GEOSCIENCE AND MINES BRANCH

An Arsenic in Well Water Risk Map for Nova Scotia based on Observed Patterns of Well Water Concentrations of Arsenic in Bedrock Aquifers

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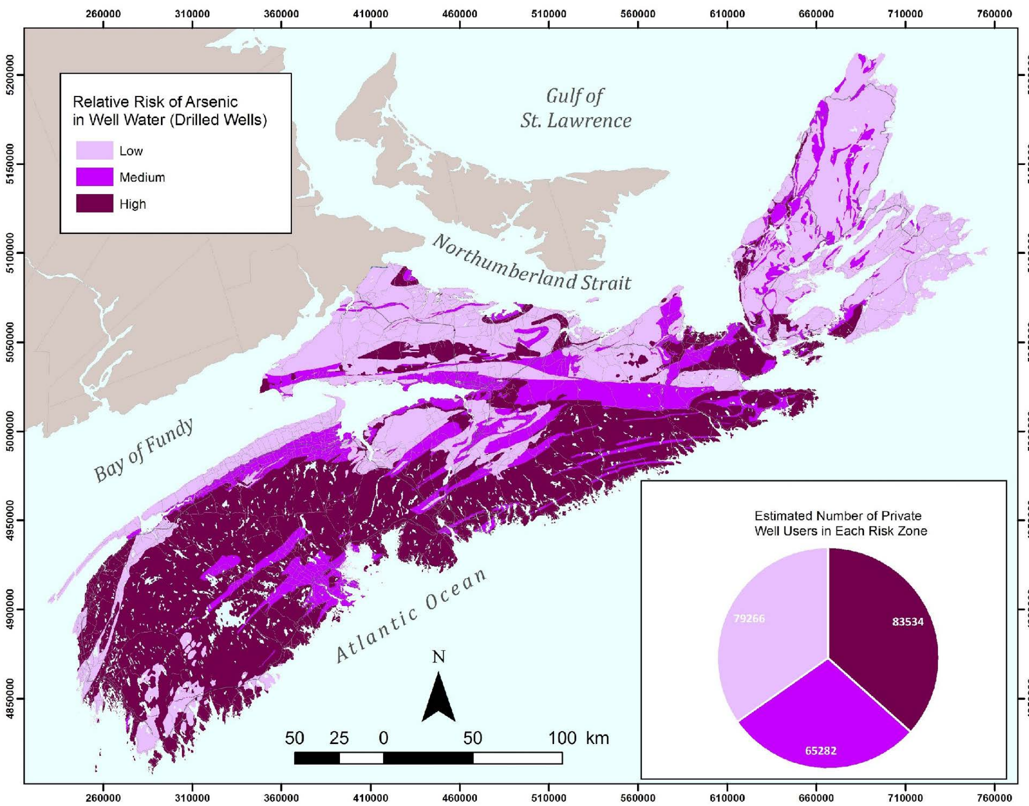

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An Arsenic in Well Water Risk Map for Nova Scotia based on Observed Patterns of Well Water Concentrations of Arsenic in Bedrock Aquifers

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Abstract

The distribution of arsenic in the province's well water compared to available bedrock geology mapping was investigated to further advance the understanding of the relationship between arsenic occurrence in well water and various bedrock units, and to develop a new risk map to communicate the risk of arsenic exposure to well owners. Arsenic exposure in well water is associated with a range of adverse health effects, including increased incidence of skin and internal cancers.

Arsenic in well water in Nova Scotia is principally attributed to geogenic sources, including arsenopyrite and arsenian pyrite, which is hosted in various bedrock types. Bedrock geology is therefore the most important provincial-scale control on the distribution of arsenic concentrations in well water, and it is mainly drilled wells that are associated with elevated levels of arsenic. Available arsenic in well water data were compiled for drilled wells and a source aquifer (i.e. bedrock unit) was assigned to each sample location. The frequency of arsenic in well water samples exceeding the Health Canada maximum acceptable concentration (MAC) of 10 µg/L was tabulated for the province's five major groundwater regions and over 60 bedrock units. Based on the observed exceedance rates of the Health Canada MAC for arsenic in well water for the various bedrock units, an arsenic in well water risk map was produced dividing the province into low- (<5% exceeding), medium- (5 to <15% exceeding) and high- (≥15% exceeding) risk zones.

Both the highest concentrations (i.e. >1000 µg/L) and exceedance rates were found in the metamorphic and plutonic groundwater regions of Nova Scotia, largely due to the presence of arsenic mineralization in these rocks. The study highlighted other potentially important controls on arsenic occurrence and mobility including metamorphic grade, contact metamorphism and the geochemical environment. Well water samples collected from Goldenville Group and South Mountain Batholith bedrock aquifers in southern Nova Scotia were associated with the highest frequency of arsenic exceeding the safe limits for drinking water (>40%), and these aquifers are situated in areas of high-density domestic groundwater use. Demographic analyses indicate that over 80,000 households (37%) are in high-risk areas, and the overall frequency of private well water exceeding safe drinking water limits in Nova Scotia may be as high as 20% (~90,000 persons). The risk map will be published in several formats to communicate risk to private well owners in Nova Scotia with the aim encouraging appropriate well water testing and treatment and preventing disease.

Introduction

Arsenic is a Class I human carcinogen and has been associated with an increased incidence of skin, lung, kidney, bladder and liver cancers (Hughes et al., 2011; Saint Jacques et al., 2014). New research has also shown an association between adverse birth outcomes and arsenic in well water (e.g. Shi et al., 2015). Arsenic is a widespread naturally occurring contaminant in Nova Scotia's groundwater and is therefore a concern for many of the province's private well owners. Recent studies have demonstrated that arsenic body burden (total amount of arsenic in a person's body) among Nova Scotians is largely related to well water exposure (Cull, 2011; Yu et al., 2013).

A review was published (Kennedy and Drage, 2016) summarizing the past forty years of research and activities related to the occurrence of arsenic in water wells in Nova Scotia since the emergence of the issue in the 1970s when a case of arsenic poisoning was detected in the community of Waverley (Grantham and Jones, 1976). During this review, a number of historical arsenic in well water surveys conducted by the province were compiled, and considerable efforts were made to generate sample location co-ordinates for the survey results. The review found that elevated levels of arsenic are associated mainly with drilled wells in bedrock aquifers, and hence the distribution of arsenic in well water in Nova Scotia was principally attributed to geogenic sources, including arsenopyrite and arsenian pyrite, which are hosted in various bedrock types. It is estimated that up to 200,000 households in Nova Scotia (~40% of the population) obtain their domestic water supply from a privately owned drilled well intercepting a fractured bedrock aquifer, and these well users are not regulated with respect to water quality.

Health Canada's (2017) maximum acceptable concentration (MAC) of arsenic in drinking water is 10 µg/L. Exceedance rates for drilled wells across the province are around 12% (overall exceedance rate for bedrock aquifers), although much higher exceedance rates (>50%) have been reported in some communities using private wells (Kennedy and Drage, 2016). Most of these communities are situated in Halifax, Hants and Lunenburg counties (Fig. 1) and are underlain by metasedimentary or granitoid bedrock. The review also found that of the two major species of arsenic found in Nova Scotia groundwater (inorganic As^{5+} and As^{3+}), the less toxic As^{5+} (or arsenate) is more prevalent based on the few available surveys that included speciation analyses (Kennedy and Drage, 2016).

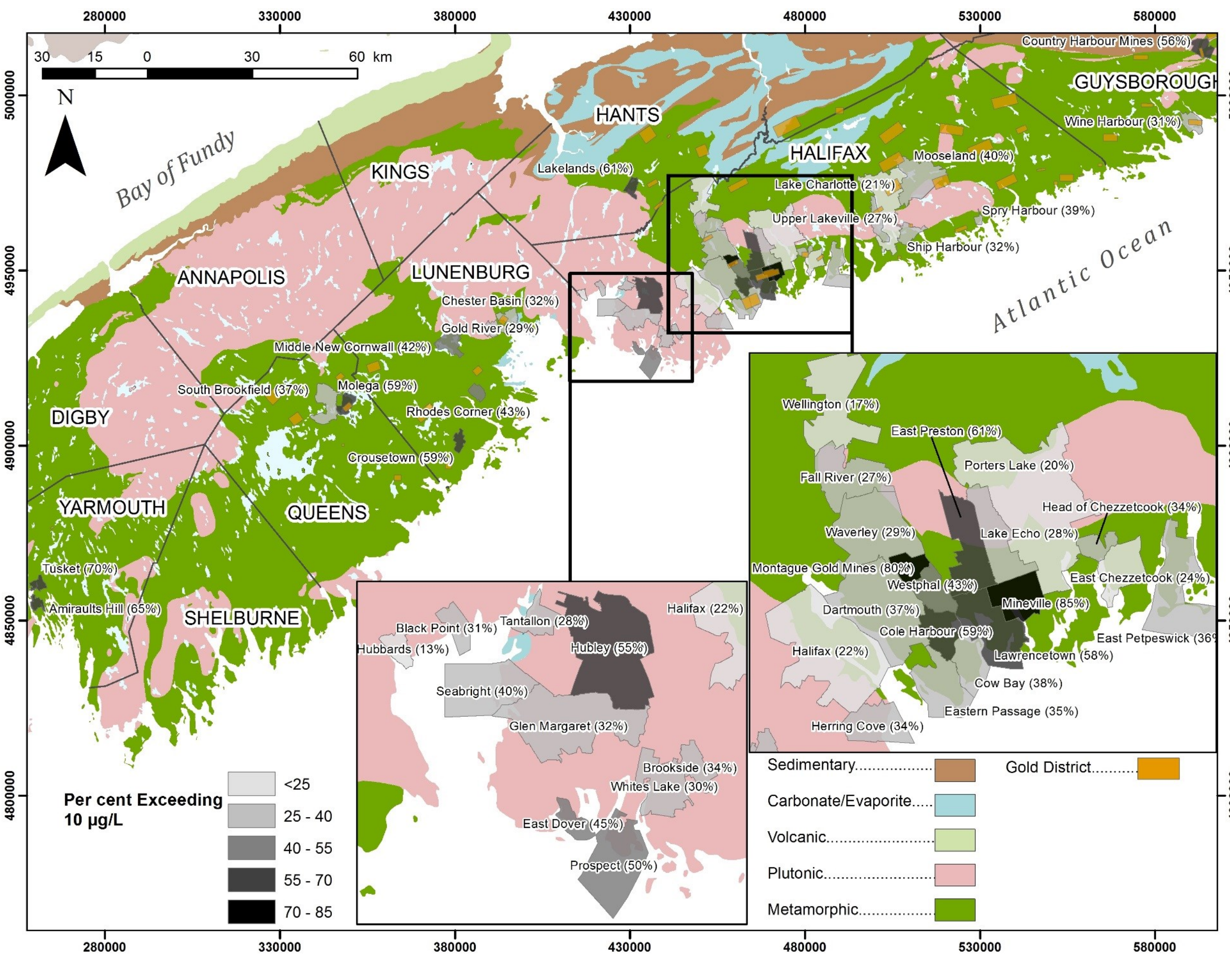

Figure 1. Per cent of samples exceeding 10 µg/L arsenic in well water for various communities surveyed in the Meguma terrane, bedrock groundwater regions and historical gold districts (*modified from* Kennedy and Drage, 2016).

Arsenic concentrations in groundwater depend on several factors, such as the availability of arsenic in the geological materials in contact with groundwater, the aquifer geochemistry (e.g. Eh, pH and the presence of iron oxides and competitive ions) and the aquifer's physical structure (e.g. fracture-flow dynamics), all of which contribute to the spatial variability of the distribution of arsenic in groundwater at local to regional scales. Iron oxides, sulphide minerals (such as pyrite) and clay mineral surfaces are all potential sources and sinks of arsenic, and the fate and species of arsenic present in groundwater is governed by pH and Eh. In addition to pH and Eh controls on arsenic mobility, growing evidence suggests that many of the processes controlling the stability and mobility of arsenic are affected by microbial activity in the subsurface (e.g. Mumford et al., 2015).

A conceptual model for arsenic occurrence in water wells in metasedimentary rocks was proposed by Yang et al. (2014). Arsenic-bearing sulphide minerals (e.g. arsenopyrite and arsenian pyrite) are oxidized in the oxic portion of the groundwater-flow path, resulting in the release of iron and arsenic, followed by oxidation of iron to form iron oxyhydroxide particulates, which are transported in fractures and sorb arsenic along the flow path until they are intercepted by boreholes. In the anoxic portions of the flow path, arsenic associated with iron oxyhydroxides may be mobilized through microbially mediated reductive dissolution. This model for arsenic mobilization in metasedimentary rock aquifers can be readily extended to other types of crystalline rock aquifers (e.g. granite) that host arsenic mineralization. Other mechanisms for arsenic occurrence in groundwater include the desorption of arsenic from clay mineral surfaces, a process largely controlled by pH and the competition for adsorption sites by ions such as phosphate (Serfes, 2005).

Risk maps for arsenic in well water have been used over the last 35 years in the province to communicate the relative risk of arsenic to private well owners in order to encourage appropriate well water testing and treatment, and to inform land development planning, groundwater supply development planning, and strategic allocation of government resources for risk mitigation. The most recent arsenic in well water risk map was published in 2005 (Fig. 2; Nova Scotia Environment, 2005). Details on the province's past arsenic in well water risk-mapping activities are in Kennedy and Drage (2016).

The main objectives of the current research are to advance the understanding of the relationship between arsenic occurrence in well water and various bedrock types in Nova Scotia, and to produce a refined risk map that will be used to communicate the risk of arsenic exposure in well water to private well owners, promote appropriate testing and treatment of well water, and to prevent disease associated with arsenic exposure. The new risk map will consider all available bedrock well-water chemistry data, including recently compiled datasets (see Kennedy and Drage, 2016) and the latest bedrock geology mapping. A considerable portion of the province's bedrock geology was recently mapped at a higher resolution than was previously available, including south-central Nova Scotia (Horne et al., 2009), southwestern Nova Scotia (White et al., 2012) and Cape Breton Island (Barr et al., in press). Although elevated levels of arsenic can also occur in surficial aquifers in Nova Scotia (e.g. dug wells), they are relatively uncommon (Kennedy and Drage, 2016), and hence these aquifers were not included in the present analysis.

Methods

The analysis of arsenic in well water in Nova Scotia relied on data from the Nova Scotia Groundwater Chemistry Database, which is maintained by the Nova Scotia Department of Natural Resources. Most of these data are publicly available (Nova Scotia Department of Natural Resources, 2016a); however, some data (including data used for this study; current to 2016) are not public due to privacy considerations. The database includes the recently compiled datasets described in Kennedy and Drage

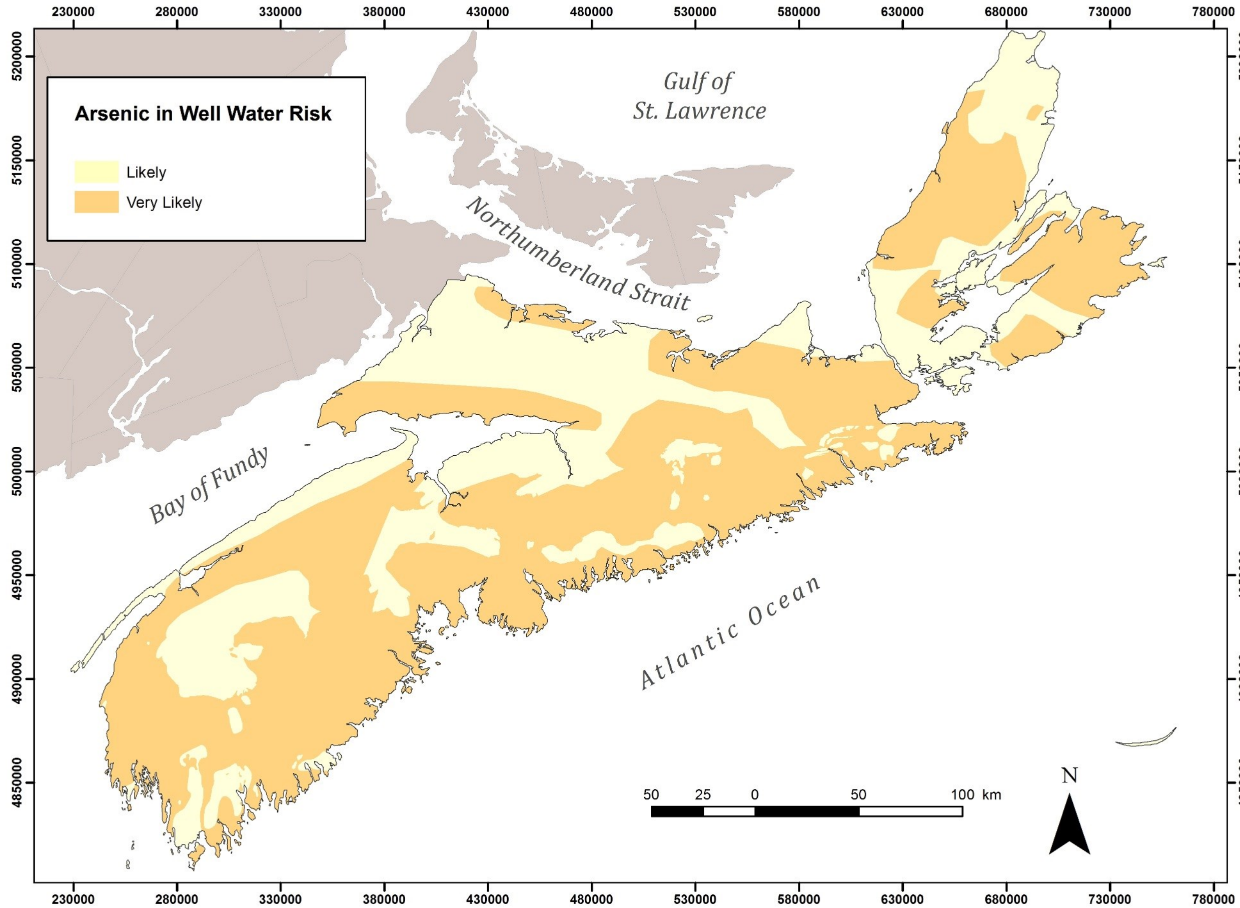

Figure 2. Arsenic in well water risk map for the province developed by the interprovincial Arsenic Working Group in 2005 (*after* Nova Scotia Environment, 2005).

(2016) (e.g. historical government well water surveys, Department of Health database). The provincial groundwater chemistry database consists mostly of non-domestic well water sample results that are believed to reflect ambient groundwater chemistry (i.e. treated well water samples and data from contaminant 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, and Nova Scotia Environment groundwater chemistry data from registered public drinking-water supplies, pumping tests, municipal groundwater systems and provincial observation wells.

Arsenic in well water data were filtered to include only data where the laboratory method detection limit was ≤5 µg/L and where there was adequate confidence in the sample location (i.e. include only water sample locations accurate to the land parcel scale), aquifer type (i.e. include only water samples from bedrock aquifers), and sample type (i.e. include only raw or suspected raw water samples). The arsenic data were plotted on the most recent bedrock geology maps for Nova Scotia (Fisher, 2006a; Fisher, 2006b; Fisher and Poole, 2006; Horne et al., 2009; White et al., 2012; White et al., 2014; Nova Scotia Department of Natural Resources, unpub. data, 2017; Barr and White, in press) in ArcGIS™10 (Esri, Inc.) geographic information system software, and a bedrock unit was assigned to each water sample.

Analysis of the relationship between bedrock geology and arsenic concentrations in well water is summarized by the province's five major bedrock groundwater regions (Kennedy and Drage, 2009). For each region, summaries of arsenic exceedance rates were generated for constituent bedrock units (or aquifers) that had a minimum of five water-sampling locations. Where multiple sample results for a given well location were available (e.g. time series), the maximum arsenic result was used for the estimation of the exceedance rate of the Health Canada MAC. Thresholds were established for low- (<5%), medium- (5 to <15%) and high- (≥15%) risk categories in terms of the percentage of well water samples exceeding the acceptable limit of 10 µg/L for a given bedrock unit, and each bedrock unit was assigned to one of the three risk categories. Non-parametric statistical tests, such as the Tarone-Ware and Kruskal-Wallis tests, were used to determine whether various groupings of samples were statistically different populations. Statistical analyses were conducted using statistical software STATA® by StataCorp, LP, and ProUCL 5.0, which was developed by the United States Environmental Protection Agency (U.S. Environmental Protection Agency, 2013).

Limitations of the Approach

The risk mapping assumed that areas of the province with significant control concerning arsenic in well-water chemistry can be extrapolated into areas without such data control but with similar bedrock geology. The spatial coverage is not regularly distributed across the province and is biased since the dataset includes surveys where arsenic in well water was investigated as a human health concern. Another limitation of the risk-mapping approach includes the assumption that bedrock geology is the dominant control, rather than other factors such as the pH and Eh environment, which may be independent of bedrock geology. The accuracy of the available bedrock geological mapping constrains the accuracy of the derivative provincial-scale risk map because bedrock maps of varying resolution (from 1:10 000 to 1:500 000) were combined to cover the province.

The compiled water chemistry data spans a period of 40 years and comes from a variety of sources ranging from samples collected by homeowners, who have shared their arsenic results with government, to well water quality surveys conducted by groundwater professionals. Hence sampling and laboratory analytical methods are not consistent across the dataset. Field measurements of relevant geochemical parameters, such as dissolved oxygen, pH and Eh, were not part of the dataset. Although the quality of the well water chemistry data used in the analysis varies, it is still considered useful for detecting regional-scale trends and to produce synoptic-scale risk maps.

Results and Discussion

A total of 2151 unique arsenic in well water sample locations were compiled, with the greatest sample density in the suburban area of the Halifax Regional Municipality (HRM) (Fig. 3). Statistical summaries of the data classified by groundwater region (Fig. 4, Table 1) show that higher concentrations of arsenic are generally associated with the plutonic and metamorphic groundwater regions, and that the Kaplan-Meier mean is about an order of magnitude higher for these bedrock aquifer types. Maximum concentrations of arsenic in well water associated with these groundwater regions exceed 1000 µg/L. Statistical comparison of arsenic in well water concentrations using the Kruskal-Wallis non-parametric test found a significant difference ($p<0.05$) between arsenic concentrations grouped by groundwater region. A summary of arsenic in well water exceedance rates of the Health Canada MAC for various bedrock units within each of the five groundwater regions and a more detailed exploration of the potential controls on arsenic occurrence in groundwater with respect to lithogeochemistry and aquifer geochemistry is presented below.

Metamorphic Groundwater Region

Distribution of Arsenic and Summary of Exceedance Rates

Low to high concentrations of arsenic are observed in the metamorphic groundwater region of Nova Scotia (Fig. 5). The overall exceedance rate of the Health Canada MAC of 10 µg/L for metamorphic

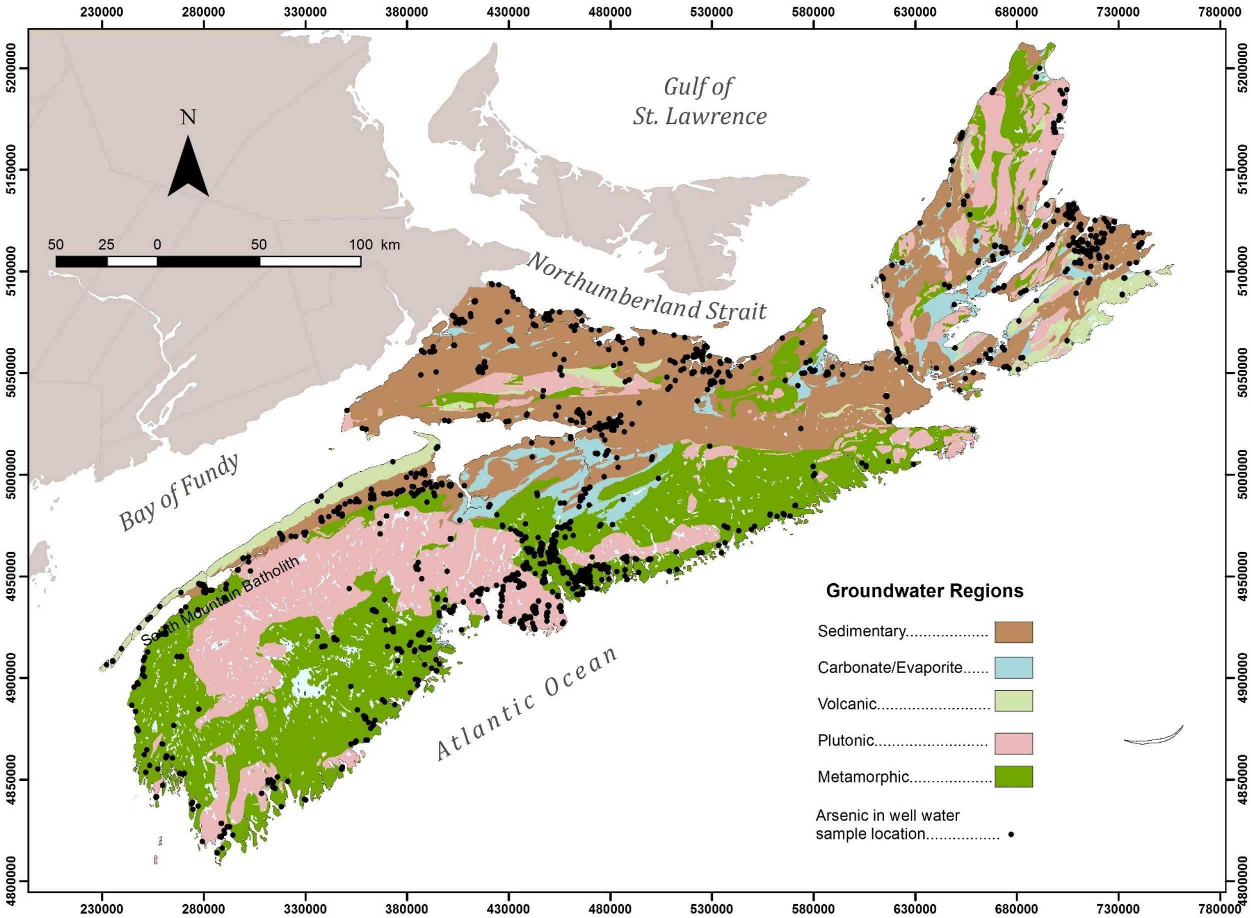

Figure 3. Distribution of arsenic in well water samples compared to the province's five groundwater regions.

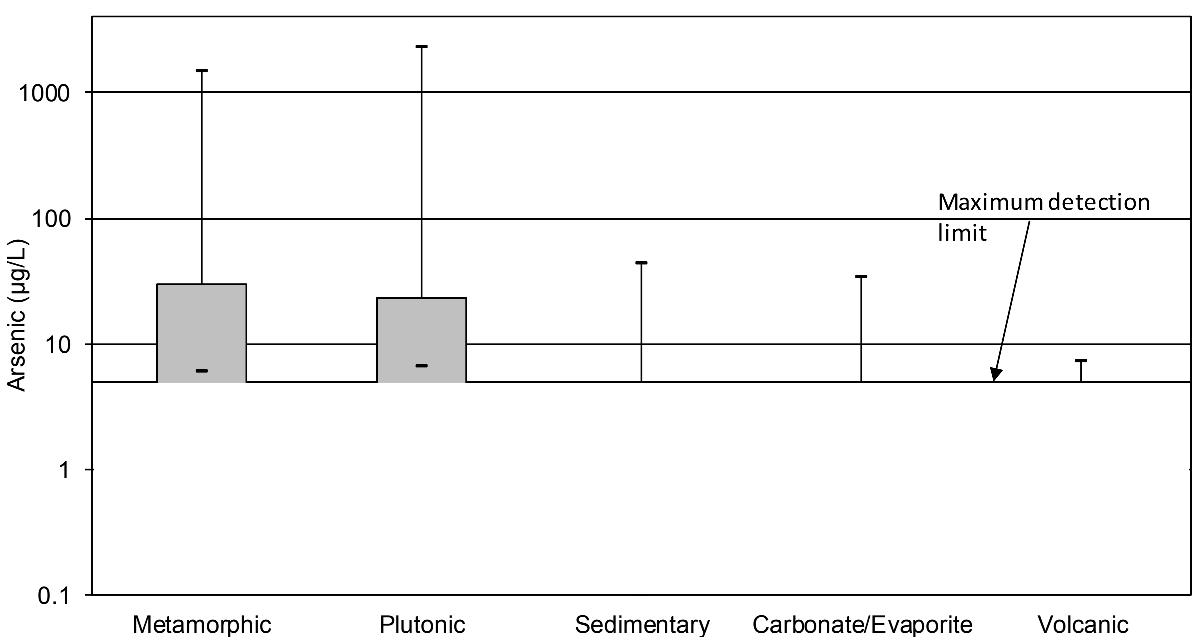

Figure 4. Censored box and whisker plot summarizing the minimum, 25%, median, 75% and maximum arsenic concentrations for the province's five groundwater regions. The portion of the plot below the maximum detection limit (e.g. 5 µg/L) is not shown.

Table 1. Statistical summary of arsenic concentrations classified by groundwater region. The highest reported laboratory detection limit is 5 µg/L.

	All Data									*Detects Only*	
Groundwater Region	**Count Observations**	**Count Detects**	**Per cent Detects**	**Per cent Exceed[1]**	**K-M[2] Mean (µg/L)**	**Min (µg/L)**	**Median (µg/L)**	**95th Percentile (µg/L)**	**Max (µg/L)**	**Mean (µg/L)**	**Median (µg/L)**
Metamorphic	903	529	58.6%	38.8%	39.9	<0.6	6.0	180.0	1460.0	67.9	20.0
Plutonic	305	224	73.4%	37.1%	64.5	<0.6	6.6	354.0	2300.0	87.6	10.9
Sedimentary	763	369	48.4%	4.7%	2.7	<0.1	2.0	10.0	44.0	5.2	3.2
Carbonate/Evaporite	141	79	56.0%	4.3%	2.6	<0.6	1.6	9.0	34.0	4.3	2.1
Volcanic	39	9	23.1%	0.0%	1.0	<0.6	1.0	4.4	7.2	3.0	2.2
TOTAL	**2151**	**1210**	**56.3%**	**23.5%**							

1. Per cent of arsenic in well water results exceeding the maximum acceptable concentration of 10 µg/L
2. Kaplan-Meier

aquifers is 39%, ranging from 0 to 80% for individual bedrock units (Table 2). Most of the province's metamorphic rocks and available well chemistry data (e.g. Table 2) are associated with the marine-deposited Cambrian to Ordovician Goldenville and Halifax groups, and hence the analysis of patterns of arsenic in well water concentrations in the metamorphic groundwater region focuses on these bedrock units.

Based on observed patterns, Goldenville Group units located in Lunenburg and Halifax counties (including the Tangier, Taylors Head, Government Point and Moshers Island formations) and Goldenville Group units located in the southwest extremity of the province in Yarmouth and Digby counties (including the Church Point and Moses Lake formations) are the most likely to have arsenic in well water exceeding acceptable levels in the metamorphic groundwater region (Fig. 5, Table 2). It is evident that there is considerable heterogeneity in arsenic in well water concentrations at both regional and local scales (Fig. 5). In some areas, where high-density well-water chemistry data are available, local-scale patterns (e.g. hots spots in a subdivision) in the distribution of arsenic in well water can clearly be observed for a given bedrock formation, whereas in other areas the distribution appears to be highly heterogeneous.

Although there is considerable heterogeneity, water wells intercepting Goldenville Group rocks are more likely to have elevated arsenic in well water compared with Halifax Group rocks (Table 2). The rate of well water samples exceeding the Health Canada MAC is approximately three-times higher in Goldenville Group aquifers compared with Halifax Group aquifers (Table 2), and statistical comparison using the Tarone-Ware rank sums non-parametric test for the two bedrock groups confirms that arsenic concentrations in well water associated with the Goldenville Group are significantly greater than the Halifax Group ($p<0.05$).

Exceedances of the safe limit for arsenic in drinking water in Halifax Group aquifers tend to be limited to small areas. For example, during a 1977 survey of the Collins Park Subdivision in Wellington (Nova Scotia Department of Environment, unpub. data, 1977), Halifax County, which is underlain by Cunard Formation bedrock, elevated levels of arsenic were detected in most of the subdivision's drilled water wells. Exclusion of these water sample results from the Table 2 summary of the Cunard Formation lowers the exceedance rate of arsenic from 22% to less than 10%. Other areas with anomalously high concentrations of arsenic in the Halifax Group include the community of Cow Bay in Halifax County.

The per cent of well water samples exceeding the Health Canada MAC for arsenic for the Goldenville and Halifax groups in the Eastern Shore area of the province (east of the urban core of HRM) could not

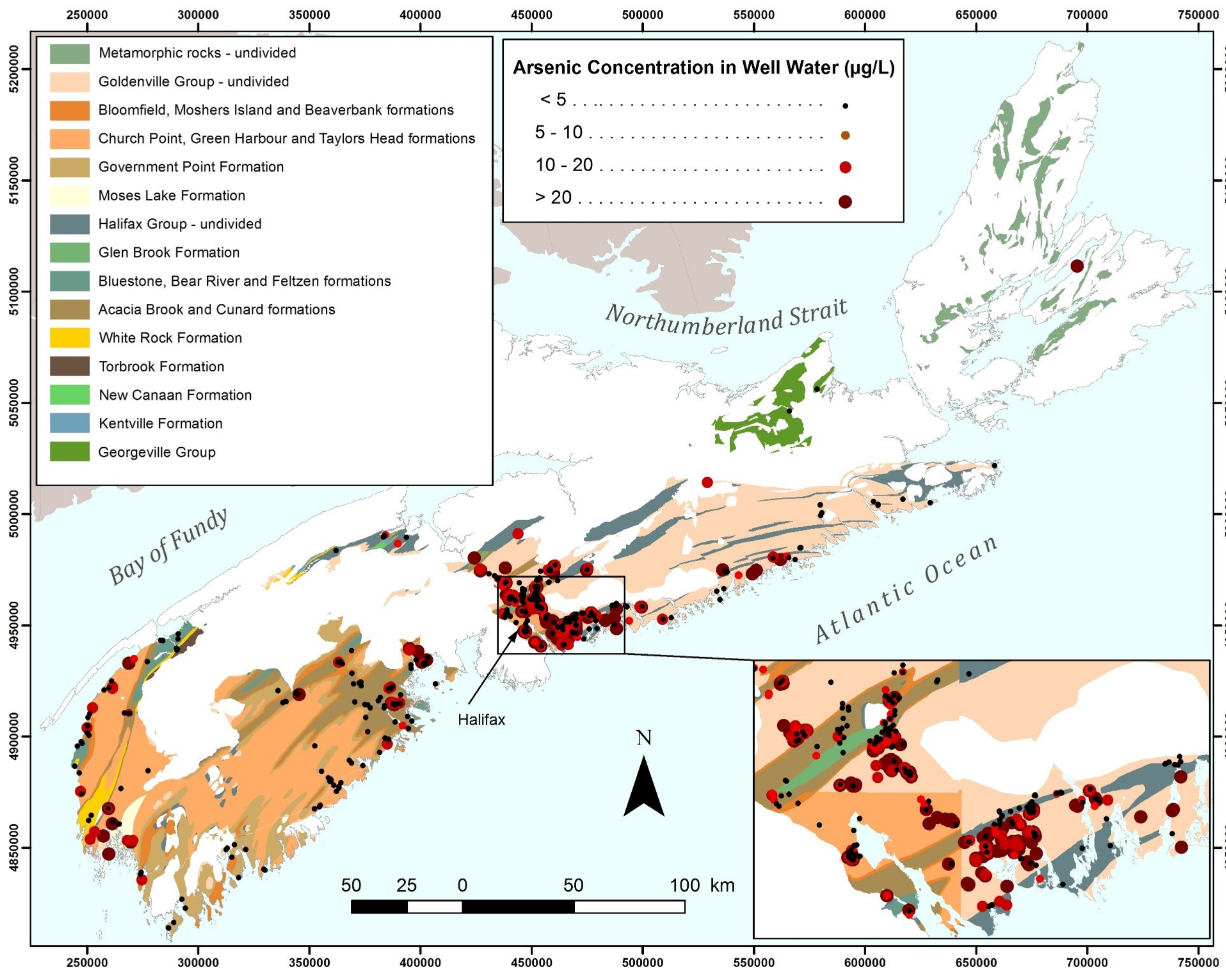

Figure 5. Concentrations of arsenic in well water in the metamorphic groundwater region compared to bedrock geology.

be reliably compiled at the formation level (e.g. Tangier, Moose River, Taylors Head, Cunard formations) due to the poor resolution of available bedrock mapping of the Meguma terrane in this area.

Relation of Arsenic to Lithogeochemistry

Goldenville and Halifax group rocks have abundant sulphide-bearing minerals (e.g. arsenopyrite) that commonly host arsenic. There is considerable heterogeneity with respect to whole-rock arsenic concentrations in Nova Scotia's metasedimentary bedrock units due to factors such as depositional environment (oxic vs. anoxic), grain size, and the redistribution of arsenic in the rock matrix from post-depositional hydrothermal activity. A review of whole-rock geochemical data for various rocks of the Goldenville and Halifax groups shows median arsenic concentrations ranging from 4 to 48 mg/kg, with generally higher concentrations associated with the Goldenville Group (Kennedy and Drage, 2016). In known mineralized zones, much higher arsenic concentrations are documented. White (2010a) reported concentrations as high as 13 000 mg/kg in the Green Harbour Formation (Goldenville Group), and Horne and Pelley (2007) reported concentrations greater than 18 000 mg/kg in the Tangier Formation (Goldenville Group).

In New England, lithogeochemistry was determined to be an important predictor of arsenic concentrations in well water. A band of calcareous metasedimentary rock units (e.g. limestone and/or dolostone protoliths), extending from Maine to Connecticut, was associated with the highest frequency of well waters exceeding 10 µg/L of arsenic (Zheng and Ayotte, 2015), and due to the absence of arsenic-rich

Table 2. Percentage of water samples exceeding 5 and 10 µg/L of arsenic in well water based on available data for various metamorphic bedrock units with at least five samples.

Bedrock Unit		Count	Per cent of As values >5 µg/L	Per cent of As values >10 µg/L
METAMORPHIC GROUNDWATER REGION		**903**	**50.8%**	**38.8%**
Goldenville Group		**657**	**60.4%**	**47.6%**
	Goldenville Group - Undivided	405	70.1%	59.0%
	Moses Lake Formation	5	80.0%	80.0%
	Church Point Formation	16	43.8%	37.5%
	Taylors Head Formation	126	54.0%	32.5%
	Moshers Island Formation	29	51.7%	31.0%
	Green Harbour Formation	29	27.6%	24.1%
	Beaver Bank Formation	11	36.4%	18.2%
	Government Point Formation	34	20.6%	14.7%
Halifax Group		**235**	**26.0%**	**15.3%**
	Halifax Group - Undivided	57	14.0%	8.8%
	Cunard Formation	131	32.8%	22.1%
	Feltzen Formation	14	14.3%	7.1%
	Acacia Brook Formation	8	25.0%	0.0%
	Glen Brook Formation	10	20.0%	0.0%
	Bear River Formation	6	0.0%	0.0%
Rock Notch Group (White Rock Formation)		**7**	**0.0%**	**0.0%**

pyrite, the source of arsenic in these rocks was interpreted to be from secondary mineral hosts redistributed during metamorphism (O'Shea et al., 2015). Similar lithogeochemical controls on the distribution of arsenic in Goldenville and Halifax group rocks have not been found. Although the lithological differences between the metasandstone-dominated Goldenville Group and the overlying slate-dominated Halifax Group are well documented (e.g. White 2010a, b; White and Barr, 2010), distinct lithogeochemical differences between the two groups (besides higher silica and prevalence of calcite cement and nodules in some Goldenville rocks), such as the concentration of sulphide or iron/manganese oxides, have not been established.

Structural differences between the Goldenville and Halifax groups may be more important in explaining the observed arsenic in well water concentration patterns in Nova Scotia. As compared to the Halifax Group, Goldenville Group bedrock hosts most of the Meguma terrane's tight anticlines, and since the hinges of these anticlines are associated with anomalous concentrations of veins arrays and arsenic and gold mineralization, most of Nova Scotia's gold districts are situated in areas underlain by Goldenville Group bedrock (Horne and Pelley, 2007). These mineralized zones may impart greater concentrations of arsenic to groundwater compared to Halifax Group bedrock.

Metamorphic grade could be another potential influence on the spatial distribution of arsenic in well water concentrations. There appears to be regional correspondence between areas of lower grade metamorphic rocks (presence of chlorite and biotite minerals) in Yarmouth, Halifax and Lunenburg counties and areas with higher rates of arsenic in well water exceeding the Health Canada MAC (Fig. 5). Similarly, Ryan et al. (2015) and O'Shea et al. (2015) found whole-rock arsenic concentrations to decrease with increasing metamorphic grade in pelitic metasedimentary rocks in New England, and metamorphic grade was therefore interpreted to be an important predictor of the distribution of arsenic concentrations in well water. The localized area of elevated arsenic in well water associated with the Cunard Formation may be related to contact metamorphism. Increases in the modal abundances of sulphide-bearing minerals in the marginal phases of the South Mountain Batholith and related contact aureole in the Cunard Formation is well documented by Clarke et al. (2009) and may contribute to the higher arsenic values in well water in these areas. For example, in the Collins Park subdivision area, contact metamorphism with the Kinsac pluton has been observed that may have caused local alteration of the Cunard Formation rocks and enrichment of arsenic. It is notable that an arsenic deposit consisting of vein-type arsenopyrite was identified in Wellington in the 1920s, when arsenic was used as an ingredient in the manufacture of pesticides (Nova Scotia Department of Public Works and Mines, 1926).

Relation of Arsenic to Aquifer Geochemistry

In addition to the heterogeneity of whole-rock arsenic concentrations, well water concentrations of arsenic depend on fracture-flow dynamics and local hydrogeochemical controls, such as Eh and pH. Most well water arsenic exceedances in the metamorphic groundwater region are associated with alkaline water (pH range of 7 to 8.5, Fig. 6). Groundwater alkalinity is influenced by such factors as groundwater interaction with calcite-bearing quartz veins (Bottomley, 1984) and carbonate-bearing tills (Utting and Goodwin, 2008). Based on available arsenic and pH well water chemistry data (n=448), approximately 80% of all well water arsenic exceedances in metamorphic aquifers are associated with pH values ranging from 7.2 to 8.2.

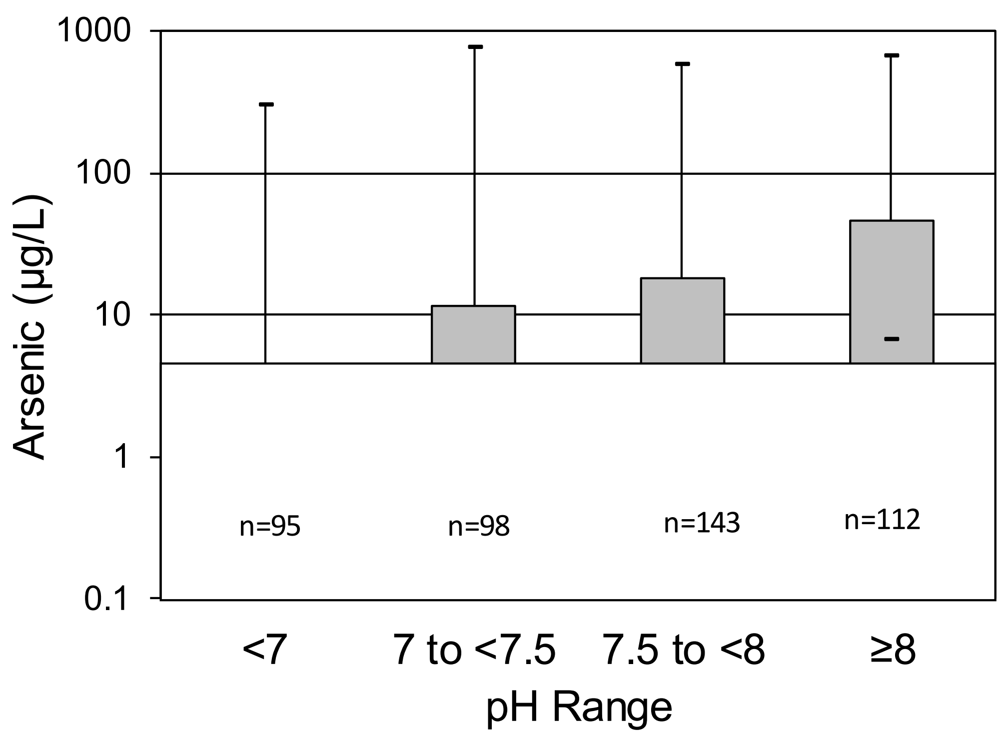

Figure 6. Censored box and whisker plot of arsenic in well water in metamorphic rock aquifers for various pH ranges. The portion of the plot below the maximum detection limit (e.g. 5 µg/L) is not shown.

Delineation of Risk Zones

Goldenville Group rocks were assigned to the high-risk category for arsenic exceeding acceptable levels in well water. The anomalous zones of high arsenic in the Cunard Formation in the Collins Park Subdivision and Cow Bay area were also assigned to the high-risk category. Metavolcanic rock aquifers (e.g. Georgeville, Coxheath, East Bay Hills groups) and the Glen Brook and Acacia Brook formations of the Halifax Group were assigned to the low-risk category, whereas all other Halifax Group rocks and metamorphic rock aquifers in the province were assigned to the medium-risk category.

Improved mapping of the Goldenville and Halifax groups east of Dartmouth would allow for the refinement of risk mapping in this part of the province. There are few available data for metamorphic aquifers located in Antigonish County and on Cape Breton Island, and additional coverage with respect to well water sampling is recommended in these areas to improve the risk analysis.

Plutonic Groundwater Region

Distribution of Arsenic and Summary of Exceedance Rates

Low to high concentrations of arsenic are observed in the plutonic groundwater region of Nova Scotia, although it should be noted that the density of available arsenic in well water data varies considerably across the province, and most well water supplies from plutonic rock aquifers are located in suburban areas of HRM underlain by the South Mountain Batholith (Fig. 7). The overall exceedance rate of the Health Canada MAC for plutonic aquifers is approximately 37%, ranging from 0 to 76% for individual bedrock units (Table 3).

Based on the observed patterns of arsenic in well water concentration, there appears to be a much higher likelihood of arsenic exceeding safe limits in well water in the South Mountain and Musquodoboit batholiths (including the Kinsac pluton) compared to granitoid rocks elsewhere in the province, especially Cape Breton Island (Fig. 7, Table 3).

Relation of Arsenic to Lithogeochemistry

Whole-rock geochemistry analyses of the South Mountain Batholith have returned arsenic concentrations as high as 9000 mg/kg (near Robinsons Corner, Lunenburg County), but median values are typically low at around 2.7 mg/kg (Kennedy and Drage, 2016). In general, higher whole-rock concentrations of arsenic are expected in the South Mountain Batholith due to the strongly peraluminous chemical affinity of these plutons, whereas those in the Antigonish and Cobequid Highlands, Cape Breton Island and the satellite plutons of the South Mountain Batholith have a more calc-alkaline affinity (Table 3).

Enrichment of arsenic in plutonic rocks in Nova Scotia may be related to the digestion of inclusions of Goldenville or Halifax group bedrock (e.g. Clarke et al. 2009), to the formation of mineralized pegmatites or to hydrothermal activity. Elevated concentrations of arsenic can be associated with pegmatite formation because, during the cooling of granitic magmas, late-stage magmatic fluids become enriched with incompatible elements such as arsenic, which are then injected into fractures and country rocks (Peters et al., 1999). Elevated concentrations of arsenic (60 mg/kg) were found in whole-rock analyses of pegmatites from New Hampshire, but much lower values were detected in surrounding schists and granites. Weak-acid leaches of these pegmatites found that about half of the arsenic was labile (Peters et al., 1999). In Nova Scotia, elevated concentrations of arsenic in pegmatites have not been reported, although localized arsenic mineralization has been documented in rocks associated with pegmatites (e.g. Kontak, 1990).

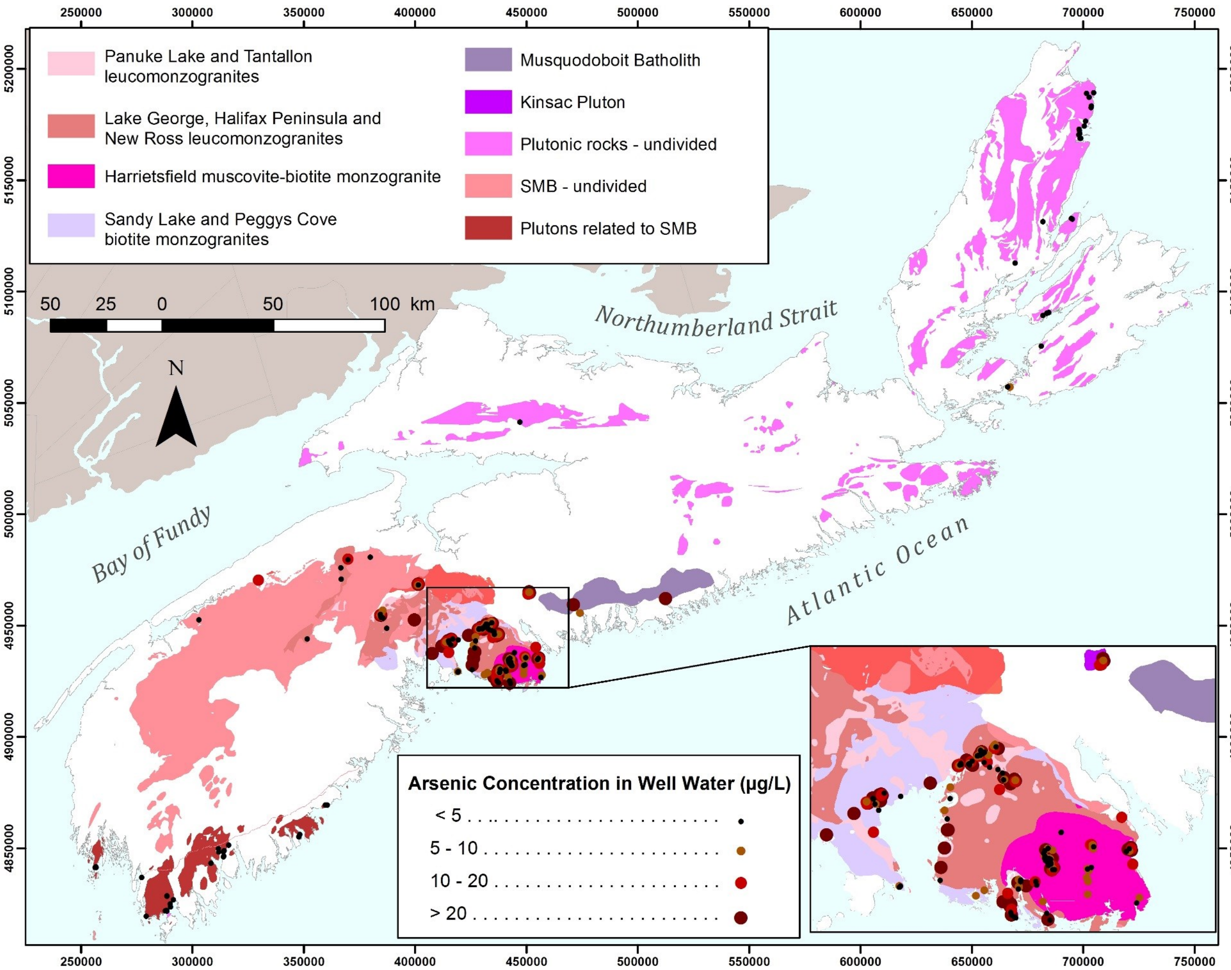

Figure 7. Concentrations of arsenic in well water in the plutonic groundwater region compared to bedrock geology. The South Mountain Batholith is denoted as SMB.

Late-stage hydrothermal activity can also produce enriched levels of arsenic in plutonic rocks because circulating hydrothermal fluids dissolve disseminated arsenic, if present, from surrounding rocks, redistributing it to zones of accumulation where it is precipitated in fractures as arsenopyrite due to changes in the Eh and pH environment (Ryan and Smith, 1998; Peters et al., 1999). The presence of arsenopyrite in quartz veins and greisens in granite is common throughout the South Mountain Batholith (Horne et al., 1988) and has been reported in areas such as East Kemptville (Kontak, 1990), Herring Cove (Kontak and Kyser, 2011) and New Ross (Nova Scotia Department of the Environment, 1978). MacDonald et al. (1992) did not present any clear trends in whole-rock concentrations of arsenic for the six major rock types associated with the South Mountain Batholith; however, average concentrations of arsenic were greatest in the muscovite-biotite monzogranites and smallest in coarse-grained leucomonzogranites and leucogranites.

Relation of Arsenic to Aquifer Geochemistry

Well water surveys of granite aquifers conducted in the 1980s by the Province of Nova Scotia in the communities of Brookside (Harrietsfield muscovite-biotite monzogranite) and New Ross (Panuke Lake and New Ross leucomonzogranite) found that the distribution of elevated arsenic in well water coincided with areas of elevated uranium in well water, which may be due to the similar geochemical behavior of the two elements in groundwater systems, especially with respect to the sensitivity of arsenic mobility to

Table 3. Percentage of water samples exceeding 5 and 10 µg/L of arsenic in well water based on available data for various plutonic bedrock units with at least five samples.

Bedrock Unit	Count	Per cent of As values >5 µg/L	Per cent of As values >10 µg/L
PLUTONIC GROUNDWATER REGION	**305**	**54.1%**	**37.1%**
Undivided Plutonic Rocks - Cape Breton Island	**22**	**0.0%**	**0.0%**
Plutons Related to South Mountain Batholith in Southwest Nova Scotia	**32**	**3.0%**	**0.0%**
Barrington Passage Pluton	10	0.0%	0.0%
Port Mouton Pluton	5	0.0%	0.0%
Shelburne Pluton	9	0.0%	0.0%
Wedgeport Pluton	6	0.0%	0.0%
South Mountain Batholith	**240**	**60.0%**	**40.4%**
Sandy Lake biotite monzogranite	45	82.2%	75.6%
Granodiorite (unnamed biotite granodiorite)	7	85.7%	57.1%
Peggys Cove biotite monzogranite	13	69.2%	61.5%
Tantallon fine to medium grained leucomonzogranite	49	63.3%	40.8%
Halifax Peninsula coarse grained leucomonzogranite	38	63.2%	39.5%
New Ross coarse grained leucomonzogranite	11	54.6%	27.3%
Harrietsfield muscovite-biotite monzogranite	63	55.2%	26.9%
Musquodoboit Batholith and Kinsac Pluton	**10**	**100.0%**	**80.0%**

pH (MacFarlane, 1983). Arsenic exceedances in plutonic rock aquifers are associated with mildly alkaline water with a pH range of about 7 to 8 (Fig. 8), and therefore subsurface conditions that tend to raise the pH of groundwater (e.g. thicker overlying till, presence of carbonate minerals) also increase the mobility of arsenic. For this reason, some of the previous well water chemistry surveys conducted by the Province (e.g. Hubbards, New Ross, Brookside) have correlated arsenic concentration in water wells with depth to bedrock (Nova Scotia Department of the Environment, 1978; MacFarlane, 1983). For example, correlation of the concentration of arsenic in well water with depth to bedrock (where corresponding water well records could be identified) in the communities of Hubbards and Brookside shows a weak positive correlation (Fig. 9). This relationship, however, is not observed consistently across the plutonic groundwater region and may be confounded by other factors. In the Hubbards area, for example, it was also suggested that the bedrock troughs may coincide with fault-controlled mineralized zones (e.g. Hubbards Cove Fault) (Nova Scotia Department of the Environment, 1978).

Delineation of Risk Zones

Based on the available data, most of the South Mountain and Musquodoboit batholiths, and other mainland granitoid rocks, were assigned to the high-risk category, whereas all other plutonic rock units in the province were assigned to the low-risk category. There are large gaps, however, in terms of data coverage (Fig. 7), and additional well water sampling is recommended in these areas to refine the risk map zones.

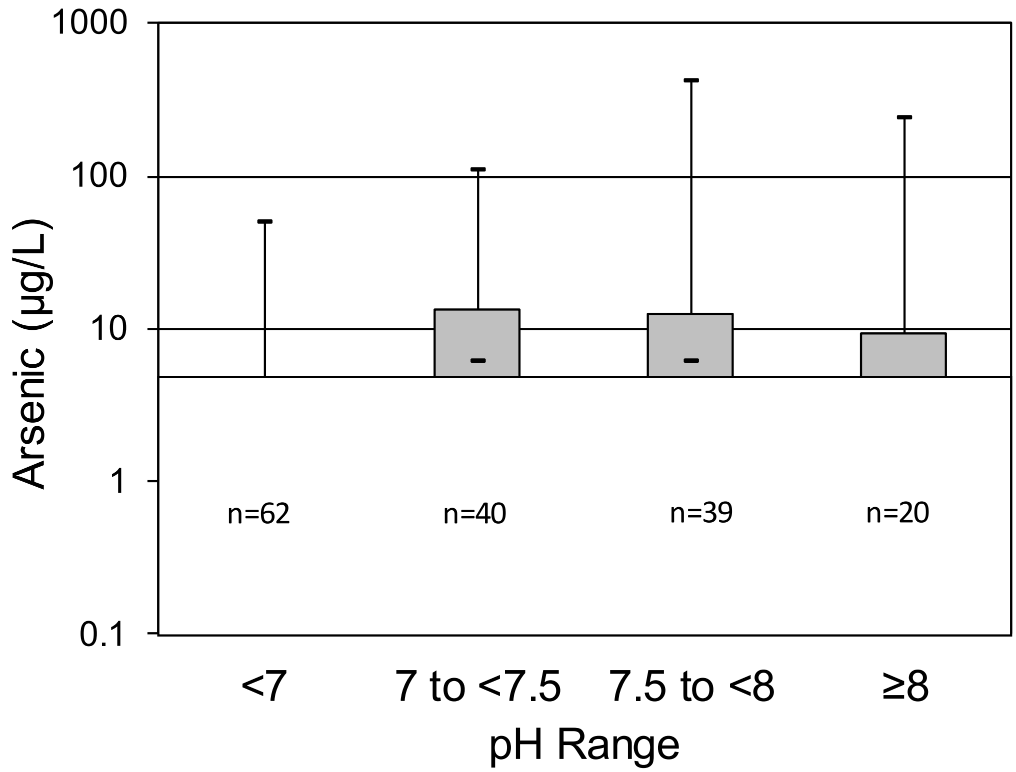

Figure 8. Censored box and whisker plot of arsenic in well water in plutonic rock aquifers for various pH ranges. The portion of the plot below the maximum detection limit (e.g. 5 µg/L) is not shown.

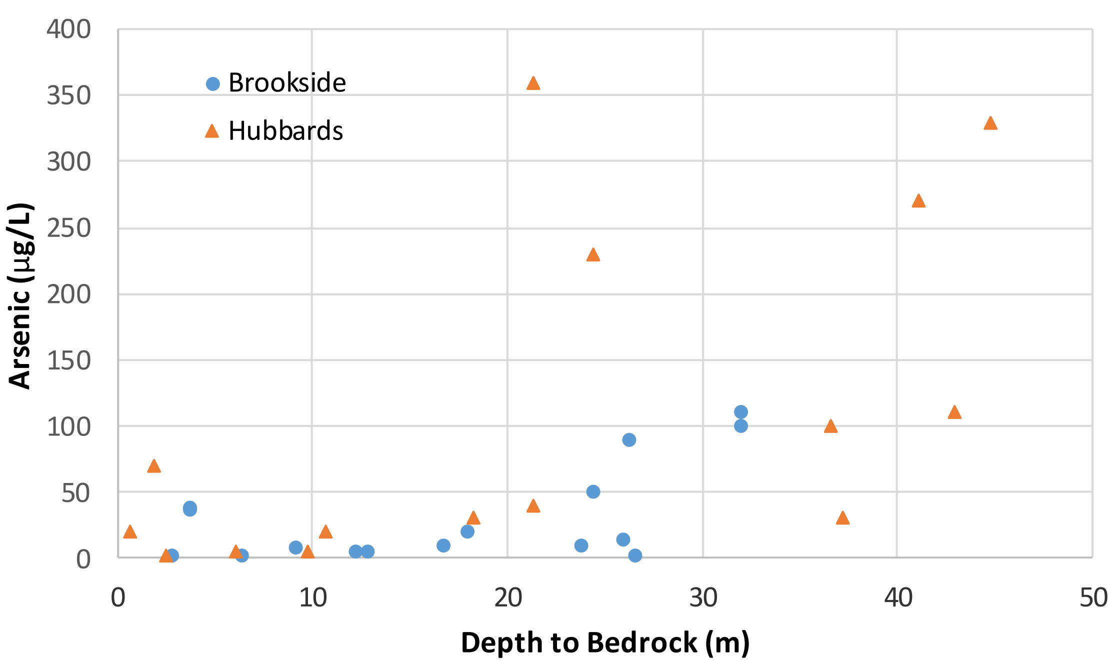

Figure 9. Correlation of depth to bedrock with well water concentrations for the communities of Brookside and Hubbards.

Sedimentary Groundwater Region

Distribution of Arsenic and Summary of Exceedance Rates

Low to moderate concentrations of arsenic are generally observed in the sedimentary groundwater region of Nova Scotia (Fig. 10). The overall exceedance rate of the 2017 Health Canada MAC for sedimentary aquifers is about 5%, ranging from 0 to 38% for individual bedrock units (Table 4). The highest exceedance rates for arsenic in well water are associated with the New Glasgow, Cheverie, Tracadie Road, Clam Harbour River, Pomquet and Hastings formations.

Relation of Arsenic to Lithogeochemistry

There are limited data available on whole-rock concentrations of arsenic in sedimentary bedrock in Nova Scotia, although a review by Kennedy and Drage (2016) reported published average concentrations in the 1 to 15 mg/kg range. In the Mesozoic Newark Basin, located in the northeast United States, enriched arsenic has been associated with both black and gray shale beds of the Lockatong Formation and the younger red mudstone and siltstone beds of the Passaic Formation (Serfes, 2005). Arsenian pyrite was determined to be a major source of arsenic in the shales (up to 240 mg/kg),

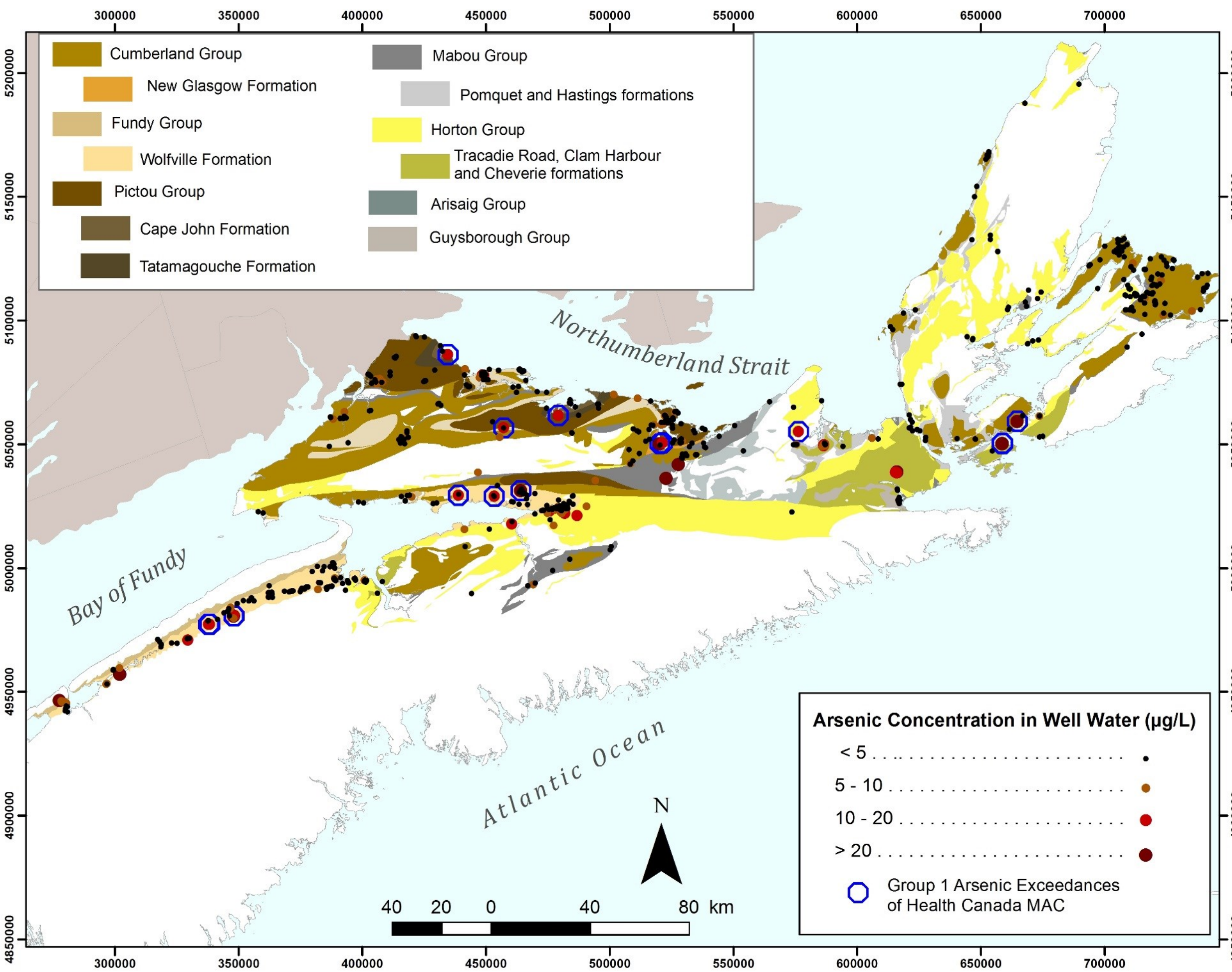

Figure 10. Concentrations of arsenic in well water in the sedimentary groundwater region compared to bedrock geology. Group 1 type exceedances of arsenic in well water shown on the map are associated with sodium bicarbonate type water, low hardness and elevated pH.

Table 4. Percentage of water samples exceeding 5 and 10 µg/L of arsenic in well water based on available data for various sedimentary bedrock units with at least five samples.

Bedrock Unit	Count	Per cent of As values >5 µg/L	Per cent of As values >10 µg/L
SEDIMENTARY GROUNDWATER REGION	**763**	**15.6%**	**4.7%**
Cumberland Group	**293**	**12.3%**	**2.0%**
New Glasgow Formation	9	33.3%	22.2%
Port Hood Formation	28	3.6%	3.6%
Malagash Formation	95	24.2%	3.2%
Joggins Formation	7	14.3%	0.0%
Boss Point Formation	7	14.3%	0.0%
Springhill Mines Formation	16	12.5%	0.0%
South Bar Formation	67	7.5%	0.0%
Ragged Reef Formation	5	0.0%	0.0%
Stellarton Formation	15	0.0%	0.0%
Sydney Mines Formation	28	0.0%	0.0%
Waddens Cove Formation	9	0.0%	0.0%
Fundy Group	**223**	**15.7%**	**4.9%**
Wolfville Formation	204	14.2%	5.4%
Blomidon Formation	13	38.5%	0.0%
McCoy Brook Formation	6	16.7%	0.0%
Guysborough Group	**8**	**0.0%**	**0.0%**
Glenkeen Formation	6	0.0%	0.0%
Horton Group	**132**	**17.4%**	**9.1%**
Clam Harbour River Formation	8	37.5%	37.5%
Tracadie Road Formation	9	44.4%	33.3%
Cheverie Formation	13	69.2%	30.8%
Horton Bluff Formation	26	23.1%	3.8%
Ainslie Formation	6	0.0%	0.0%
Grantmire Formation	54	0.0%	0.0%
Mabou Group	**52**	**28.8%**	**9.6%**
Pomquet Formation	17	52.9%	17.6%
Hastings Formation	13	38.5%	15.4%
Lismore Formation	7	0.0%	0.0%
Pictou Group	**50**	**18.0%**	**4.0%**
Tatamagouche Formation	5	20.0%	20.0%
Cape John Formation	7	14.3%	14.3%
Balfron Formation	37	16.2%	0.0%

whereas hematite and clay minerals (up to 14.8 mg/kg) were inferred to be a possible source of arsenic in the red beds. In New Brunswick (Ordovician Tetagouche Group) and other areas of New England (e.g. Silurian Waterville Formation), it is predominantly black shale units that have been associated with elevated whole-rock concentrations of arsenic (Klassen et al., 2009; Ryan et al., 2015). The sedimentary rock units associated with the highest arsenic in well water exceedance rates in Nova Scotia (e.g. New Glasgow, Cheverie, Tracadie Road, Clam Harbour River, Pomquet and Hastings formations) all locally contain significant beds of black carbonaceous material (e.g. Ténière et al., 2005), although there are few data on arsenic concentrations in these rocks. The compilation of a database of whole-rock geochemistry for arsenic for various sedimentary units/rock types would provide an improved understanding of the source of arsenic in sedimentary bedrock aquifers.

Relation of Arsenic to Aquifer Geochemistry

Aquifer geochemistry appears to have an important role in controlling the mobilization of arsenic from sedimentary bedrock. In general, a relationship between well water arsenic concentration and laboratory pH can be observed in sedimentary bedrock aquifers, especially at pH greater than 8.5, with arsenic levels in water wells increasing with pH (Fig. 11). More alkaline groundwater is observed in some sedimentary bedrock aquifers compared to crystalline bedrock aquifers (Fig. 6 and 8), and there appears to be a shift in the pH-arsenic relationship. Based on the available data, most (60%) of the highest concentrations of arsenic (e.g. >25 µg/L) in sedimentary aquifers are associated with pH levels above 8.5 (n = 10), whereas there are no exceedances of the Health Canada MAC for arsenic in crystalline bedrock aquifers at pH levels above this threshold.

Where arsenic in well water is elevated in sedimentary bedrock aquifers, two dominant geochemical facies are evident, which can be grouped as follows:

- Group 1: Sodium bicarbonate ($NaHCO_3$)– or sodium bicarbonate chloride–type waters with elevated pH (e.g. >8.5) and low hardness (e.g. <50 mg/L) found in localized areas of various sedimentary rock types (Fig. 10). Water samples in this group also tend to have high sodium-calcium and sodium-chloride ratios.
- Group 2: Select sedimentary geological units (e.g. Mabou and Horton group bedrock units) associated with stratification (alternating red and grey sedimentary layers) indicating a variable redox depositional environment. Occurrences of elevated arsenic for this group tend to be concentrated along bedrock contacts (Fig. 10) and the associated water chemistry generally consists of calcium bicarbonate ($CaHCO_3$)–type water with comparatively lower pH (7–8.5) and higher hardness (e.g. >100 mg/L).

Group 1 There is no clear association of the Group 1 geochemical facies with a particular sedimentary geological unit in Nova Scotia, but rather this type of water is detected in water wells intercepting various sedimentary geological units, including the Wolfville Formation (Fundy Group), the Tracadie Road Formation (Horton Group), the New Glasgow Formation (Cumberland Group) and the Cape John Formation (Pictou Group) (Fig. 10). Approximately 50% of the exceedances for arsenic in well water (>10 µg/L) from sedimentary bedrock aquifers are associated with Group 1 groundwater (where a corresponding major-ion analysis is also available). In Figure 12, boxplots of pH, hardness and arsenic are compared for Group 1 well water samples (where the concentration of arsenic > 5 µg/L) (n=23) and all other well water samples collected from sedimentary bedrock aquifers (n=709).

Previous regional groundwater studies in Pictou, Cumberland and Cape Breton counties have noted the occurrence of $NaHCO_3$ groundwater, especially in finer grained (e.g. argillaceous) sedimentary rocks (Gibb and McMullin, 1980; Vaughan and Somers, 1980; Baechler, 1986). Vaughan and Somers (1980)

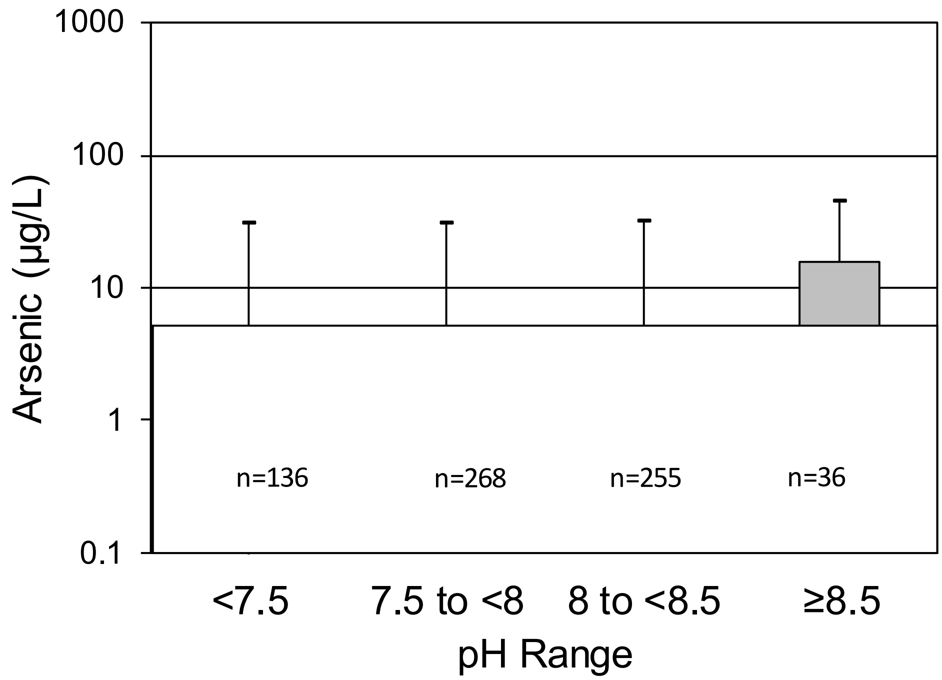

Figure 11. Censored box and whisker plot of arsenic in well water in sedimentary rock aquifers for various pH ranges. The portion of the plot below the maximum detection limit (e.g. 5 µg/L) is not shown.

observed that shale sequences of Mabou, Pictou and Cumberland group rocks can produce highly alkaline and very soft groundwater containing high pH, a wide range of chloride salts, and high total dissolved solids (TDS) concentrations. The soft character of the water was attributed to cation exchange, whereby sodium ions (which do not contribute to hardness) from the shale sediment are exchanged with magnesium and calcium ions in groundwater. Similarly, Baechler (1986) related $NaHCO_3$ groundwater with fine-grained sediments in the Cumberland Group in Cape Breton, and attributed the soft, alkaline character of the water to a natural softening process where calcium ions are replaced by sodium ions. Few details are presented in these studies, however, that might explain the source of both the sodium and elevated groundwater alkalinity.

Other studies (e.g. Cerling et al., 1989; Ayotte et al., 2003) have reported that chemical weathering of rocks containing a large number of sodium ions adsorbed onto ion exchange sites due to deposition under marine conditions (or affected by marine incursion following deposition) by generally calcium-rich freshwater recharge results in the preferential adsorption of calcium and the release of sodium. Ayotte et al. (2003) further suggested that this ion exchange mechanism promotes the dissolution of calcite, resulting in a pH increase and a corresponding increase in arsenic mobility in New England metasedimentary rock aquifers. Many of the Group 1 type occurrences of elevated arsenic in Nova Scotia well water, however, do not correspond to rocks that were deposited under marine conditions.

Another mechanism that might explain the association of elevated arsenic with $NaHCO_3$ type water involves membrane filtration. White (1965) and Van Everdingen (1968) suggested that lower permeability clay-rich layers, such as shales (argillaceous rock), can behave similarly to reverse osmosis units in the groundwater flow field, increasing concentrations of larger ions on the hydraulically upgradient (high concentration) side of the low permeability unit. The clay minerals therefore act as a filter, permitting ions of sufficiently small diameter (e.g. Na^+) and bicarbonate molecules to pass through; however, larger ions, such as Ca^{2+} an Cl^-, are rejected. Hence the concentration of salts in the groundwater passing through the argillaceous layer is diluted, reducing the amount of chloride, calcium and magnesium in the groundwater while enriching it with sodium and bicarbonate ions. White (1965) expected H^+ to move

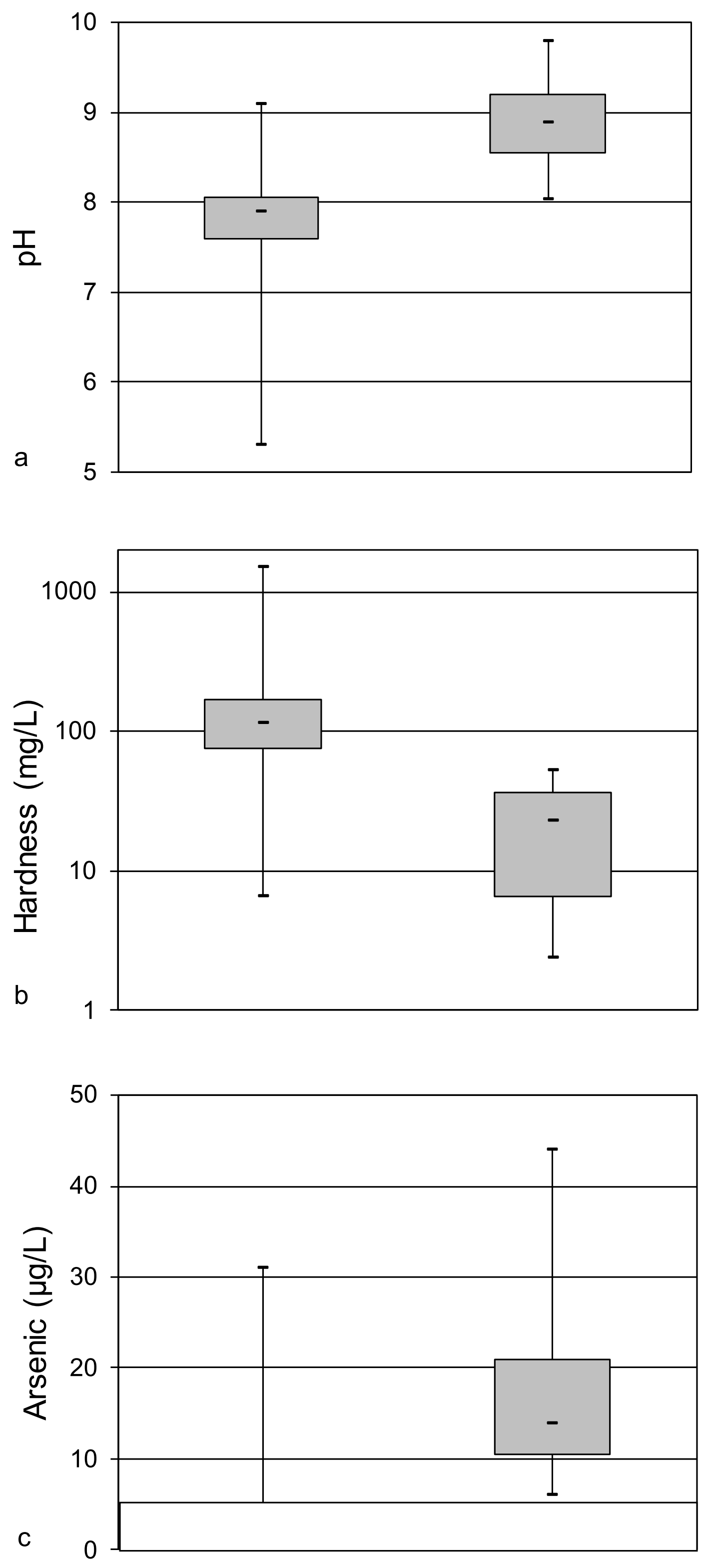

Figure 12. Box and whisker plot of (a) pH, (b) hardness and (c) arsenic for Group 1 type exceedances of arsenic in well water compared to all other data on arsenic in well water from sedimentary aquifers.

from the low-concentration side to the high-concentration side of the semi-permeable membrane to compensate for the electrical potentials created by the movement of Na^+ to the low-concentration side. This movement of H^+ may result in more alkaline conditions at the low-concentration side of the barrier. Given the sensitivity of arsenic mobilization to pH, the increase in pH on the low concentration side of the membrane may cause a concomitant release of arsenic hosted in sedimentary bedrock. Several studies have also shown that arsenic mobility is sensitive to the bicarbonate ion, which can mobilize arsenic either from sulphide minerals (Kim et al., 2000) by forming aqueous $As\text{-}HCO_3$ complexes or from iron/manganese oxyhydroxides through substitution (Appelo et al., 2002; Anawar et al., 2004).

The available water well stratigraphy information is inadequate for correlating Group 1 type occurrences of elevated arsenic with bedrock stratigraphy, although it should be noted that this geochemical facies has been detected in areas underlain by sedimentary bedrock units with a relatively low percentage of fine-grained shales (e.g. Wolfville Formation). Several possibilities that might explain the Group 1 elevated levels of arsenic in sedimentary bedrock aquifers are presented herein; however, it is evident that additional hydrogeochemical testing is needed to determine which mechanism best explains the observed patterns.

Group 2 The other dominant grouping of elevated arsenic in sedimentary bedrock aquifers appears to be associated with bedrock units belonging to the Clam Harbour River, Tracadie Road and Cheverie formations of the Horton Group and the Pomquet and Hastings formations of the Mabou Group, especially near bedrock contacts (Fig. 10). This group accounts for approximately 74% (n=14) of the remaining exceedances for arsenic in well water (i.e. excluding Group 1 type exceedances) from sedimentary bedrock aquifers, despite representing only 12% of all of the water sample locations in sedimentary bedrock aquifers (n=734). Group 2 type occurrences of elevated arsenic tend to be associated with $CaCO_3$ type water, which is representative of most sedimentary bedrock aquifers across the province. Bedrock stratigraphy from these formations is characterized by alternating red (oxidized) and grey (reduced) sandstone and siltstone beds, suggesting variable redox conditions and a potential redox control on arsenic occurrence in well water. The grey beds are characterized by organic-rich, pyritic deep-water lake strata and more anoxic conditions compared to the red beds. The destabilization of metal oxide surfaces as a function of changing redox potential could result in the desorption and mobilization of arsenic from these surfaces. The proximity of arsenic exceedances to bedrock contacts may also indicate changing geochemical conditions along the groundwater-flow path and a potential redox control on arsenic mobilization in sedimentary bedrock aquifers. Measurement of vertical and lateral (across bedrock contacts) geochemical gradients is needed in these types of aquifers to better understand the role of redox conditions on arsenic mobilization.

Delineation of Risk Zones

A different approach to assigning risk zones was employed for Group 1 (e.g. $NaHCO_3$ type groundwater) exceedances of the Health Canada MAC compared to the province's other groundwater regions, where geological boundaries were used to define risk zones. Group 1 type occurrences of elevated arsenic in well water appear to be localized, making it difficult to delineate zones where this geochemical facies is present. To capture Group 1 type occurrences of elevated arsenic in the risk zones, an arbitrary buffer of 500 m was generated around each location of an identified Group 1 exceedance of the Health Canada MAC, and the resulting polygon was assigned to the high-risk category.

Bedrock units associated with Group 2 occurrences of elevated arsenic in well water (i.e. Clam Harbour River, Tracadie Road and Cheverie formations of the Horton Group and the Pomquet and Hastings formations of the Mabou Group), the New Glasgow Formation (Cumberland Group) and the Tatamagouche Formation (Pictou Group) were also assigned to the high-risk category. The Wolfville Formation (Fundy Group) and Cape John Formation (Pictou Group) were assigned to the medium-risk category and all other sedimentary bedrock units were assigned to the low-risk category.

Carbonate/Evaporite Groundwater Region

Distribution of Arsenic and Summary of Exceedance Rates

Low to moderate concentrations of arsenic are observed in the carbonate/evaporite groundwater region of Nova Scotia (Fig. 13). The overall exceedance rate of the Health Canada MAC for carbonate/evaporite aquifers is about 4%, ranging from 0 to 30% for individual bedrock units (Table 5), which is similar to the exceedance rate and range of per cent exceedances reported for the sedimentary groundwater region (Table 4).

The highest exceedance rate of the Health Canada MAC was observed in the Pugwash Mines Formation, near the contact with the Malagash Formation of the Cumberland Group (Table 5, Fig. 13). Elevated concentrations of arsenic were also detected along the Windsor Group–Goldenville Group contact in central Nova Scotia (Fig. 13).

Relation of Arsenic to Aquifer Geochemistry and Lithogeochemistry

Average concentrations of arsenic in carbonate/evaporite rocks in the Walton area of the Carboniferous Windsor Basin published by Boyle and Jonasson (1973) ranged from 2 to 5 mg/kg. Few other published surveys of arsenic whole-rock geochemistry for these rocks in Nova Scotia could be located.

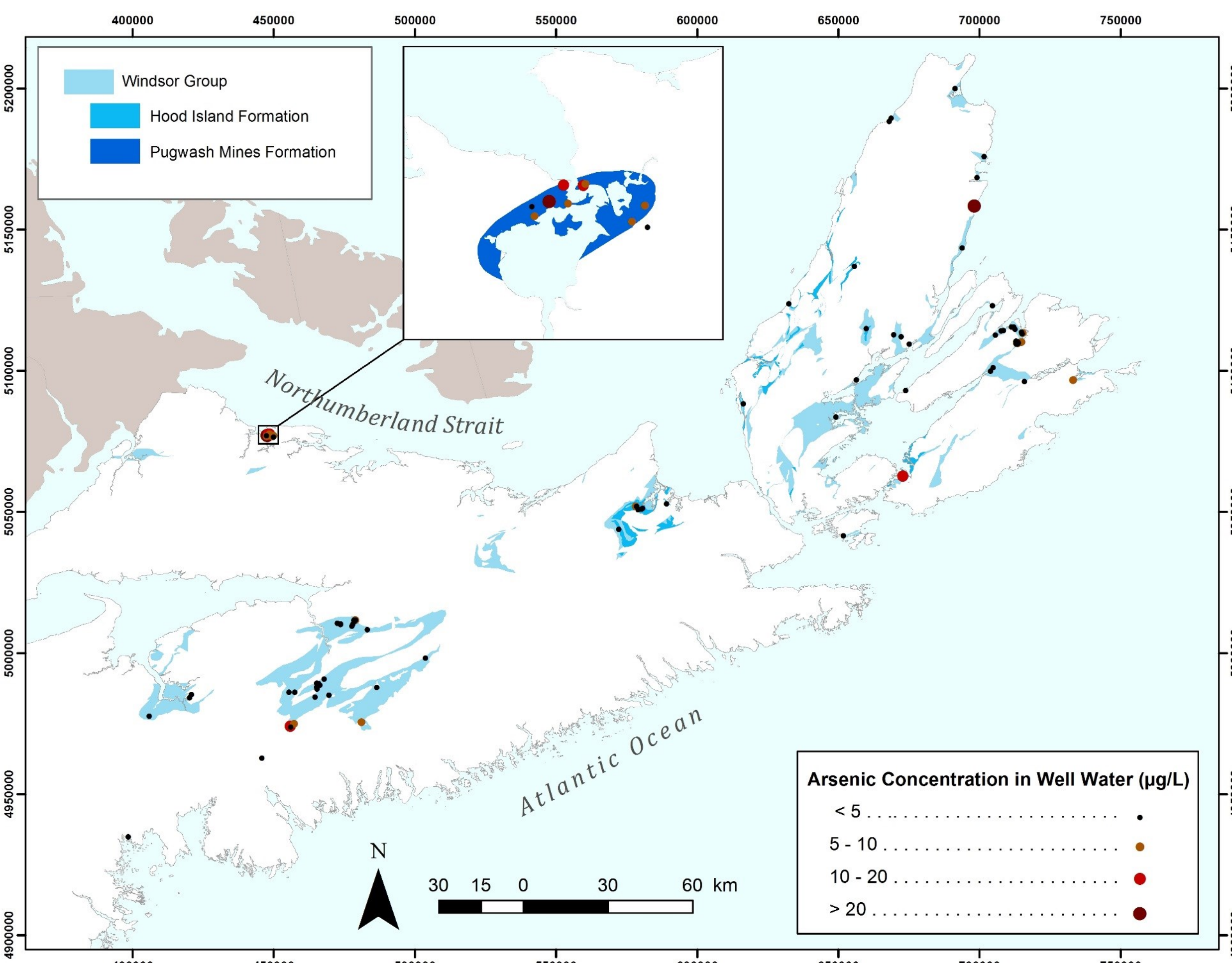

Figure 13. Concentrations of arsenic in well water in the carbonate/evaporite groundwater region compared to bedrock geology.

Table 5. Percentage of water samples exceeding 5 and 10 µg/L of arsenic in well water based on available data for various carbonate/evaporite bedrock units with at least five samples.

Bedrock Unit	Count	Per cent of As values >5 µg/L	Per cent of As values >10 µg/L
CARBONATE/ EVAPORITE GROUNDWATER REGION (Windsor Group)	**141**	**15.6%**	**4.3%**
Pugwash Mines Formation	10	80.0%	30.0%
Hood Island Formation	13	15.4%	7.7%
Woodbine Road Formation	10	20.0%	0.0%
Macdonald Road Formation	5	20.0%	0.0%
Meadows Road Formation	21	14.3%	0.0%
Sydney River Formation	36	2.8%	0.0%
Green Oaks Formation	20	0.0%	0.0%
Mainland Nova Scotia - all	**54**	**24.1%**	**7.4%**
Cape Breton Island - all	**87**	**10.3%**	**2.3%**

An exploration of the geochemical controls of arsenic occurrence in carbonate/evaporite aquifers is constrained by the small size of the data set compared to sedimentary aquifers. There are fewer data due to the smaller extent of these aquifers and because dug wells are often preferred in areas underlain by carbonate/evaporite rocks due to the unsuitable water quality (e.g. high sulphates, hardness and TDS) of bedrock aquifers for water supply. It is expected, however, that the shale-rich bedrock units of the Windsor Group would behave similarly with respect to arsenic occurrence and mobilization as the Group 2 type occurrences of elevated arsenic in well water described in the preceding section.

It should also be noted that well water samples collected from the relatively soft Windsor Group rocks can have a high amount of particulates, depending on the rock type and sampling protocol. When a total-water digest is performed during sample analysis, especially for recently completed wells that may not have been fully developed, elevated arsenic values can be detected that may not be representative of routine well operation.

Delineation of Risk Zones

Based on the exceedance rates presented in Table 5, the Pugwash Mines Formation was assigned to the high-risk category, and the Hood Island Formation was assigned to the medium-risk category. All other Windsor Group rocks were assigned to the low-risk category for arsenic in well water.

Volcanic Groundwater Region

Distribution of Arsenic and Summary of Exceedance Rates

Low concentrations of arsenic are typically found in the volcanic groundwater region of Nova Scotia (Fig. 14), and based on the available data, no arsenic in well water samples exceeding the Health Canada MAC of 10 µg/L were reported in volcanic rock aquifers (Table 6).

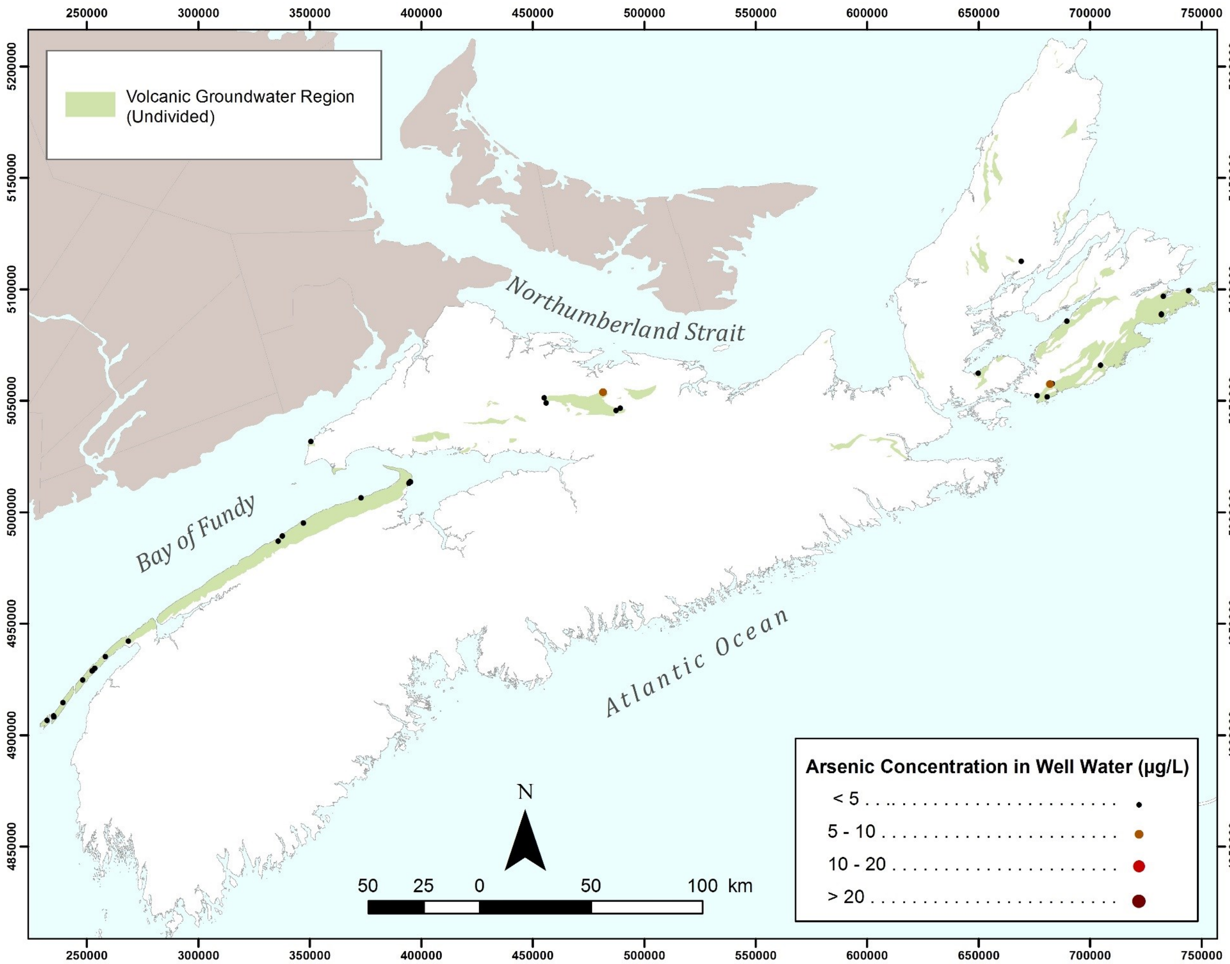

Figure 14. Concentrations of arsenic in well water in the volcanic groundwater region compared to bedrock geology.

Relation of Arsenic to Aquifer Geochemistry and Lithogeochemistry

Boyle and Jonasson (1973) reported a concentration of <1 mg/kg arsenic for a sample of basalt collected from the North Mountain Formation. Few published regional surveys with geochemical data for arsenic in volcanic rocks are available, although some of the recent work in the Cobequid Mountains (Nova Scotia Department of Natural Resources, unpub. data, 2016) have shown whole-rock arsenic concentrations up to 72 mg/kg in mineralized zones, and levels generally less than 5 mg/kg in country rock. Similar arsenic concentrations have been detected in volcanic rock hosted mineral deposits in Cape Breton Island (Nova Scotia Department of Natural Resources, unpub. data, 2016).

Although favourable conditions for arsenic mobilization could develop in volcanic rock aquifers, groundwater concentrations of arsenic in most volcanic rocks in Nova Scotia are likely limited by the low levels of arsenic in the host bedrock.

Delineation of Risk Zones

Since the available data shows that there are no exceedances for arsenic in well water from volcanic rock aquifers in Nova Scotia, these bedrock units were categorized as low risk for arsenic in well water exceeding the Health Canada MAC. There are few well water chemistry results available, however,

Table 6. Percentage of water samples exceeding 5 and 10 µg/L of arsenic in well water based on available data for various volcanic bedrock units with at least five samples.

Bedrock Unit	Count	Per cent of As values >5 µg/L	Per cent of As values >10 µg/L
VOLCANIC GROUNDWATER REGION	39	5.1%	0.0%
Fountain Lake Group	6	20.0%	0.0%
Fourchu Group	5	0.0%	0.0%
Fundy Group (North Mountain Formation)	21	0.0%	0.0%

which is partly due to the low population density of areas underlain by volcanic aquifers. Additional sample coverage is recommended to address this data gap, especially in Cape Breton Island and the Cobequid and Antigonish highland volcanic rock aquifers.

Arsenic in Well Water Risk Map for Bedrock Aquifers and Demographics of Risk

The Nova Scotia Department of Health (1983) produced the first arsenic in well water risk map for the province, identifying areas of Nova Scotia that are most likely to have elevated levels of arsenic in groundwater, with the 'most likely' category generally corresponding to the bedrock units of the Goldenville and Halifax groups. This map was later refined in 2005 by the Arsenic Working Group (Nova Scotia Environment, 2005) based on arsenic concentration data for various media types, and divided the province into areas that are 'very likely' and 'likely' to have elevated levels of arsenic in well water (Fig. 2).

A new risk map, the main objective of the current study, is presented in Figure 15. The risk map is based on the exceedance rates of arsenic in well water (compared to the 2017 Health Canada MAC) for various bedrock units (Tables 2 to 6), dividing the province into low- (<5% exceeding), medium- (5 to <15% exceeding) and high- (≥15% exceeding) risk zones.

Statistical comparison of the sample populations intersecting each of the three zones using the Kruskal-Wallis non-parametric test found a significant difference ($p<0.05$) between the three categories of risk. The high-risk category covers the largest area of the province (Table 7) and captures approximately 95% of the exceedances of the Health Canada MAC reported in the study dataset (Table 7). Comparison of the risk zones with arsenic in well water data that were rejected due to insufficient information regarding the aquifer type (e.g. drilled vs. dug well) or location (e.g. inadequate location accuracy) shows similar efficacy in terms of capturing exceedances in the highest risk category (Table 7), although it should be noted that these 'rejected' data are highly concentrated in suburban areas of HRM. The revised risk map offers significant refinement compared to the older version, which had only two categories of risk. For example, 18% of arsenic results ≤2.5 µg/L are within the highest risk zone in the revised map, compared to 48% in the 2005 version.

A count of households using private wells in each of the three risk zones shows a relatively similar distribution, (Fig. 15), although the high-risk zone comprises the largest area and has the highest percentage (37% or ~80,000 households) of the province's private well users. The ten communities with

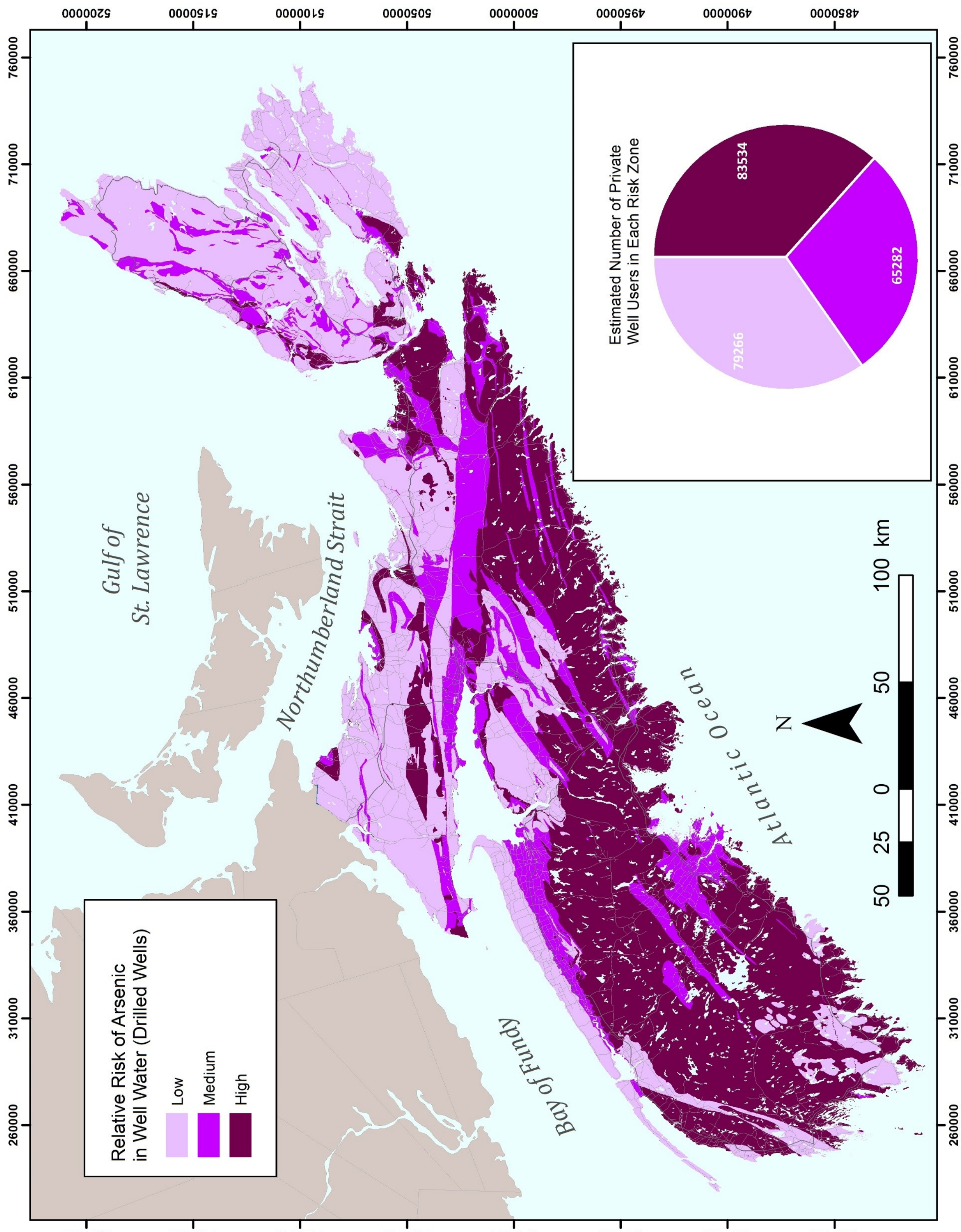

Figure 15. Risk map for arsenic in well water from bedrock aquifers in Nova Scotia. The estimated number of private well users in each risk zone is also shown.

Table 7. Demographics of the three risk zones, including area, estimated number of unserviced households and per cent of arsenic in well water exceedances of the Health Canada MAC captured in the risk zone. In both the accepted and rejected datasets there were approximately 500 samples exceeding 10 µg/L of arsenic in well water. The demographic analyses assumes that 90% of private well users are supplied by bedrock aquifers.

Risk Classification	Area of Coverage (km^2)	Per cent of Samples >10 µg/L (n = 505; Accepted Data)	Per cent of Samples >10 µg/L (n = 516, Rejected Data)	Count of Unserviced Civic Addresses
High	24,905	95%	92%	85,534
Medium	9,113	4%	7%	65,282
Low	21,061	1%	1%	79,266

the largest number of households using private wells are underlain by bedrock of the Goldenville Group or South Mountain Batholith (Table 8) and are situated in suburban HRM. It is estimated that wells intercepting the Goldenville Group or South Mountain Batholith account for 24% of private well users in the province. Using the exceedance rates estimated for the five groundwater regions (Tables 2–6) and the distribution of private wells relative to the groundwater regions, the percentage of private well users with arsenic exceeding the acceptable limit of 10 µg/L may be as high as 20% (~90,000 persons) across Nova Scotia (Fig. 16).

Summary

Bedrock geology is the most important provincial-scale control on the distribution of arsenic concentrations in well water. Both the highest concentrations (i.e. >1000 µg/L) and exceedance rates (37–39%) were observed in the metamorphic and plutonic groundwater regions of southern Nova Scotia, largely due to the presence of arsenic mineralization in these rocks. In particular, water well samples

Table 8. The ten communities with the largest number of private well users located in a high-risk zone for arsenic in well water.

Community	Underlying High-Risk Bedrock Unit	County	Count of Unserviced Civic Addresses
Hammonds Plains	Goldenville Group and SMB Granites	Halifax	1521
Upper Tantallon	SMB Granites	Halifax	1430
Porters Lake	Goldenville Group	Halifax	1370
Sackville	Goldenville Group	Halifax	1104
Lawrencetown	Goldenville Group	Halifax	1074
Stillwater Lake	SMB Granites	Halifax	853
Hubley	SMB Granites	Halifax	794
Lake Echo	Goldenville Group	Halifax	770
Brookside	SMB Granites	Halifax	729
Head of St. Margarets Bay	SMB Granites	Halifax	706

SMB: South Mountain Batholith

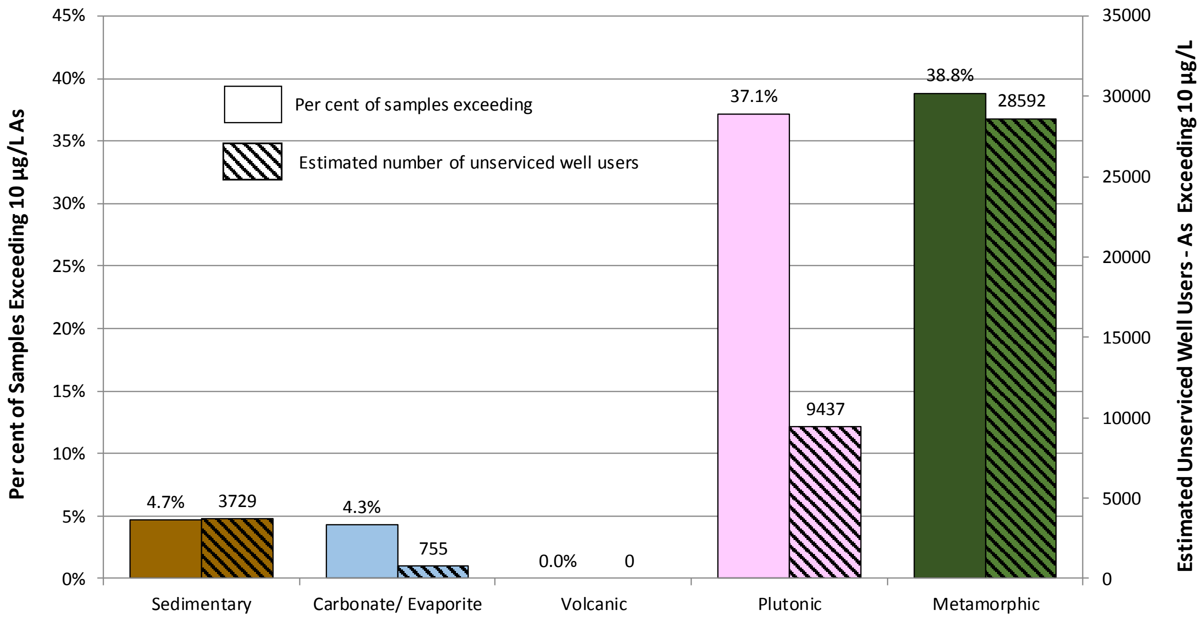

Figure 16. Exceedance rates of Health Canada's maximum acceptable concentration (MAC) of 10 µg/L in drinking water for Nova Scotia's five major groundwater regions. Based on these exceedance rates, the number of affected private well owners (households) in each groundwater region is estimated.

collected from Goldenville Group and South Mountain Batholith bedrock aquifers were associated with the highest frequency of arsenic exceeding the safe limits for drinking water.

Generally, both whole-rock and well water concentrations of arsenic are much lower in sedimentary aquifers compared to crystalline aquifers, and groundwater concentrations of sedimentary aquifers appear to be more strongly influenced by geochemical conditions. Based on the available data, well water supplies in carbonate/evaporite aquifers exceed the Health Canada MAC at a similar rate to sedimentary aquifers (4–5%), however, these aquifers host comparatively fewer private and public water supplies. There are no recorded exceedances of arsenic in well water for volcanic rock aquifers. Favourable conditions for arsenic mobilization could develop in these aquifers, however, well water concentrations of arsenic from these aquifers are likely limited by the low levels of arsenic in the host bedrock.

In addition to arsenic concentrations in the source rock, the study highlighted several examples of other potentially important controls on arsenic occurrence and mobility including metamorphic grade, contact metamorphism and geochemical conditions. The focus of the present analysis with respect to geochemical controls on arsenic mobility was the influence of pH, and it was generally found that elevated levels of arsenic in groundwater were associated with a pH range of 7 to 8, except in sedimentary aquifers with $NaHCO_3$ type water, where elevated arsenic was associated with pH > 8. Both Eh and pH were identified as key controls on the mobilization of arsenic in sedimentary aquifers, and the observed patterns suggest that the aquifer geochemistry is influenced by sedimentary stratigraphy.

Although the present analysis demonstrated regional-scale trends, the distribution of arsenic in well water shows significant spatial heterogeneity. At the local scale, patterns of elevated arsenic are difficult to predict and are likely controlled by heterogeneities with respect to the groundwater flow path (e.g. fracture flow, recharge vs. discharge) and groundwater interaction with geological media (e.g. till thickness, mineralized veins in contact with water-bearing fractures, stratigraphy).

The revised arsenic in well water risk map (Fig. 15) developed during this study relies on observed exceedance rates of arsenic in well water samples for over 60 individual bedrock units, dividing the province into low-, medium- and high-risk zones. It is estimated that 37% (Fig. 15) of the province's private well users are in the high-risk zone for arsenic exceeding the Health Canada MAC, and the overall proportion of private wells with arsenic exceeding the Health Canada MAC may be as high as 20% (~90,000 persons). The communities surrounding the urban core of HRM, where residential growth is concentrated, have the greatest density of private well water use. Groundwater in these communities is obtained from crystalline rock aquifers such as the Goldenville Group and the South Mountain Batholith, which have the highest rate of arsenic in well water exceedances of the Health Canada MAC and provide domestic water to approximately a quarter of Nova Scotia's private well users.

According to a risk-perception study by Chappells et al. (2014), existing risk communication efforts are largely failing to reach target audiences, the quality of the information presented through formal channels is below average, and there is poor compliance with recommended standards for private well-water monitoring. The new risk map offers a more refined characterization of risk across Nova Scotia compared to the previous version, and is better aligned with recent provincial risk-map products (e.g. radon in indoor air), which also include three levels of risk (Nova Scotia Department of Natural Resources, 2016b). The risk zones, however, should be subject to continuous evaluation as new groundwater chemistry data becomes available. The arsenic in well water risk map will serve as a foundational tool to raise risk awareness among private well users in Nova Scotia. Effective public awareness is essential to manage the risk of arsenic exposure and prevent disease.

The arsenic in well water risk map will be published as a web-map application that will target private well owners, making it easier to access information about risk levels and ways to mitigate arsenic in well water. In conjunction with the development of a new web-map application, relevant public-facing webpages and fact sheets will also be updated with the aim of encouraging well owners to regularly (once every two years) test their water.

Further Research

Several knowledge gaps and anomalies were identified during the preparation of the risk map. Additional targeted research would help to further develop the understanding of the distribution of, and the various hydrogeochemical controls on, arsenic in well water in Nova Scotia. For example, detailed borehole-scale research in various types of rocks associated with elevated arsenic would provide a more thorough understanding of source of arsenic in the rock, the influence of stratification on geochemical gradients and arsenic mobilization (e.g. sedimentary aquifers) and heterogeneity with respect to geochemical conditions (e.g. pH and Eh). Although arsenic concentrations in sedimentary aquifers are lower than in crystalline aquifers, an improved understanding of the controls on arsenic mobilization in these aquifers is still useful because they are important sources of groundwater to municipal water supplies around the province and are often targeted for water supply development.

A regional-scale analysis is also recommended to investigate whether metamorphic grade might explain why lower concentrations of arsenic are generally observed in Goldenville Group bedrock aquifers at the eastern and western ends of the province. Additional sampling for arsenic in well water is recommended where there are gaps in water sample coverage, especially where there is a large number of private well users relying on the aquifer for domestic water supply. The development of higher resolution mapping of the Goldenville and Halifax groups along the eastern shore of Nova Scotia would allow refinement of the risk map in this area.

Another important gap regarding our understanding of the risk of arsenic exposure to private well owners is the lack of survey information describing the percentage of well users with unsafe levels of arsenic in their raw drinking water that have implemented successful and sustainable mitigation measures. There are also few data available on how risk communication and personal risk assessment affects the behaviours of private well owners with respect to implementing and maintaining appropriate water treatment systems (Chappells et al., 2014). This information is critical to ensure that risk communication efforts are designed effectively.

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