similar gradient as LSI with respect to groundwater region (i.e. high calcium, alkalinity and pH for carbonate aquifers and low calcium, alkalinity and pH for crystalline aquifers and most surficial aquifers; Fig. 4-6, Table 2). The lowest values for LSI tend to be observed in surficial aquifers because shallow well water is less geochemically evolved and has had lesser opportunity for neutralization through contact with geologic materials (i.e. lower pH and alkalinity).



**Figure 4.** Boxplot of calcium (plotted on a logarithmic axis) for the seven major aquifer types assessed.



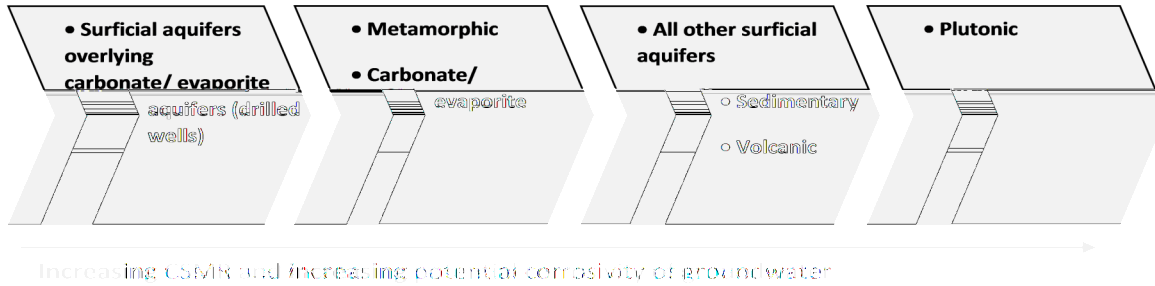
**Figure 5.** Boxplot of alkalinity (plotted on a logarithmic axis) for the seven major aquifer types assessed.



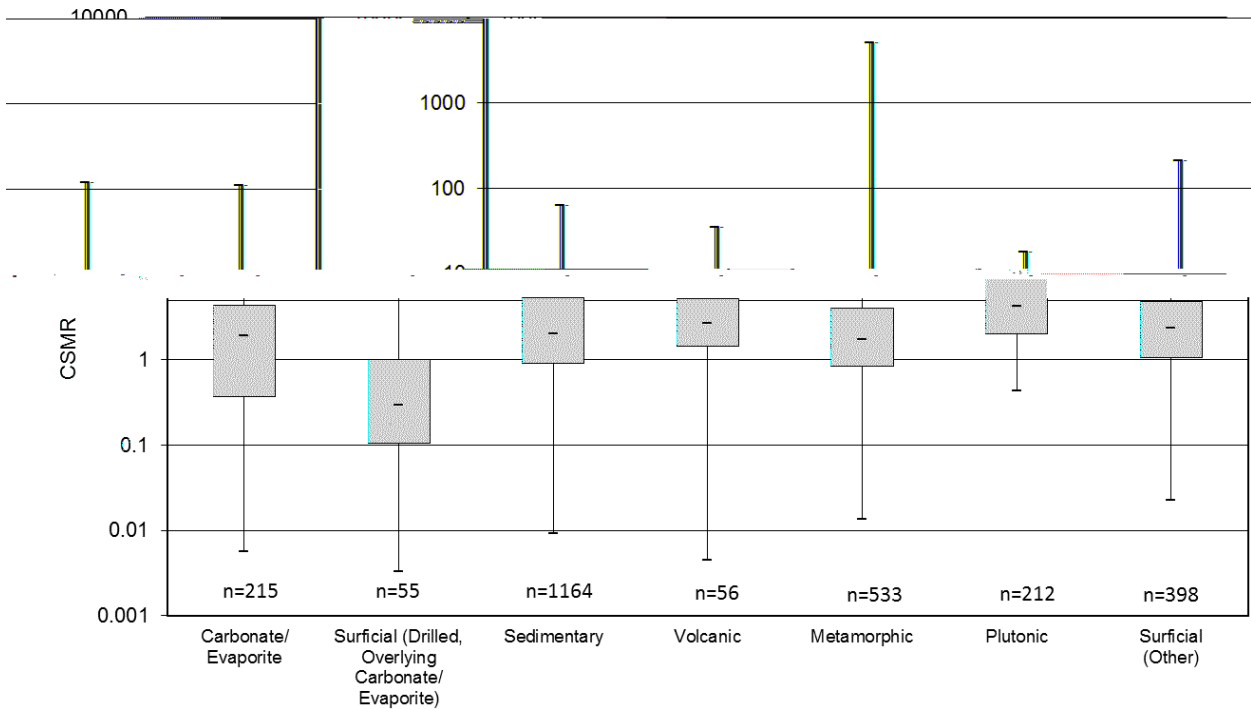
**Figure 6.** Boxplot of pH for the seven major aquifer types assessed.

### Chloride-to-Sulphate Mass Ratio

Compared to plutonic bedrock aquifers, lower chloride-to-sulphate ratios were observed in carbonate/evaporite aquifers and surficial aquifers in contact with these aquifers, largely due to the influence of sulphate-bearing gypsum rocks on groundwater chemistry (Fig. 7, Table 2). Metamorphic aquifers were associated with a lower CSMR compared to other crystalline bedrock aquifer types which may be due to the presence of sulphide-bearing slates or the influence of sulphate-bearing gypsum units in contact with metamorphic aquifers, especially in central areas of the province (Fig. 1). Kruskal-Wallis non-parametric analysis of the distribution of these data shows that they are statistically different populations ( $p < 0.0001$ ) and implementation of the Dunn test shows that the major aquifer types can be grouped as follows ( $p < 0.05$ ):



Because anthropogenic activities can result in elevated concentrations of chloride in aquifers (e.g. groundwater extraction inducing seawater intrusion or the application of road de-icing agents), CSMR is sensitive to these types of activities. Inputs of chloride can also cause the complexation of naturally occurring lead and increase its dissolution and mobility in groundwater (Sun et al., 2015).

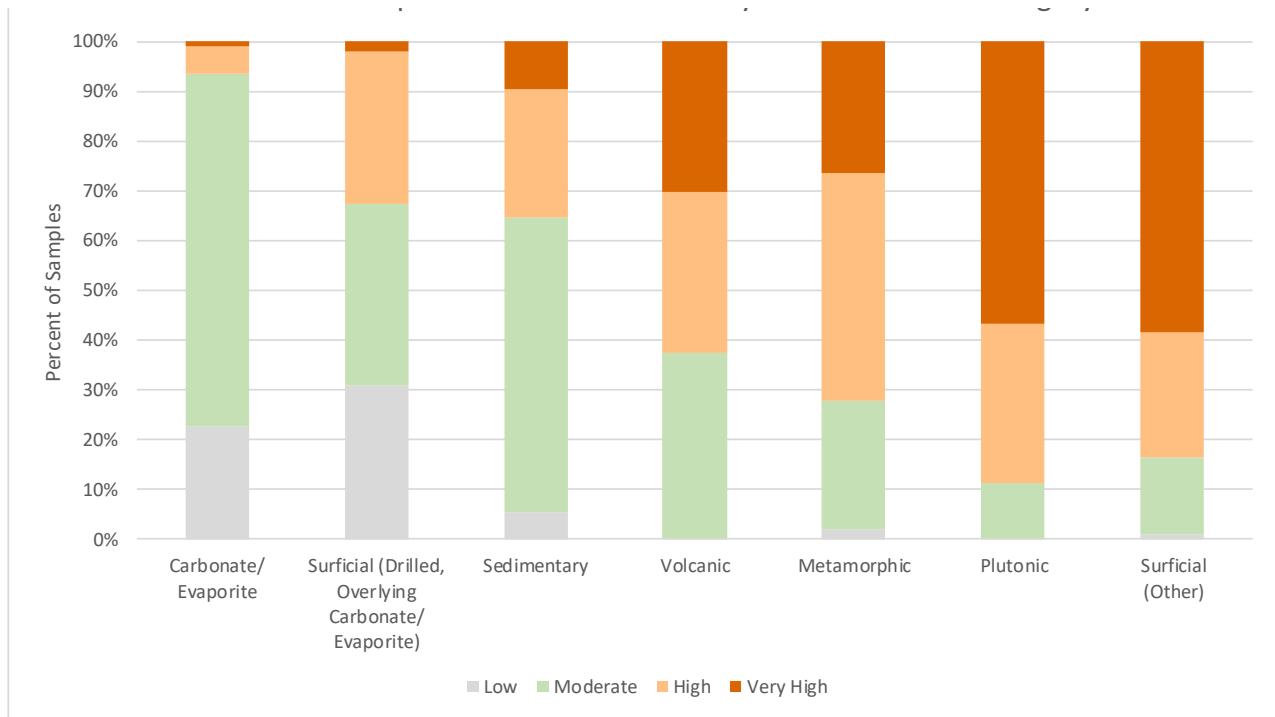
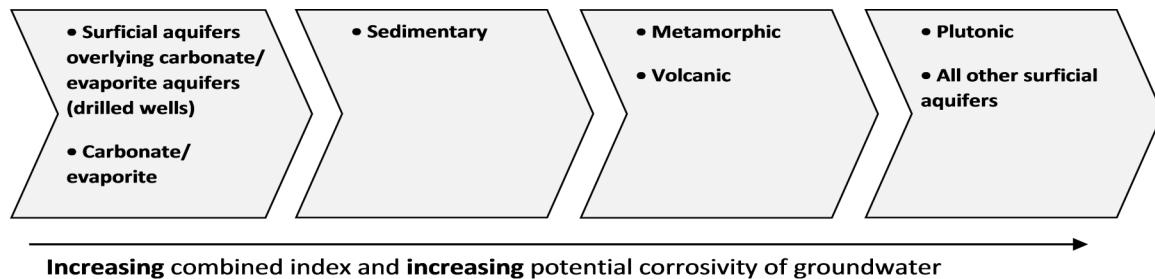


**Figure 7.** Boxplot of chloride-to-sulphate ratio (plotted on a logarithmic axis) for the seven major aquifer types assessed.

### Combined Index

When the two individual indices (LSI and CSMR) are combined into an overall index (Fig. 2), a high potential for corrosive groundwater is observed throughout Nova Scotia, except in carbonate/evaporite aquifers and surficial aquifers in contact with these rocks. In areas where metamorphic aquifers showed low potential for corrosivity, the well water quality appeared to be influenced by the presence of nearby carbonate aquifers or fluvial materials derived from carbonate rocks preserved in bedrock depressions (Kennedy and Utting, 2011).

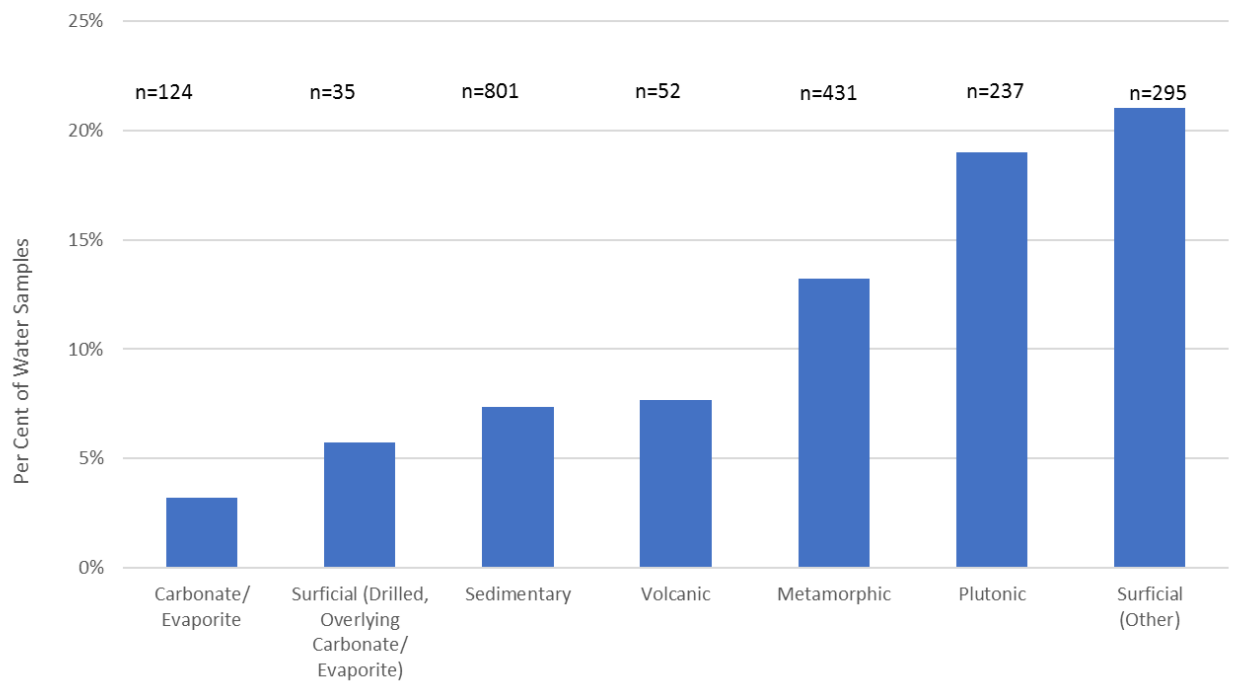
The percent of samples in each of the four overall index categories describing the potential corrosivity of groundwater, from low to very high (Fig. 2), was estimated for each of the seven major aquifer types (Fig. 8). The percent of water samples with a high to very high combined potential corrosivity ranking is shown in Figure 10 for the seven major aquifer types across Nova Scotia. Assigning the combined index category an ordinal variable from 1 (low) to 4 (very high) (Fig. 2), and comparing these ordinal scores using the Kruskal-Wallis test, showed a statistically significant difference between the seven major aquifer types ( $p < 0.0001$ ) and implementation of the Dunn test indicates an overall grouping as follows ( $p < 0.05$ ):



**Figure 8.** Per cent of water samples in each of the four categories of the potential corrosivity of groundwater combined index for the seven major aquifer types assessed.

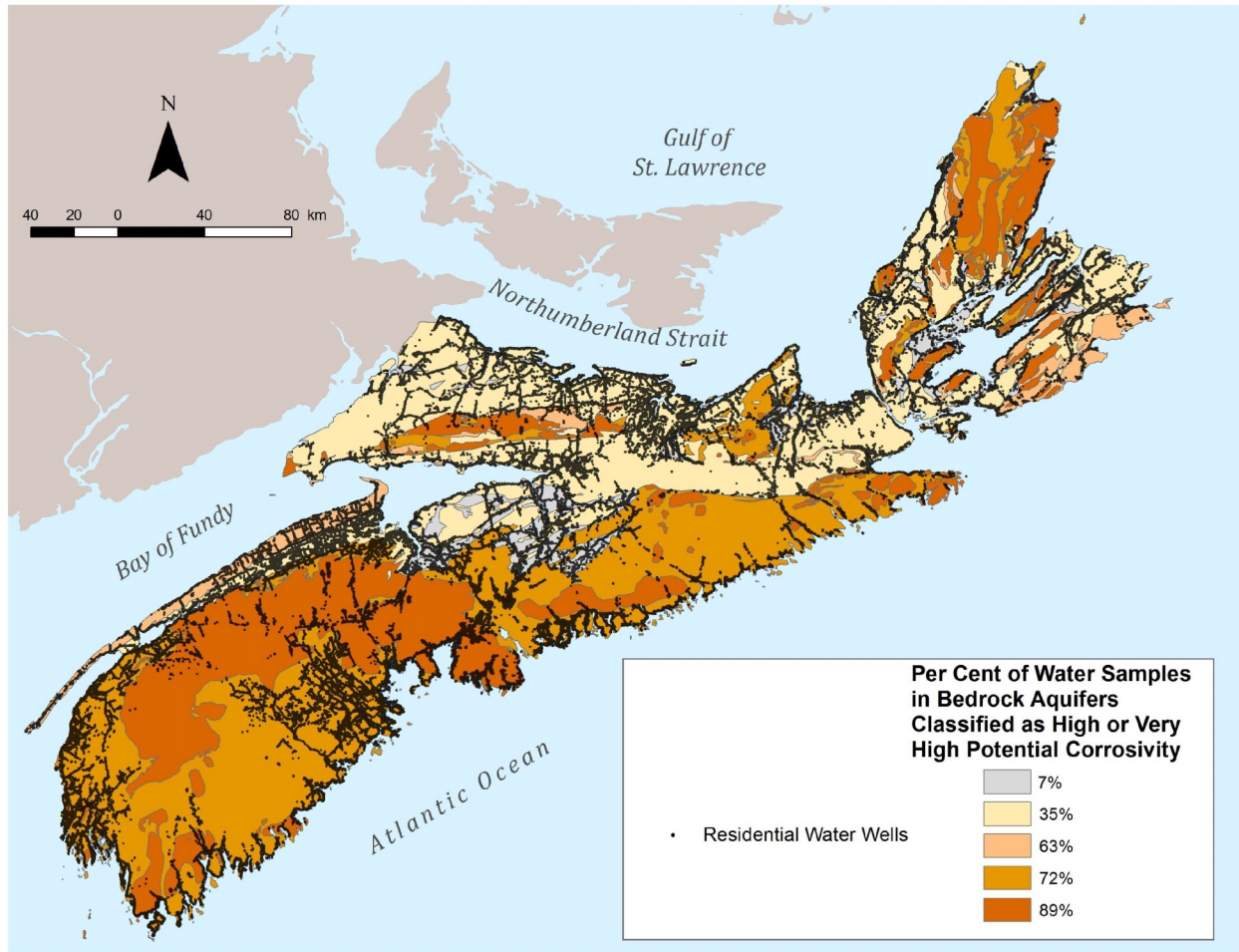
Surficial aquifers and crystalline bedrock aquifers, especially plutonic aquifers, are associated with the highest potential for corrosive groundwater. The warmer groundwater temperatures associated with shallow surficial aquifers relative to bedrock aquifers during the summer may exacerbate the susceptibility of private wells in these aquifers to lead exposure during this period. Only carbonate/evaporite aquifers had less than 10% of water samples in the high or very high combined index category for potential corrosivity of groundwater (Figs. 9 and 10). These aquifers, however, tend to be associated with other water quality characteristics that make them unsuitable for domestic water supply, such as high hardness, TDS, and sulphates. According to the statewide classification by Belitz et al. (2016), Nova Scotia shows a similar prevalence of potentially corrosive groundwater as the northeastern states, which has a higher risk of corrosive groundwater than most other areas in the USA.

Despite the issues with respect to the use of secondary data and the limitations of the use of LSI as an indicator for corrosion, regional-scale patterns with respect to potential corrosivity of groundwater are evident, and these regional-scale patterns are controlled by aquifer hydrogeochemistry. Although LSI may be a poor indicator of corrosion at a site-scale, it may be useful in determining regional groundwater chemistry patterns which, combined with CSMR, can indicate which aquifers or areas are associated with a higher relative vulnerability to corrosion of private well plumbing components. Because water temperature is not typically measured during routine sampling, and LSI is not especially sensitive to TDS, the use of a simpler index that combines key parameters known to affect the mobility of lead, such as alkalinity and pH, could be considered. For example, the application of the aggressive index (e.g. Zhang et al., 2014), which is defined as  $pH + \log[(\text{alkalinity} + \text{hardness})]$ , could be investigated as an alternative to LSI. The index might also be improved by using dissolved inorganic carbon, which is considered a more direct indicator of the available carbonate species for the formation of a passivating film compared to alkalinity (Schock, 1989), but is rarely interpreted during routine water sampling.



**Figure 9.** Per cent of water samples exceeding the Health Canada MAC (i.e. 5 µg/L) for lead for each of the seven major aquifer types assessed.





**Figure 10.** The per cent of water well samples that were classified as having high or very high groundwater potential corrosivity are shown for the five major bedrock aquifer types assessed. Surficial aquifers overlying carbonate/evaporite aquifers had 33% of water samples in these categories, whereas all other surficial aquifers had 83% of water samples in these categories.

## Lead

The percentage of lead exceedances of the Health Canada MAC (5 µg/L) in well water for each of the seven major aquifer types assessed in the study follows a similar trend as the percentage of sample results with a very high combined index for potential corrosivity for each of the aquifer types (Table 3; Fig. 9). The highest percentages of lead exceedances are associated with shallow surficial aquifers and crystalline bedrock aquifers, especially plutonic aquifers. Approximately 20% of water samples for lead exceeded the MAC in these aquifer types compared to less than 5% in carbonate/evaporite aquifers. Analysis of a subset of samples where lead data, in addition to the general chemistry parameters used to derive the combined potential corrosivity index, are available (n=1780), shows that approximately 3.5% (n=847) of the samples in the low to moderate potential corrosivity of groundwater categories exceed the MAC for lead, whereas 10.8% (n=933) of samples in the high to very high categories exceed the MAC.

Although it is not possible to differentiate the sources of lead using the existing well water chemistry dataset, groundwater concentrations of lead collected from wells located in hydrostratigraphic units associated with lead mineralization (e.g. Cumberland Group) were compared to samples collected from

**Table 3.** Statistical summary of lead concentrations classified by the seven major aquifer types assessed. The highest reported laboratory detection limit in the dataset is 5 µg/L. 1=Kaplan-Meier; 2=coefficient of variation; min=minimum; max-maximum.

Aquifer Type	Count Observations	Count Detects	Percent Detects	Percent Exceed 5 µg/L	K-M <sup>1</sup> Mean	K-M CV <sup>2</sup>	Min	Median	95%ile	Max	Detects Only	
											Mean	Median
Carbonate/ Evaporite	124	49	39.5%	3.2%	1.9	3.8%	<0.5	2.0	4.6	76.0	4.4	2.0
Surficial (Drilled, Overlying Carbonate/ Evaporite)	35	19	54.3%	5.7%	1.5	2.3%	<0.5	1.2	3.8	20.0	2.6	1.2
Sedimentary	801	309	38.6%	7.4%	4.6	8.7%	<0.5	1.8	7.7	700.0	11.7	1.8
Volcanic	52	21	40.4%	7.7%	2.1	2.6%	<0.5	2.2	8.3	36.0	4.9	2.2
Metamorphic	431	276	64.0%	13.2%	7.1	9.9%	<0.5	1.6	13.4	1420.0	11.0	1.6
Plutonic	237	180	75.9%	19.0%	6.9	3.9%	<0.5	1.6	28.5	260.0	9.1	1.6
Surficial (Other)	295	228	77.3%	21.0%	7.1	4.3%	<0.5	2.3	17.3	416.0	9.1	2.3
<b>TOTAL</b>	<b>1975</b>	<b>1082</b>	<b>54.8%</b>	<b>11.8%</b>								

wells located in other sedimentary aquifers. A significant difference between the two populations was not detected ( $p=0.23$ ), which suggests that geology is not a strong regional-scale control on lead concentrations in well water.

To investigate the relationship between lead and copper in well water, which can also be leached from system plumbing, a correlation was performed using the Kendall-Tau test. A significant positive correlation was found between the two variables ( $\tau=0.36$ ,  $p=0.001$ ). The Health Canada limit for acceptable concentrations of copper in drinking water is currently under review (Health Canada, 2018), and a new maximum acceptable concentration (MAC) of 2000 µg/L has been proposed. Approximately 67% of the wells with copper >2000 µg/L also had lead >5 µg/L.

### Relative Risk Map of Potentially Corrosive Groundwater

Based on the four distinct groupings identified during the comparison of the combined index (LSI and CSMR) for the seven major aquifer types assessed during this study, a simplified relative risk map comprising three risk levels (low, medium, and high) was developed for Nova Scotia (Fig. 11). The simplified risk framework is consistent with other risk maps produced by the Geological Survey Division (e.g. arsenic, Kennedy and Drage, 2017). Although the simplified risk characterization reflects regional patterns with respect to indicators of potential corrosivity and lead concentrations, it should be highlighted that corrosion of private water system components containing lead can occur throughout Nova Scotia, and all private well users should routinely test their well water to ensure it is safe for drinking. For example, the limited survey of four communities (n=55) in Nova Scotia by Maessen et al. (1985) found similar exceedance rates of acceptable lead levels in both carbonate/evaporite aquifers and plutonic aquifers, although the plutonic aquifers had higher mean concentrations.

Kennedy and Polegato (2017) found that about 42% (197,395 well users or 386,408 persons) of Nova Scotian households use private wells for their water supply. The largest concentration of these users is in suburban areas of Halifax, which is underlain by crystalline bedrock (Fig. 10). Analysis of the distribution of private wells relative to the simplified risk zones shows that approximately 56% (~111,100) of private well water supplies obtain groundwater from the highest risk aquifers for potentially corrosive groundwater, while approximately 36% of private well water supplies obtain groundwater from aquifers in the medium relative risk category (Fig. 11).

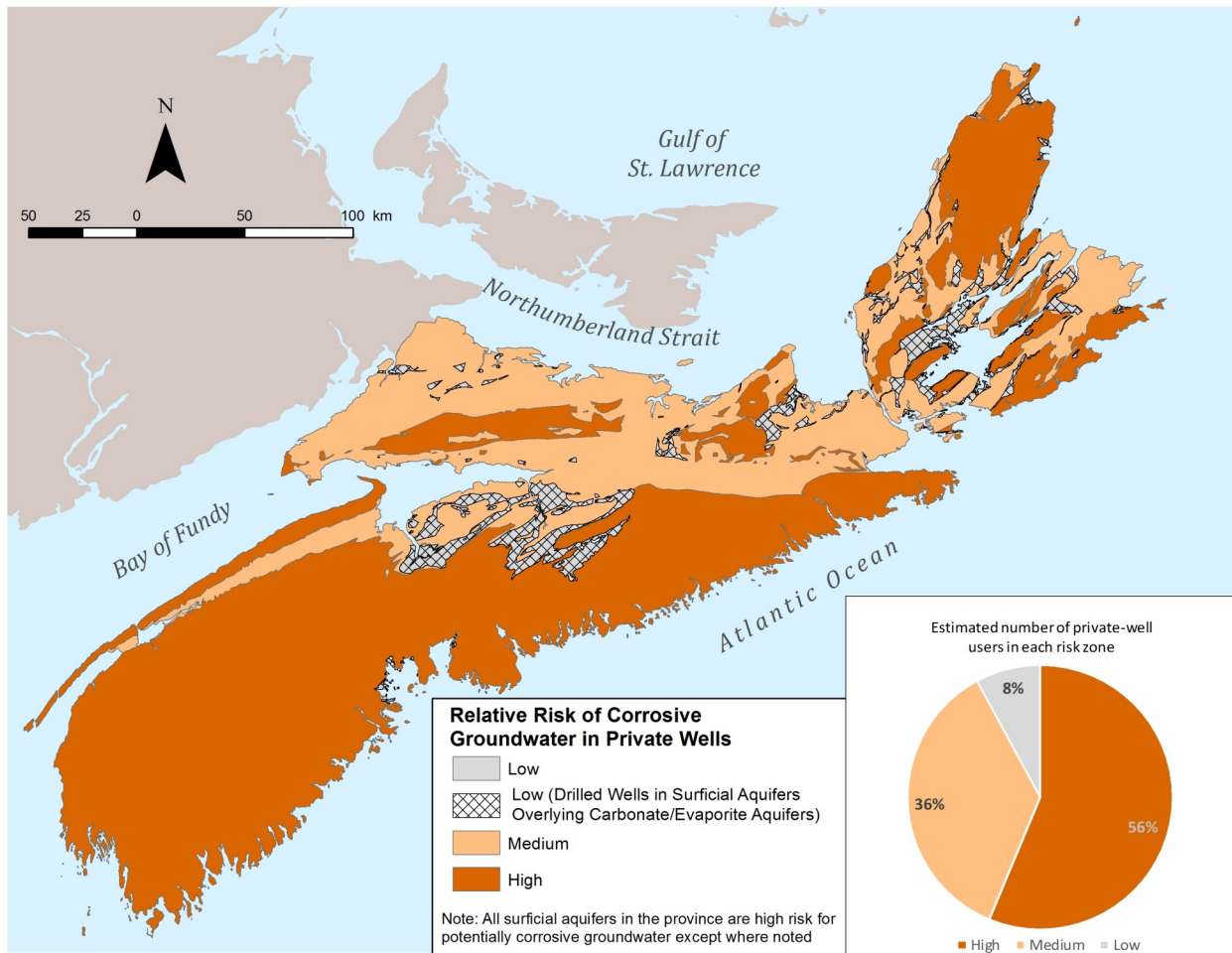


Figure 11. Simplified relative risk map of potentially corrosive groundwater in private well water supplies.

In Nova Scotia, it is recommended that private well users regularly (every two years, or sooner, if a noticeable change is detected) test their water for lead and other metals, and general chemistry parameters by taking a flushed sample (i.e. running the faucet for five to ten minutes before collecting the sample). Because most private well users have a high to very high risk of corrosive groundwater (Fig. 10), and flushing the system may underestimate the risk associated with lead exposure, it is also recommended that well users should collect a separate first draw water sample (i.e. after a six-hour period of stagnation) to adequately assess the risk of lead associated with household plumbing (e.g. Pieper et al., 2015b). If lead is present above the Health Canada MAC in either of the two samples (first draw or flushed), further investigation of the source of lead and/or mitigation measures are recommended. Further investigation may include lead profiling, which involves the collection of a series of water samples to help determine the source of the lead in the water system plumbing materials. Mitigation measures recommended by Nova Scotia Environment (2019) include the replacement of older plumbing materials, water treatment using National Sanitation Foundation (NSF) certified treatment systems, or the use of alternative drinking water sources.

## Summary

Given the low mobility of lead at pH levels typical of Nova Scotia aquifers, and the absence of a statistical correlation between lead and rock units associated with lead mineralization, most lead exceedances recorded in the Nova Scotia Groundwater Chemistry Database can be attributed to leaching of lead from plumbing materials.

There is a relatively high potential for corrosive groundwater to occur throughout most of Nova Scotia, except for carbonate/evaporite aquifers and surficial aquifers in contact with these aquifers, which tend to have lower chloride-to-sulphate ratios, and higher pH and alkalinity. Well water concentrations of lead associated with these aquifers are generally lower compared to other aquifers in Nova Scotia, suggesting that the groundwater's chemical composition may be inhibiting corrosion of plumbing components, resulting in a lower likelihood of elevated lead in private wells. In many cases, however, carbonate/evaporite aquifers are also associated with geochemical characteristics (e.g. high sulphate, TDS and hardness) that make them unsuitable for domestic groundwater supply development, and shallow dug wells, which have a greater potential for corrosive groundwater, tend to be favoured in these areas of the province. Crystalline bedrock aquifers, especially plutonic aquifers, and most of the province's surficial aquifers, have the highest potential for corrosive groundwater, and therefore the greatest risk of elevated lead in well water supplies. It is estimated that approximately 56%, or 111,100, of the province's private well users rely on aquifers associated with a high potential for corrosive groundwater.

Although regional patterns may exist, the evidence shows that corrosion processes are complex and corrosion of metals from plumbing components can occur anywhere in the province, and therefore private well users should regularly test their water to ensure their drinking water is safe. The replacement of older plumbing materials or water treatment systems is generally recommended for private wells to mitigate elevated lead in well water.

About 42% of Nova Scotians use private wells for their water supply, and therefore the risk of lead exposure in these supplies, in addition to municipal water supplies, should be considered to achieve public health objectives. The risk map of potentially corrosive groundwater can be used to help communicate risk and raise awareness, which is important to prevent diseases associated with lead exposure in private wells.

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