GEOSCIENCE AND MINES BRANCH

A Review of Activities Related to the Occurrence of Arsenic in Nova Scotia Well Water

G. W. Kennedy and J. Drage

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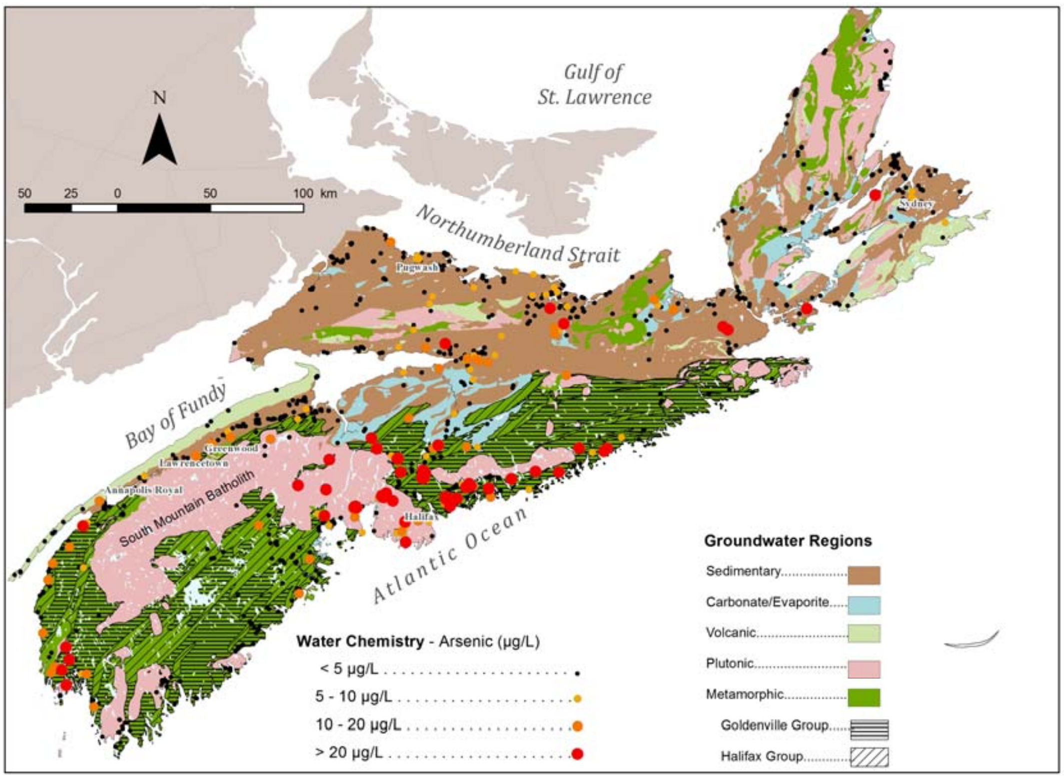

NOVA SCOTIA
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Introduction

The presence of naturally occurring arsenic in drinking water in Nova Scotia was first identified in 1976 when a patient at the Victoria General Hospital was diagnosed with symptoms of chronic arsenic intoxication (Grantham and Jones, 1977). Subsequent water testing indicated that the dug well at the patient's home in Waverley, Nova Scotia, had an arsenic concentration of 5000 μg/L, which is 500 times higher than the current Canadian drinking water guideline maximum acceptable concentration (MAC) of 10 μg/L (Health Canada, 2014).

The detection of arsenic in well water in Waverley led to the formation of a provincial multi-disciplinary task force that developed and implemented a program to investigate the occurrence of arsenic in water wells in Nova Scotia (Province of Nova Scotia, 1976). Waverley is located near a past producing gold district, and since the province's historic gold districts were known to contain tailings and waste rock with high levels of arsenic-bearing minerals, the task force's investigation initially focused on private wells located in heavily mined, densely populated gold districts of the Meguma terrane between Kemptville and Goldboro (Fig. 1) (Province of Nova Scotia, 1976). Although the source of arsenic in the Waverley patient's dug well was attributed to groundwater flow through mine tailings and waste rock deposits near the patient's residence, the problem was discovered to be more widespread (Grantham and Jones, 1977). The task force also attributed the arsenic detected in water wells to a geological source, concluding that the weathering of soil and rock containing naturally occurring arsenic can result in concentrations in groundwater exceeding acceptable levels for drinking water supply. These findings were published in an interim report (Province of Nova Scotia, 1976).

Arsenic is now considered the most prevalent naturally occurring groundwater contaminant in the province, and research over the past four decades has indicated that bedrock geology is the most important control on arsenic concentrations in well water in Nova Scotia (e.g. Bottomley, 1984; Dummer *et al.*, 2015) as well as in the broader region of the northern Appalachian orogen (Ayotte *et al.* 2003; Robinson and Ayotte, 2006; Peters, 2008; Klassen *et al.*, 2009; Yang *et al.*, 2009).

The purpose of this report is to summarize the available research related to arsenic occurrence in water wells in Nova Scotia, including the following: (1) human health effects and exposure pathways of arsenic; (2) the distribution of arsenic in Nova Scotia groundwater and the population at risk; (3) controls on the occurrence and mobility of arsenic in well water; and (4) strategies for managing the risk of arsenic exposure in private wells in Nova Scotia.

Health Effects and Exposure

Arsenic, a toxic metal(loid), is classified by the International Agency for Research on Cancer (IARC) as a Class I human carcinogen that causes skin, bladder, kidney, and lung cancer (IARC, 2004). The understanding of arsenic toxicology, and the corresponding maximum acceptable concentration (MAC) of arsenic in drinking water published in the Canadian Drinking Water Quality Guidelines (Health Canada, 2014), has decreased from 50 μg/L in the 1970s to 10 μg/L today (Fig. 2). It is important to note

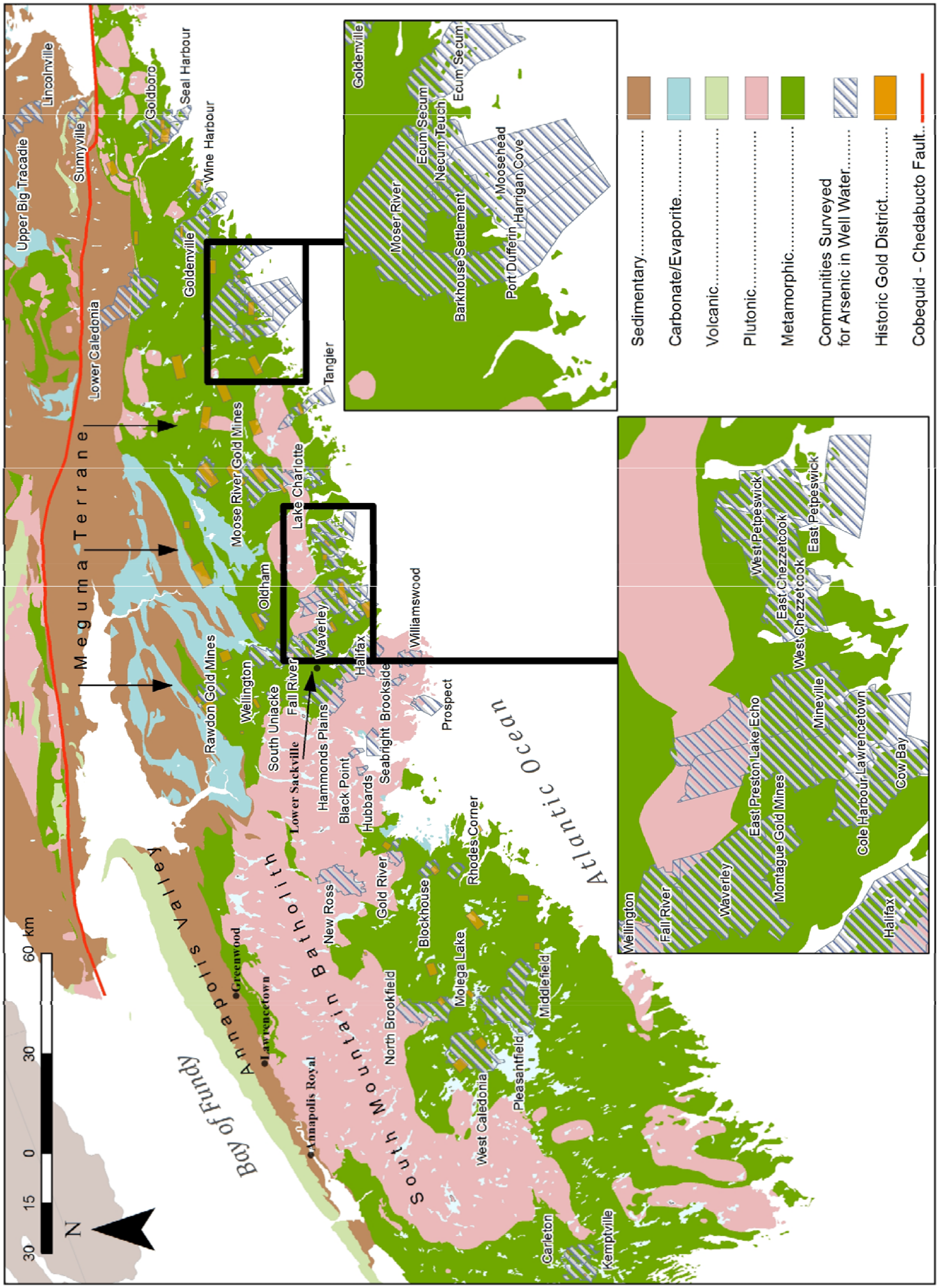

Figure 1. Locations of arsenic in well water surveys conducted by the Province of Nova Scotia, historical gold districts and the Meguma terrane.

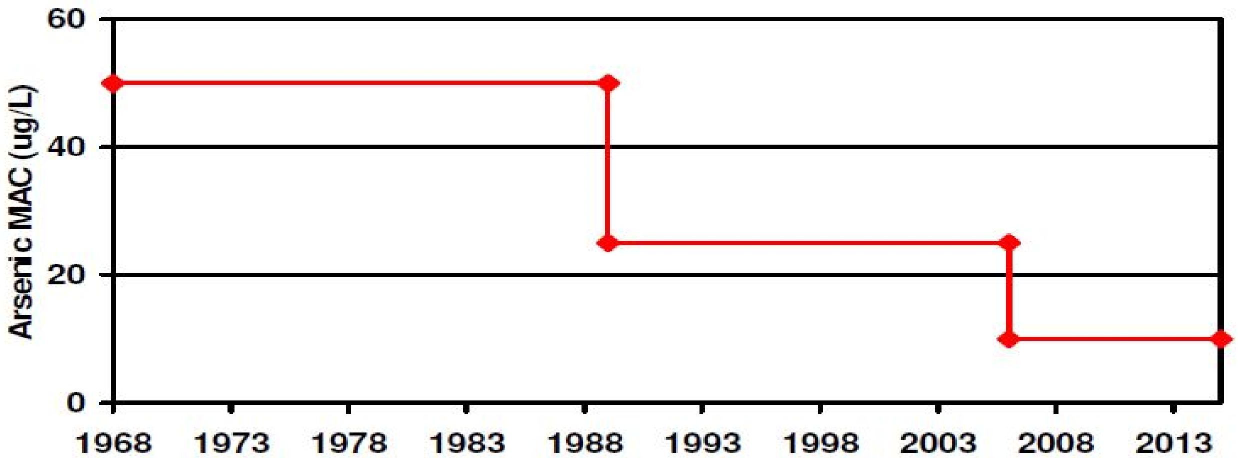

Figure 2. Change in Health Canada (2014) maximum acceptable concentration (MAC) for arsenic in drinking water over time from 50 µg/L to 10 µg/L.

that the present MAC of 10 µg/L arsenic is based on treatment achievability, and Health Canada (2014) recommends that arsenic levels in drinking water be reduced to levels as low as reasonably achievable.

Common pathways for human exposure to arsenic include air, water, soil and food (e.g. fish, shellfish and rice), but drinking water is the pathway considered to have the greatest impact on human health (Smedley and Kinniburgh, 2002; Orloff *et al*., 2009), and is the focus of the current review. Private well users account for approximately 40% of domestic water use in Nova Scotia, and these water users are unregulated when it comes to drinking water quality. It is the responsibility of individual well owners to ensure that their drinking water is safe and to mitigate any potential health risks. Owners of public water supplies in Nova Scotia (defined as systems having at least 15 service connections, regularly serving at least 25 people per day for 60 days of the year or serving daycares, restaurants and commercial accommodations for at least 60 days of the year) are responsible for the provision of safe drinking water.

Following ingestion, most inorganic arsenic will be absorbed and transformed in the liver through methylation processes into organic forms of arsenic, which are more water soluble and therefore easier to eliminate from the body through excretion. Methylation capacity varies among individuals, and recent epidemiological studies have shown that inefficient methylation capacity, characterized by a higher proportion of urinary intermediate metabolites of arsenic methylation, is related to increased susceptibility to arsenic-caused diseases (Li *et al.,* 2015). When inorganic forms of arsenic are not methylated and excreted they can affect human health by interfering with sulfhydral groups in proteins and enzymes (Saha *et al.,* 1999).

Short-term, high-dose oral exposure to high levels of arsenic can cause a range of adverse health effects, such as gastrointestinal irritations, accompanied by difficulty in swallowing, thirst, low blood pressure and convulsions, whereas long-term low-dose exposure to arsenic is associated with hypertension (Li *et al.,* 2015) and an increased risk of cancer (Saha *et al.,* 1999). Arsenic exposure through ingestion has been clearly linked to an increased risk of skin cancer and there is growing evidence that it also increases the risk of internal cancers (lung, liver, bladder and kidney) (Saint-Jacques *et al*., 2014). At low to moderate levels of exposure to arsenic in drinking water there is more uncertainty regarding the health risks, although a recent meta-analysis conducted by Saint-Jacques *et al*. (2014) found that exposure to arsenic in drinking water wells at concentrations exceeding 10 µg/L was associated with a doubling of bladder cancer risk. Nova Scotia's urinary tract cancer rates rank among the highest in Canada, with age-standardized incidence rates for males and females reported to be 33 and 9 per

100,000, respectively, for bladder cancer, and 22 and 13 per 100,000, respectively, for kidney cancer (Canadian Cancer Society's Steering Committee on Cancer Statistics, 2015). The toxicity of arsenic also depends on the species of arsenic that is ingested, with trivalent arsenic (As^{3+} valence state) estimated to be 10 times more toxic than pentavalent arsenic (As^{5+} valence state) (Vaclavikova *et al.*, 2008).

Following the detection of arsenic in well water in the community of Waverley, Hindmarsh *et al*. (1977) conducted clinical studies and found chronic arsenicalism in the community's population. The clinical sampling program collected hair, nail, blood, and urine samples from a cohort of 86 people in the community. Approximately 94% of the people with high arsenic (>100 µg/L) in their well water also had high arsenic in their hair, and approximately 70% of these people exhibited mild symptoms consistent with arsenic poisoning, such as peripheral neuritis. High levels of arsenic (270 to 1460 µg/L) were detected in 1977 in a cluster of eight drilled wells located in the community of Lower Sackville, with some of the residents reporting symptoms consistent with arsenic poisoning. Records indicate that limited hair, blood, urine, and nervous system tests were conducted although a published account of the clinical findings could not be located.

More recent epidemiological work by Yu *et al*. (2013) evaluated the relationship between arsenic concentrations in drinking water and toenail clippings among a cohort of Nova Scotians. A total of 960 men and women aged 35 to 69 years provided household drinking water and toenail clipping samples to researchers with the Atlantic Partnership for Tomorrow's Health (PATH) project. The study participants came from across the province although the majority of participants were located in the Halifax and Sydney area population centres. A significant relationship was detected between drinking water and toenail arsenic concentrations among the participants with levels of arsenic ≥1 µg/L in their drinking water, and the study concluded that arsenic ingested in the drinking water of private wells is the most important contributor to arsenic body burden. Similarly, a joint study by researchers at Health Canada and Acadia University of 108 households in the communities of Hubbards and Fall River, two rural communities reliant on private well water in areas known to be associated with elevated concentrations of arsenic, found that arsenic levels in toenail and hair samples showed the best correspondence with arsenic in well water (Cull, 2011). In general, the biomonitoring study found that long-term exposure indicators (hair and nails) were better correlated with arsenic in well water, whereas short-term exposure indicators (urine) were better correlated with food consumption.

Distribution of Arsenic in Nova Scotia

Arsenic in Well Water Surveys – Meguma Terrane

Extensive surveys have been conducted by the Province of Nova Scotia to characterize the distribution of arsenic in well water, and hence the magnitude of the issue and the population at risk. Most of these surveys were conducted between 1976 and 1988 and were located in the metamorphic rocks of the Cambrian to Ordovician Goldenville and Halifax groups and their associated gold districts, and the granitoid rocks of the Devonian South Mountain Batholith. The survey locations are shown in Figure 1 and selected data from these surveys were compiled from Nova Scotia Environment (NSE) files and are presented in Tables 1a and 1b.

The Arsenic Task Force that formed in 1976 initially tested over 600 private wells located in 21 gold districts in six counties across southern Nova Scotia, and approximately 200 wells in areas located outside of these gold districts (Grantham and Jones, 1977). Although there are a total of 64 gold districts, all within the Meguma terrane (Smith and Goodwin, 2009), only the gold districts considered by the task force to have the greatest risk of private well contamination by historical mining activity were included in the initial survey (Province of Nova Scotia, 1976). A critical finding of the task force

Table 1a. Summary of exceedance rates (>MAC) compiled from arsenic in well water surveys conducted by the province in gold district areas.

Area	County	Count of Wells Sampled		>50 µg/L		>10 µg/L		Exceedance Rate (>10 µg/L)		Rationale for Survey	Source
		Dug	Drilled	Dug	Drilled	Dug	Drilled	Dug	Drilled		
Barkhouse Settlement	Guysborough	4	0	0	0	1	0	25%	-	Gold District	a
Blockhouse	Lunenburg	7	0	0	0	0	0	0%	-	Gold District	a
Blockhouse	Lunenburg	13	19	0	0	0	2	0%	11%	Gold District and homeowner/ developer concern	e
Caribou Gold Mines	Halifax	6	0	0	0	2	0	33%	-	Gold District	a
Carleton	Yarmouth	2	1	0	0	0	0	0%	0%	Gold District	a
Chezzetcook area	Halifax	11	5	0	0	0	2	0%	40%	Gold District	b
Cow Bay	Halifax	4	10	0	0	0	0	0%	0%	Gold District	a
Drumhead Harbour	Guysborough	4	0	1	0	1	0	25%	-	Gold District	a
Dufferin Mines	Guysborough	3	0	0	0	0	0	0%	-	Gold District	a
Ecum Secum/Necum Teuch	Halifax	18	4	0	0	0	1	0%	25%	Gold District	a
Gold River	Lunenburg	3	2	0	0	0	1	0%	50%	Gold District	a
Goldboro	Guysborough	8	1	0	0	0	0	0%	0%	Gold District	a
Goldenville	Guysborough	18	0	6	0	14	0	78%	-	Gold District	a
Harrigan Cove	Guysborough	4	0	0	0	0	0	0%	-	Gold District	a
Isaac's Harbour	Guysborough	4	0	0	0	0	0	0%	-	Gold District	a
Kemptville	Yarmouth	4	0	0	0	0	0	0%	-	Gold District	a
Lake Charlotte	Halifax	3	1	0	0	0	0	0%	0%	Gold District	b
Lawrencetown area (incl. Mineville)	Halifax	53	45	0	12	6	23	11%	51%	Gold District	a
Lower Caledonia	Guysborough	2	1	0	0	0	0	0%	0%	Gold District	b
Mineville - Dempster Cres and Mineville Rd	Halifax	11	27	0	5	0	11	0%	41%	Gold District and homeowner/ developer concern	c
Molega Lake	Queens	5	0	0	0	0	0	0%	-	Gold District	a
Montague Mines	Halifax	9	10	1	2	5	5	56%	50%	Gold District	a
Montague Mines	Halifax	0	8	0	1	0	3	-	38%	Gold District	d
Moose River Gold Mines	Halifax	11	0	0	0	2	0	18%	-	Gold District	a
Moosehead and Moser River	Halifax	3	1	0	1	0	1	0%	100%	Gold District	a
North Brookfield	Queens	3	5	0	1	0	1	0%	20%	Gold District	a
Oldham	Halifax	32	24	7	1	15	5	47%	21%	Gold District	a
Petpeswick (East and West)	Halifax	0	2	0	1	0	2	-	100%	Gold District	b
Pleasantfield	Queens	1	1	0	0	0	0	0%	0%	Gold District	a
Rawdon	Hants	3	1	0	0	0	0	0%	0%	Gold District	a
Rawdon Gold Mines	Hants	2	3	0	0	0	1	0%	33%	Gold District	a
Seal Harbour	Guysborough	6	0	0	0	0	0	0%	-	Gold District	a
South Uniacke	Hants	14	2	0	0	1	2	7%	100%	Gold District	a
Tangier	Halifax	37	5	4	3	8	4	22%	80%	Gold District	a
Waverley	Halifax	101	77	7	23	30	43	30%	56%	Gold District	a
West Caledonia	Queens	4	5	0	0	0	0	0%	0%	Gold District	b
Wine Harbour	Guysborough	8	0	0	0	0	0	0%	-	Gold District	a
		TOTALS		**PER CENT OF TOTALS**							
		421	**260**	**6%**	**19%**	**20%**	**41%**				

a. Arsenic Task Force (Grantham and Jones, 1977)

b. 1976 ATF Survey - Internal Files

c. 1981 Survey - Internal Files

d. Health Canada (unpublished data 2009-2011)

e. Arsenic and Uranium Concentrations Well Water Supplies Rhodes Corner and Blockhouse Areas (Nova Scotia Department of Health, 1989)

Table 1b. Summary of exceedance rates (>MAC) compiled from arsenic in well water surveys conducted by the province in areas not designated as gold districts.

Area	County	Count of Wells Sampled		>50 µg/L		>10 µg/L		Exceedance Rate (>10 µg/L)		Rationale for Survey	Source
		Dug	Drilled	Dug	Drilled	Dug	Drilled	Dug	Drilled		
Black Point	Halifax	39	6	0	3	0	5	0%	83%	Homeowner/developer concern	f
Brookside	Halifax	5	23	0	3	0	7	0%	30%	Homeowner/developer concern	c
Cole Harbour - Caldwell Rd and Ritcey Cres	Halifax	9	7	0	4	0	7	0%	100%	Homeowner/developer concern	f
East Preston	Halifax	1	3	1	0	1	3	100%	100%	Homeowner/developer concern	f
Fall River area	Halifax	14	326	0	5	0	33	0%	10%	Epidemiological study	d
Halifax - Birch Cove	Halifax	13	106	0	12	0	33	0%	31%	Homeowner/developer concern	g
Hammonds Plains - Lucasville Rd	Halifax	15	14	0	0	2	0	13%	0%	Homeowner/developer concern	a
Hubbards	Lunenburg	134	45	0	23	1	30	1%	67%	Homeowner/developer concern	j
Hubbards area	Lunenburg	32	278	0	7	0	44	0%	16%	Epidemiological study	d
Lake Echo - Pine Grove, Forest Park S/D	Halifax	3	62	0	7	0	25	0%	40%	Homeowner/developer concern	h
Lake Echo - Ponderosa S/D	Halifax	22	33	2	4	2	10	9%	30%	Homeowner/developer concern	c
Lincolnville	Guysborough	2	14	0	3	0	6	0%	43%	Homeowner/developer concern	b
Lower Sackville	Halifax	0	12	0	10	0	11	-	92%	Homeowner/developer concern	g
Middlefield	Queens	1	1	0	0	0	0	0%	0%	Homeowner/developer concern	b
New Ross	Lunenburg	7	5	0	1	0	1	0%	20%	Homeowner/developer concern	a
Prospect	Halifax	9	33	1	4	1	21	11%	64%	Homeowner/developer concern	f, i
Purcells Cove	Halifax	6	2	0	1	0	1	0%	50%	Homeowner/developer concern	f
Rhodes Corner	Lunenburg	65	30	0	5	0	5	0%	17%	Homeowner/developer concern	e
Seabright	Halifax	47	22	0	7	0	9	0%	41%	Homeowner/developer concern	f, h
Sunnyville	Guysborough	0	6	0	0	0	0	-	0%	Homeowner/developer concern	b
Upper Big Tracadie	Guysborough	0	4	0	0	0	0	-	0%	Homeowner/developer concern	b
Waverley - Silversides S/D	Halifax	2	37	0	6	0	12	0%	32%	Homeowner/developer concern	a
Waverley - Waverley Road	Halifax	12	15	0	0	1	3	8%	20%	Homeowner/developer concern	a
Wellington - Collins Park S/D	Halifax	12	35	2	15	6	20	50%	57%	Homeowner/developer concern	a
Williamswood	Halifax	0	4	0	0	0	0	-	0%	Homeowner/developer concern	c
		TOTALS		**PER CENT OF TOTALS**							
		450	**1123**	**1%**	**11%**	**3%**	**25%**				

f. 1980 Survey - Internal Files

g. 1977 DOH Survey - Internal Files

h. 1979 Survey - Internal Files

i. 1983 Survey - Internal Files

j. Report on Arsenic Survey, Hubbards, Nova Scotia (Nova Scotia Department of the Environment, 1978) was that elevated levels of arsenic also occur in areas that are not close to known gold-producing areas (e.g. Waverley 'Silversides' and Wellington 'Collins Park' subdivisions) (Grantham and Jones, 1977). The task force study also found that drilled wells were more likely to have elevated arsenic levels, but some dug wells near mine tailing piles and rock dumps exhibited arsenic levels exceeding drinking water quality guidelines. Tables 1a and 1b show average exceedance rates of the current MAC for arsenic of 20% and 41% for dug and drilled wells, respectively, located in gold district areas. By comparison, exceedance rates of 3% and 25% are estimated for all dug and drilled wells, respectively, located in areas not designated as gold districts. The data compiled in these tables indicate that high exceedance rates (e.g. >30%) of the MAC for arsenic in drinking water are observed for drilled wells both inside and outside of gold district areas, whereas high exceedance rates for dug wells in areas not designated as gold districts are less common, which may reflect the influence of till geochemistry and/or the presence of waste rock and tailings.

Following the initial Arsenic Task Force survey, the province conducted additional arsenic in water well surveys where arsenic problems were detected during routine water testing. For example, a well water investigation by Nova Scotia Environment (NSE) in 1978 surveyed 179 wells (134 dug wells and 45 drilled wells) located in granitoid rock of the South Mountain Batholith in the Hubbards area after a well was found to have elevated levels of arsenic (Nova Scotia Department of Environment, 1978). Approximately 67% of the drilled wells surveyed had arsenic levels above 10 µg/L whereas only one of the dug wells (1%) had arsenic levels greater than 10 µg/L (Table 1b). Prior to this investigation, it was believed that elevated arsenic levels were restricted to wells located in rocks of the Goldenville and Halifax groups. The investigation also found that arsenic levels generally increased with well depth, and the majority of wells with high arsenic also had pH levels above 7.0.

Other arsenic in well water surveys conducted by the province following the initial task force survey were located in areas mostly underlain by metasandstone of the Goldenville Group, such as the communities of Lake Echo, Birch Cove, Rhodes Corner, Blockhouse and Cole Harbour, and areas underlain by granitoid rock of the South Mountain Batholith, such as the communities of Prospect, Brookside, Seabright and Black Point (Fig. 1; Table 1a and 1b). These surveys were generally initiated in response to concerns identified by municipalities, subdivision developers and homeowners. More recently, the Clean Annapolis River Project (2014) conducted a rural private well survey in the Annapolis Valley in areas underlain primarily by sedimentary rocks of the Wolfville and Blomidon formations, and found that approximately 6.5% of the drilled wells tested (n=124) exceeded the Health Canada MAC for arsenic in drinking water. The Health Canada/Acadia University and Atlantic PATH surveys (Table 1b) were conducted to better understand the epidemiology of arsenic exposure via well water and its relationship to arsenic body burden. Reported exceedance rates for arsenic in drilled well water samples were 9% for the Atlantic PATH province-wide survey (n=3500, Chappells *et al*., 2014), and 10% for the Health Canada/Acadia survey in the communities of Fall River and Hubbards (n=604, L. White, Health Canada, personal communication).

Arsenic Speciation in Well Water Surveys – Meguma Terrane

In addition to surveys of total arsenic in well water, there have been several surveys of arsenic speciation (McCurdy, 1980; Guernsey *et al*., 2004; Cull, 2011) to characterize the distribution of the various forms of arsenic in the province's groundwater, since trivalent arsenic is the species of most concern to human health. Arsenic can be present in many different forms and valencies in Nova Scotia's aquifers, however dissolved arsenic occurs mainly in two valence states: +3, known as trivalent arsenic or As(III), and +5, known as pentavalent arsenic or As(V). Although inorganic arsenic tends to be the dominant form of arsenic in groundwater, both As(III) and As(V) can form stable bonds with carbon and, therefore, arsenic may also be present in organic compounds (Vaclavikova *et al*., 2008).

Table 2. Summary of arsenic in well water speciation results.

Survey	Well Type	Count of Wells Tested	Average Percentage of As(III) of Total As	Per cent exceeding 10 µg/L for As(III)	Maximum Concentration of As(III) (µg/L)	Source
School wells located in Meguma terrane and South Mountain Batholith	Drilled	12	10%	8%	735	Guernsey et al. (2004)
Hubbards and Fall River areas	Drilled	67	14%	16%	249	Cull (2011)
Waverley	Dug	55	10%	5%	2900	McCurdy (1980)
Waverley	Drilled	78	20%	26%	470	McCurdy (1980)

The first survey of arsenic species in well water was conducted by McCurdy (1980). Samples were collected from a total of 213 wells in Waverley and analyzed for total arsenic and general chemistry. Detectable concentrations of arsenic (i.e. >5 µg/L) were measured in 133 of the wells sampled (55 dug wells and 78 drilled wells) and these samples were further analyzed for As(III) and As(V) concentrations. The results generally show that As(V) is the dominant species, with the average percentage of As(V) accounting for 90% of total arsenic in dug wells and 80% of total arsenic in drilled wells (Table 2). In six of the drilled wells, however, the reverse trend was observed with As(III) representing the majority of total arsenic. Similarly, speciation work by Bottomley (1984) of a test well in Waverley showed that 90% of total inorganic arsenic was present as the more toxic As(III) species, amounting to 333 µg/L of As(III). McCurdy (1980) also conducted a weekly sampling program over a one year period at four stations in Waverley that included analyses of inorganic and organic (monomethyl and dimethyl) arsenic, and found that inorganic arsenic represented more than 99% of the total arsenic in the sample results.

In 2004, a study of arsenic in school groundwater supplies was prepared for NSE to investigate how arsenic species are partitioned in the well water (Guernsey *et al*., 2004). The study involved a limited sampling program of the province's school water wells located in areas known to be associated with elevated concentrations of arsenic (metamorphic and plutonic rocks of the Meguma terrane). Raw water from drilled wells at 12 schools in five counties was sampled for total arsenic, As(III) and As(V). The results from the sampling program indicated that As(V), the less toxic form of arsenic, was the dominant form of arsenic present in 11 of the 12 school wells surveyed, and in these 11 wells the average proportion of As(III) in the water sample was approximately 10% of the total arsenic (Table 2). Nine of the 12 school wells had As(V) above 10 µg/L and one of the 12 school wells had As(III) above 10 µg/L. The highest total arsenic concentration (829 µg/L) was detected in the well where As(III) was dominant, partitioned as 735 µg/L of As(III) and 94 µg/L of As(V). Speciation of selected samples (n=67) collected during the arsenic in well water surveys of the Fall River and Hubbards areas found that the average contribution of As(III) to total arsenic in drilled wells was around 14% (Table 2) (Cull, 2011).

These surveys indicate that most dissolved arsenic in groundwater occurs as inorganic As(V) in the metamorphic and plutonic rocks of the Meguma terrane, typically accounting for more than 80% of the total arsenic concentration in well water samples, although in some wells the more toxic As(III) form of arsenic can be dominant. Arsenic in well water speciation data have not been collected for other areas of the province.

Sources and Management of Arsenic in Well Water Data

The province developed a centralized inventory (hard copy ledger book) for tracking arsenic results when the issue of elevated arsenic in well water first emerged. The inventory included arsenic results from well surveys completed by staff and arsenic results collected directly from homeowners between 1976 and the mid-1980s. Some of these data were then entered into a digital database called NAQNSWELL by NSE around the mid-1980s. The NAQNSWELL database was also used to record well water analyses from contaminated sites investigations around the province and is therefore not considered a representative dataset of ambient groundwater geochemistry.

A separate digital database was maintained by the Nova Scotia Department of Health (DOH) whereby concerned homeowners, realtors and developers were invited to voluntarily send in arsenic water results to the DOH between 1981 and 1994, and DOH staff compiled the results into the database. The data are biased because they consist mainly of arsenic results submitted by homeowners or realtors located in known arsenic hazard areas of suburban Halifax where development activity was concentrated. The well type (dug vs. drilled) and water type (raw vs. treated) were not recorded in the database, although it is expected that the majority of results submitted to the DOH consist of raw water samples from drilled wells since most new subdivisions in these areas relied on drilled wells, and homeowners were seeking advice on treatment options based on the arsenic levels present in their raw water.

In the late 1990s NSE requested available arsenic data from the Queen Elizabeth II hospital (QEII) laboratory, which performs many of the residential well water analyses in the province, for publication in a ‘State of the Environment’ report (Nova Scotia Department of Environment, 1998). The dataset contained more than 10,000 records from water analyses completed between 1990 and 2000, although owner and site location data were removed from the dataset by the QEII to protect well owners’ privacy. This dataset is also biased because in addition to poor location information there are a number of duplicate, treated water, and surface water samples that cannot be filtered from the dataset.

Despite the data quality issues noted above, these datasets contain a large number of samples, and community level exceedance rates can provide useful information regarding the spatial distribution of arsenic in well water. Arsenic concentrations in communities with some of the highest rates of exceedances based on the DOH and QEII datasets are summarized in Appendix A. The compiled results show that arsenic can range over four orders of magnitude in Nova Scotia groundwater under natural conditions, with arsenic levels exceeding 1000 μg/L in the communities of Hubbards, Prospect, East Preston, Montague Gold Mines and Head of Chezzetcook. The highest median arsenic levels for communities with at least 20 well water samples include Prospect, Eastern Passage, East Preston, Mineville and Lawrencetown (Halifax County). The overall percentage of samples exceeding the arsenic MAC in the communities listed in Appendix A is 33%, with exceedance rates surpassing 60% in the communities of Amiraults Hill, Montague Gold Mines, East Preston and Mineville (combined datasets, $n \geq 20$) (Fig. 3 and Appendix A).

Distribution of Arsenic in Nova Scotia’s Bedrock Groundwater Regions

The Groundwater Regions Map of Nova Scotia (Kennedy and Drage, 2008, 2009) broadly classifies the groundwater regions of Nova Scotia into five major categories based on the dominant rock type of the various bedrock units interpreted on the provincial bedrock geology compilation map (Keppie, 2000). The distribution of arsenic in Nova Scotia’s bedrock groundwater regions was first published as part of a series of seventeen provincial bedrock groundwater chemistry maps in 2011 (Kennedy and Finlayson-Bourque, 2011). The data used in the map are from the provincial groundwater chemistry database (Nova Scotia Department of Natural Resources, 2015), which 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). An updated distribution map of arsenic in Nova Scotia’s bedrock groundwater regions is shown in Figure 4, with the metamorphic rocks of the

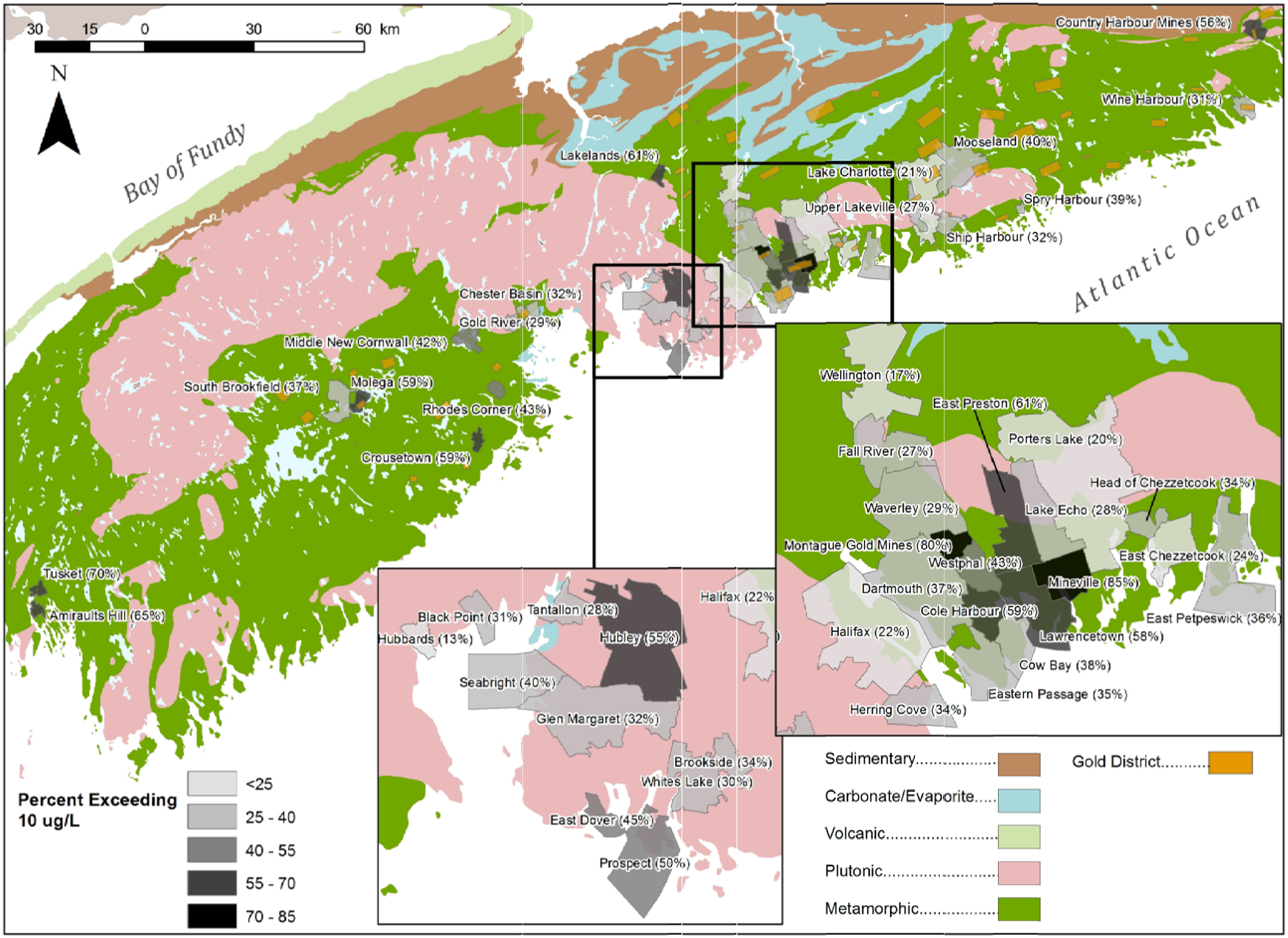

Figure 3. Per cent of samples exceeding 10 µg/L arsenic in well water for various communities surveyed in the Meguma terrane, bedrock groundwater regions, and historical gold districts.

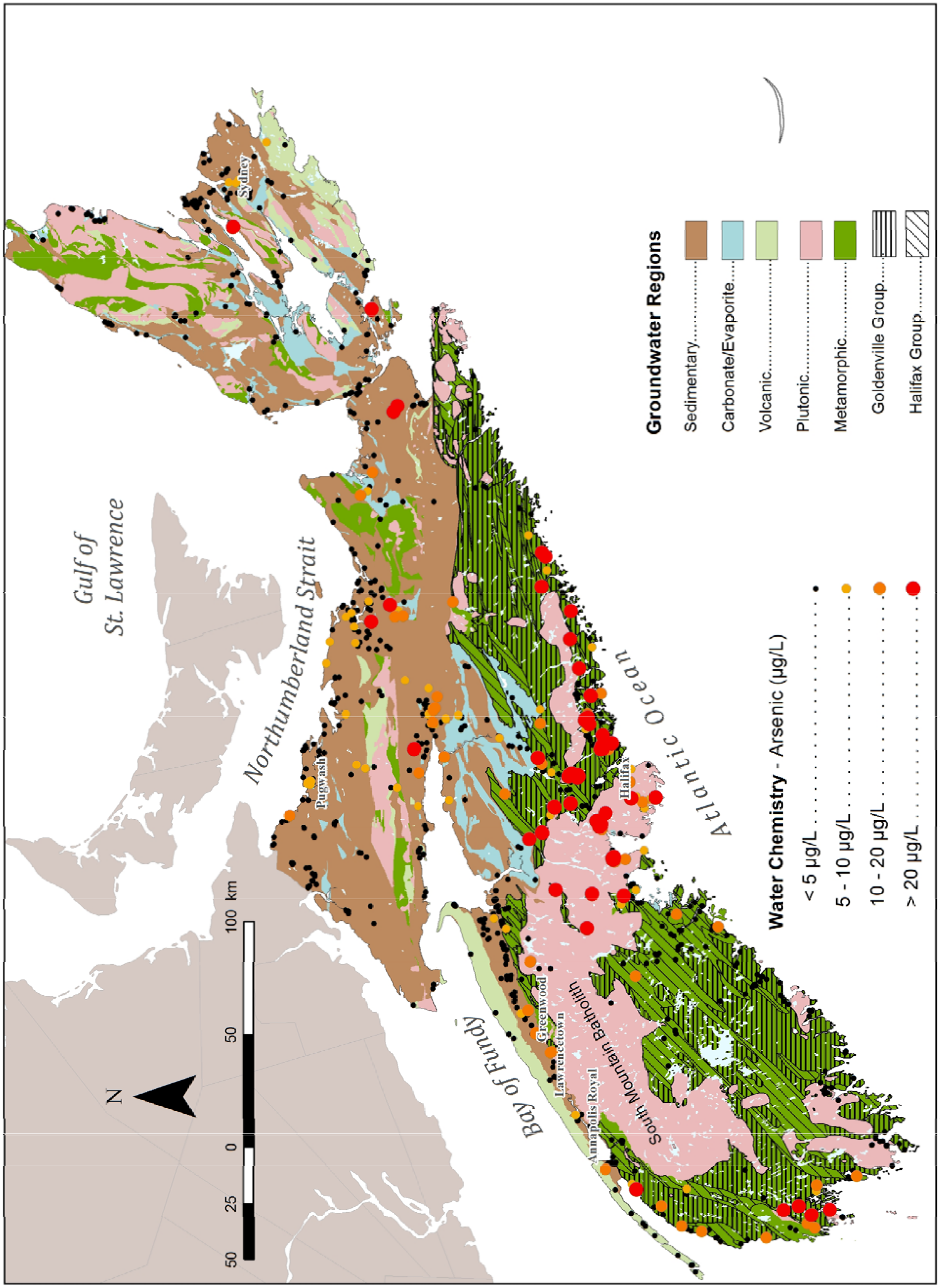

Figure 4. Distribution map of arsenic concentrations in well water from the Nova Scotia Groundwater Chemistry database compared to Nova Scotia's bedrock groundwater regions, with the metamorphic rocks of the Meguma terrane divided into the Halifax and Goldenville groups. Where multiple sample results were available for a given well, the most recent sample concentration is shown.

Meguma terrane divided into the Halifax and Goldenville groups. Arsenic concentrations range from less than 2 µg/L to 710 µg/L and approximately 12% of raw water samples (n=1264) exceed the Health Canada MAC of 10 µg/L. In comparison only 2% of raw water samples exceed the arsenic MAC in the province's surficial aquifers (n=199) (Nova Scotia Department of Natural Resources, 2015).

Arsenic levels in groundwater exceeding the current drinking water MAC (Health Canada, 2014) have been reported in all of Nova Scotia's groundwater regions, with the exception of the volcanic groundwater region. In localized areas of the sedimentary and carbonate/evaporite groundwater regions of the province, clusters of elevated arsenic concentrations in drilled wells have been reported. For example, arsenic in well water issues have been recorded in sedimentary bedrock aquifers of the Wolfville Formation near the contact with the South Mountain Batholith between the communities of Greenwood and Annapolis Royal, in the sedimentary bedrock aquifers of the Mabou Group around central Pictou County, and in areas of the Village of Pugwash underlain by Windsor Group evaporite bedrock. The metamorphic rocks of the Goldenville and Halifax groups and granitoid rocks of the South Mountain Batholith, however, generally have the highest exceedance rates of arsenic in well water compared to other bedrock types (Nova Scotia Department of Health, 1983; Kennedy and Drage, 2009; Chappells *et al*., 2014; Dummer *et al*., 2015).

In comparison, the highest concentrations of arsenic in New England (USA) well waters is associated with sulphidic and calcareous metasedimentary rocks (Ayotte *et al*., 2003; Yang *et al*. 2009; O'Shea *et al*., 2014). In New Brunswick, elevated arsenic in well waters is associated with black shale and iron-rich metasedimentary bedrock, and bedrock characterized by sulphide mineralization, manganese oxides, uranium, phosphorite and coal (Klassen *et al*., 2009). Many of these bedrock types exist in northern mainland Nova Scotia and Cape Breton Island, but there are limited arsenic in well water data available for these areas of the province.

Origin and Controls of Arsenic in Well Water

A number of studies have been conducted in the northern Appalachian orogen to better understand the origin and controls of arsenic in well water since the issue first emerged around the 1970s. Generally arsenic concentrations in groundwater have been found to be heterogeneous and the result of complex hydrogeochemical processes, although there is broad correspondence between groundwater chemistry and bedrock composition.

Geogenic Sources of Arsenic

In Nova Scotia, the major geogenic forms of arsenic include arsenopyrite, where arsenic is present in the crystal structure of the sulphide mineral as a substitute for sulphur (i.e. FeAsS), and arsenic-rich pyrite, where soluble arsenic is incorporated during the formation of the pyrite mineral. Bedrock hosting sulphide minerals and pyritic rocks, such as the Goldenville and Halifax groups, are therefore the main sources of arsenic in the province. Other less common minerals containing arsenic found in Nova Scotia include scorodite (a hydrated iron arsenate), particularly within oxidized zones of arsenopyrite-rich tailings (Walker *et al*. 2009; DeSisto *et al*., 2011). Arsenopyrite is associated with gold, lead-zinc and nickel-copper mineralization and arsenic concentration is often measured in geochemical surveys as a pathfinder element (Kerswill, 1991). For example, in the Meguma terrane, arsenopyrite is preferentially concentrated with gold in the hinges of anticlines (Ryan and Smith, 1998).

Smedley and Kinniburgh (2002) reviewed arsenic concentrations in various metamorphic rocks worldwide and found that most contain around 5 ppm total arsenic, reflecting levels present in their protoliths; however, marine protoliths such as Goldenville and Halifax groups were associated with

Table 3. Statistical summaries of (a) arsenic concentrations in various rocks of the Goldenville and Halifax groups, and the South Mountain Batholith, and (b) arsenic concentrations in other types of media in the province.

Table 3a

	Laboratory				Portable XRF				
	Min	**Max**	**Median**	**n**	**Min**	**Max**	**Median**	**n**	**Sources**
Goldenville Group	**0.1**	**13000**	**20**	**565**	**0.5**	**481**	**5.7**	**177**	A, B, C, D
Beaverbank	0.5	433	32.4	80	0.5	481	8	34	
Bloomfield	1.3	105	26.9	6					
Church Point	0.2	136	13	79					
Government Point	0.1	278	11	95					
Green Harbour	0.3	13000	48	149					
Moses Lake	0.5	86	9	10					
Moshers Island	0.5	527	45	76					
Tangier	4	17	10.5	12					
Taylors Head	1	59	9.6	58	0.5	43	5	143	
Halifax Group	**<DL**	**3559**	**13**	**93**	**<DL**	**928**	**4**	**134**	A, B, C, D
Acacia Brook	2.7	52	30.75	8					
Bluestone	0.5	69.2	4.75	10	0.5	74.6	4	27	
Cunard	<DL	3559	14.8	70	<DL	928	4	107	
South Mountain Batholith	**<DL**	**9000**	**2.7**	**652**	**<DL**	**10.2**	**2.25**	**39**	A, J

Table 3b

	Min	**Max**	**Median**	**n**	**Sources**
Till Surveys	0.1	12900	10	9418	H, I, J, K, L, M, N, O
Soil Surveys	<DL	2000	9	4840	H, I, P, Q, R
Biogeochemical Surveys	0.25	100	3.3	2496	S, T, U, V, W
Surface Water and Sediment Surveys	<DL	1200	3.5	15061	X, Y, Z, AA, H, AA, AB, AC

Notes
All values reported in ppm arsenic
DL: Method detection limit

Data Sources

A	White, 2010	P	Carrick Gold Resources Ltd., 1990
B	Farmer and White, 2015	Q	Goodwin, 2005
C	White et al., 2014	R	Goodwin et al, 2009
D	White and Goodwin, 2011	S	NSDNR, 2006d
E	Hill, 1987	T	NSDNR, 2006e
F	Graves and Zentilli, 1988	U	NSDNR, 2006f
G	Kerswill, 1993	V	Dunn et al., 1994a,
H	NSDNR, Unpublished	W	Dunn and Balma, 1997
I	NSDNR, 2006a	X	NSDNR, 2006g
J	NSDNR, 2006b	Y	NSDNR, 2006h
K	NSDNR, 2006c	Z	NSDNR, 2006i
L	McClenaghan et al., 1992	AA	NSDNR, 2006j
M	Goodwin et al., 2002	AB	Murdoch and Sandilands, 1978
N	GSC, Unpublished	AC	Environment Canada, unpublished
O	Goodwin, 2003		

arsenic concentrations of about 18 ppm. Median arsenic concentrations range from 4 to 31 ppm for the Halifax Group and 5 to 48 ppm for the Goldenville Group, based on a compilation of whole rock laboratory and portable XRF analyses, but concentrations may be significantly higher in mineralized zones, with a maximum concentration of 13,000 ppm observed in a sandstone sample from the Green Harbour Formation of the Goldenville Group (Table 3a) (White, 2010).

Arsenic in granitoid rocks of the South Mountain Batholith may be derived from digested inclusions of Goldenville and Halifax group bedrock or from hydrothermal quartz veins in the granite (Ryan and Smith, 1998). The geochemical effects of granitic plutons intruding the surrounding rocks may also enhance arsenic occurrence in the host rock, with late stage magmatic and hydrothermal fluids associated with intrusive plutonic activity concentrating arsenic in pegmatites and along fractures in adjacent rocks. The arsenic in well water survey conducted in Hubbards found that the highest concentrations of arsenic were located along a fault-controlled zone (Hubbards Cove) where the granitic intrusions were youngest (Nova Scotia Department of Environment, 1978). The median arsenic concentration of granitoid rocks of the South Mountain Batholith, based on a compilation of whole rock laboratory and portable XRF analyses, is in the range of 2.3 to 2.7 ppm (Table 3a), which is comparable to the average literature level reported by Ure and Berrow (1982) of 1.5 ppm for various types of igneous rock.

Arsenic concentrations in sedimentary rocks vary by rock type, with typical concentrations reported to be in the range of 1-4 ppm for sandstones and carbonate/evaporite rocks, and 13 ppm for argillaceous deposits (Smedley and Kinniburgh, 2002). Boyle and Jonasson (1973) published average arsenic concentrations of 1-7 ppm in sandstone and sandy shale units in the Walton area of the Windsor Carboniferous basin of Nova Scotia, compared to concentrations of 2-15 ppm for argillaceous deposits and 2-5 ppm for carbonate/evaporite rocks. The highest concentrations of arsenic reported in the literature tend to be found in coal and bituminous deposits, ironstones and iron-rich rocks (Smedley and Kinniburgh, 2002).

In northern mainland Nova Scotia and Cape Breton Island pyritic metasedimentary rocks exist that are similar to units in the Goldenville and Halifax groups. In southeastern Cape Breton Island (Barr *et al.,* 1996) and in the northern Antigonish Highlands (White, 2012), Cambrian to Ordovician marine slate units locally contain significant sulphide-bearing rocks with minor ironstone units. In addition, throughout the Cobequid and Antigonish highlands, Silurian marine sedimentary rocks containing significant sulphide-rich beds and ironstone deposits are abundant (White, 2012; MacHattie and White, 2013). Although there are limited available data, these units have the potential to host significant concentrations of arsenic.

A number of regional surveys of arsenic and other pathfinder elements (e.g. Stea and Fowler, 1979; Woodman *et al*., 1994; Dunn and Balma, 1997) have been conducted in soils, tills, sediment, and biological media throughout the province for reconnaissance mineral exploration (Table 3b). Arsenic is commonly enriched in soil and tills down-ice from known mineralized zones due to mechanical dispersion (erosion, transportation and deposition) of the material by advancing glaciers, with dispersal distances ranging from hundreds of metres to several kilometres (Stea and Finck, 2001). Other regional soil surveys in Nova Scotia have focused on establishing background concentrations of arsenic (e.g. Goodwin *et al*., 2009; Parsons and Little, 2015) in comparison to the Canadian Soil Quality Guidelines (CSQG) (Canadian Council of the Ministers of the Environment, 2001). These surveys demonstrate that arsenic in soil and till is commonly found at levels exceeding the CSQG limit of 12 ppm arsenic (residential, parkland, agriculture, commercial and industrial lands) (Table 3b). Goodwin *et al*. (2009) reported that 56% of the sites sampled across the province (n=52) had arsenic concentrations greater than 12 ppm, with the highest concentrations generally found in soils overlying the Goldenville and

Halifax groups of southern mainland Nova Scotia. Bedrock geology has been identified as the main control on the variability of the regional distribution of arsenic in soils and tills (Grosz *et al*., 2004; Goodwin *et al*., 2009).

Anthropogenic Sources of Arsenic

Anthropogenic sources of arsenic in Nova Scotia include tailings (Grantham and Jones, 1977), wood preservation activities (Bamwoya *et al*., 1991), coal combustion and processing, and arsenical pesticide application, all of which can be locally important sources of arsenic in groundwater. Tailings are the main anthropogenic source of arsenic in groundwater documented in the literature, although there have been few studies in Nova Scotia investigating the transport of arsenic from tailings to regional groundwater systems (Drage, 2015). Arsenic concentrations well above background have been documented in tailings at various gold districts in the province, with concentrations as high as 312,000 mg/kg measured in arsenopyrite concentrates co-disposed with tailings in the Lower Seal Harbour gold district (Parsons *et al*., 2012). Pore water samples (n=181) collected from nine of the past-producing gold districts between 2003 and 2005 show median dissolved arsenic concentrations of 117 µg/L (range 0.2 to 6580 µg/L), compared to background levels of <25 µg/L (Parsons *et al*., 2012). DeSisto (2014) reported a median arsenic concentration of 620 µg/L (range <30 to 45,100 µg/L) for pore water samples (n=10) collected from the saturated zone in tailings at the Montague and Goldenville gold districts, and the maximum concentrations measured in shallow monitoring wells (i.e. <6 m deep) at these sites were 3,100 µg/L and 450 µg/L for Montague and Goldenville, respectively (C. J. MacLellan & Associates Inc., 2009). Recent studies have examined the stability of arsenic sequestered in hardpan tailing deposits (DeSisto *et al.,* 2011), and the bioavailability of the various forms of arsenic contained in the tailings (Meunier *et al*., 2010).

Elevated arsenic in soils (2 – 157 ppm) surrounding the former Sydney tar ponds and coke ovens site in Cape Breton County have been attributed to past coal combustion and emissions from steel-making activities (Lambert and Lane, 2004). A review of the available literature indicates that the relationship between coal combustion activities and groundwater contamination from arsenic has not been investigated in the vicinity of Nova Scotia's coal fired generators. Dummer *et al*. (2015) examined the relationship between agricultural land use and well water arsenic concentrations and did not find a significant regional correlation.

Geochemical Controls of Arsenic in Well Water

For arsenic to be present in well water, both a source of arsenic and the geochemical conditions for arsenic to be mobile in the groundwater system are needed. Arsenic concentrations exceeding drinking water guidelines in well water are relatively common in Nova Scotia because arsenic is abundant in various types of earth materials, mobile under the range of typical pH and redox conditions observed in our aquifers, and has a low drinking water limit relative to its abundance.

Microbially mediated and pH controlled oxidative dissolution of iron and arsenic sulphide minerals releases iron and arsenic into solution. For example, the most common mechanism for the solubilization of arsenic in Nova Scotia is the oxidation of arsenopyrite, releasing Fe(II), As(III) and SO_4^{2-} according to the following (Yu *et al.,* 2007):

$$FeAsS(s) + 11/4\ O_2 + 3/2\ H_2O \rightarrow Fe^{2+} + H_3AsO_3^0 + SO_4^{2-}$$

The Fe(II) is unstable in an oxic environment, and further oxidation releases Fe(III) and As(V):

$$Fe^{2+} + 1/4\ O_2 + H^+ \rightarrow Fe^{3+} + 1/2\ H_2O$$

In an alkaline environment, the Fe(III) will form hydrous ferric oxide particulates due to the low solubility of Fe(III) in these conditions, which can sorb or co-precipitate As(III) and As(V). Colloidal iron oxyhydroxide particulates are small enough to be transported in groundwater flowing through bedrock fractures and sorb arsenic along the groundwater flow path (Yang *et al*., 2014). Competition between arsenic and other sorbates such as PO_4^{3-} and SO_4^{2-} on clay and oxide minerals can therefore affect its mobility. At lower pH levels, the iron will remain in solution and arsenic will more readily sorb to other surfaces (e.g. clay minerals), which explains why an inverse relationship is often reported between iron and pH/arsenic at a given Eh (e.g. McCurdy, 1980). Under anoxic conditions, microbially mediated reductive dissolution of iron oxyhydroxides will release adsorbed or co-precipitated arsenic. Seawater intrusion to Nova Scotia's coastal aquifers could promote reducing conditions favourable for arsenic mobilization by this mechanism (Nath *et al*., 2008), potentially exacerbating arsenic in well water issues in areas of the province vulnerable to seawater intrusion.

The concentration and mobility of arsenic in groundwater will therefore be strongly influenced by groundwater chemistry, especially iron concentrations, Eh and pH, which evolve in the subsurface depending on the aquifer structure, mineralogy and regional groundwater flow dynamics. The regional hydrogeology of the province consists of a thin, discontinuous sediment cover, comprised mainly of till from the last glaciation, overlying shallow, fractured bedrock aquifers. Most water supply wells (~90%) in Nova Scotia are drilled wells in bedrock aquifers with complex fracture patterns, accessing groundwater from a depth of less than 100 m (Nova Scotia Environment, 2015). Groundwater flow patterns in fractured bedrock aquifers are controlled by the orientation, density, and hydraulic properties of the fractures, which vary by rock type and structural setting. For example, bedrock aquifers in crystalline rocks hosting arsenic store and transmit water only through intersecting fractures, some of which are coincident with mineralized veins, resulting in mixing of groundwater of varying chemistry from multiple flow paths and ages.

Arsenic concentrations are positively correlated with residence time in host aquifers since rock-water interactions tend to consume H^+, resulting in increased pH along the groundwater flow paths and the desorption of arsenic from iron oxyhydroxides under alkaline conditions. Laboratory studies have shown that arsenate desorption from a variety of substrates, including iron oxides and clay minerals, increases with increasing pH, while arsenite desorption from these same substrates requires higher pH conditions (Ayotte *et al.,* 2003; Stollenwerk, 2003). Studies have generally demonstrated that total arsenic concentration increases with well depth, since deeper wells are associated with longer flow paths and older, more alkaline groundwater (Nova Scotia Environment, 1978; McCurdy, 1980). Bottomley (1984) found, however, that elevated arsenic levels occurred over a range of different groundwater ages (less than 10 yr to 10,000 yr) based on isotopic analysis of groundwater from three sites underlain by Goldenville Group bedrock, indicating that younger groundwater is still susceptible to elevated levels of arsenic.

Analysis of temporal variation in arsenic in the Waverley study (McCurdy, 1980) found that arsenic levels in well water varied weekly in an unpredictable manner in the wells that were monitored over a year, which was attributed to variability in groundwater recharge, Eh, and pH conditions. The average variation in total arsenic concentration was 28% (based on 20 wells with arsenic levels ranging from 40 to 580 µg/L). The proportion of As(III) to As(V) in the samples, however, remained relatively stable.

Sampling and Analysis

Sampling and laboratory methods can also influence the concentration of arsenic measured in a well water sample. Bottomley (1984) found that dissolved arsenic concentrations increased with time of pumping based on pump test results from test wells installed in Waverley, Nova Scotia, and in Harvey, New Brunswick. Standing water sampled within the casing using a peristaltic pump was found to have much lower arsenic levels than formation water sampled below the casing using a submersible pump (after several well volumes removed), and was inversely related to iron. Bottomley (1984) attributed the observed relationship to the influence of the casing, with arsenic in the standing water adsorbing onto iron particulates from the casing, which were filtered out during sample collection. More recent research has found that Eh changes in the borehole as a result of pumping cycles (i. e. water level drawdown and recovery) can mobilize arsenic from the borehole and near-borehole rock and mineral surfaces (e.g. pyrite) (Ayotte *et al*., 2011).

Bottomley (1984) and McCurdy (1980) compared various sampling techniques, and showed that sample filtration and acidification yields the most representative concentrations of dissolved arsenic in groundwater, and is required to stabilize the As(III) to As(V) ratios in the water sample. It is recommended, however, that unfiltered acidified samples be collected for toxicological assessment of arsenic exposure to humans. Standard field and laboratory filtration methods have been found to remove around 5 – 10% of total arsenic (mostly arsenate) (Bottomley, 1984; Ayotte *et al*., 2003; Yang *et al*. 2014). Several early studies conducted in Nova Scotia evaluated laboratory techniques for the reliable measurement of inorganic arsenic in well water samples (e.g. McCurdy, 1980; Meranger *et al*., 1984). Early techniques involved the extraction of As(III) and As(V) using chemical reagents and measurement using atomic adsorption-spectrophotometric procedures, while today the speciation of arsenic is commonly measured using high performance liquid chromatography – inductively coupled plasma – mass spectrometry.

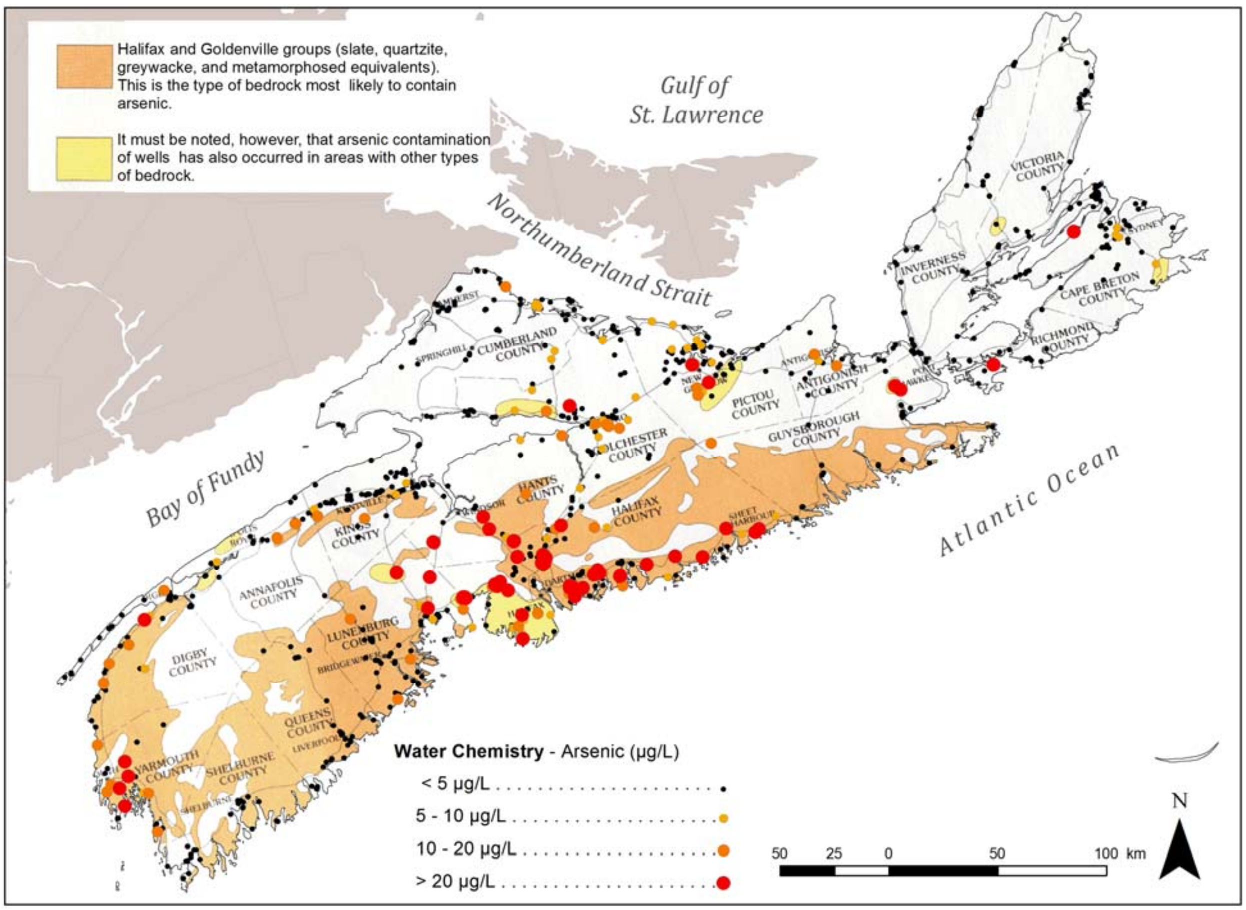

Figure 5. First generation arsenic in well water risk map for the province developed by the Department of Health. The distribution of arsenic concentrations in well water from the Nova Scotia Groundwater Chemistry database is shown for comparison.

Risk Management

Various activities have been conducted by the Province of Nova Scotia to manage the risk of arsenic in well water. These include well water surveys and risk mapping, assessment of potential arsenic in well water hazards as part of subdivision planning and approval, provision of water servicing to arsenic impacted areas, assessment of small-scale arsenic treatment technology, and outreach and education, including the promotion of routine water testing by homeowners.

Risk Mapping

In addition to helping well owners identify areas that are more likely to have elevated arsenic levels, risk maps can be useful for land development planning, groundwater supply development planning, risk communication, and strategic allocation of government resources for risk mitigation. Since new housing development using private wells continues to be concentrated in areas where groundwater may contain elevated levels of arsenic, especially in Halifax, Hants and Lunenburg counties, risk maps are important tools for land-use planning and communication of the relative risk of arsenic in well water to homeowners.

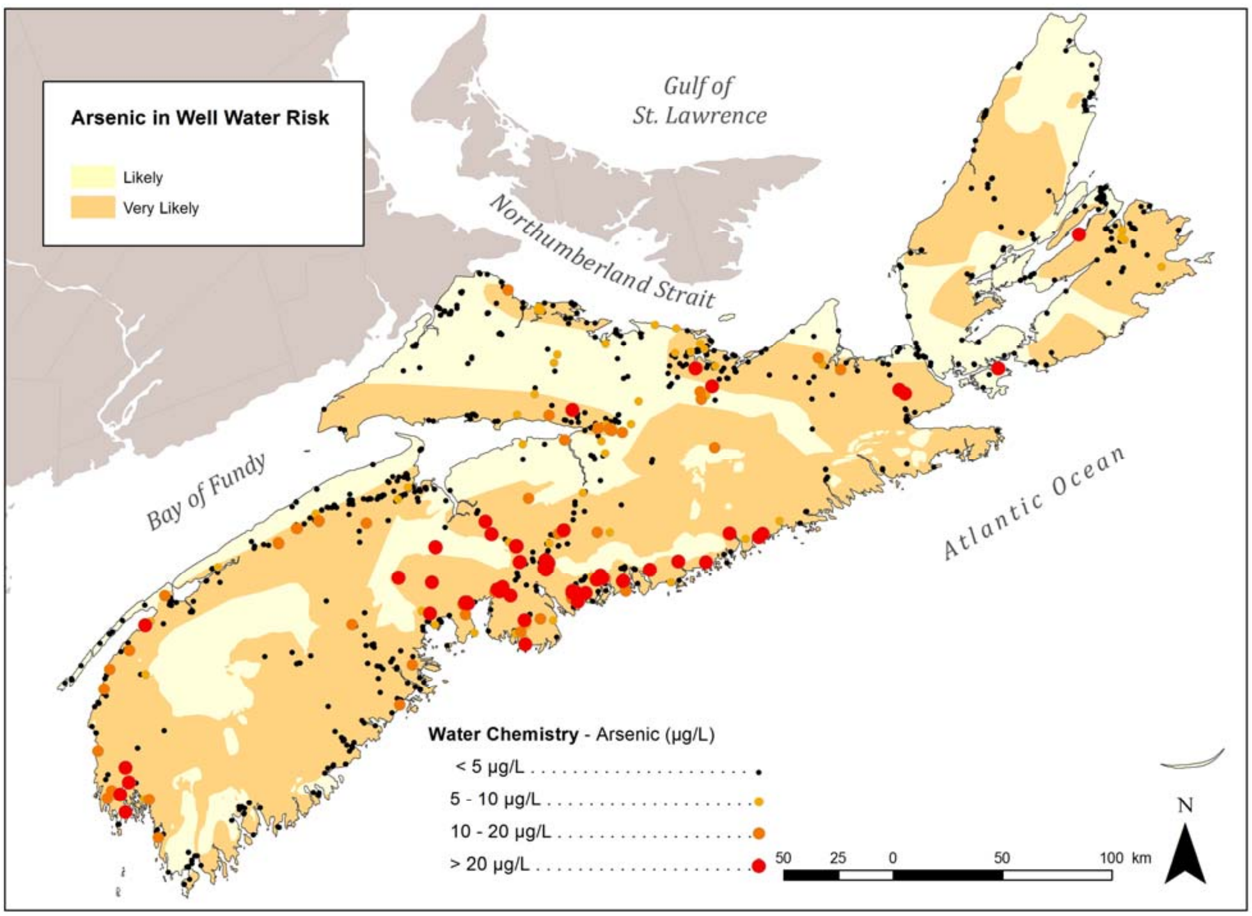

Figure 6. Second generation arsenic in well water risk map for the province developed by the Arsenic Working Group. The distribution of arsenic concentrations in well water from the Nova Scotia Groundwater Chemistry database is shown for comparison.

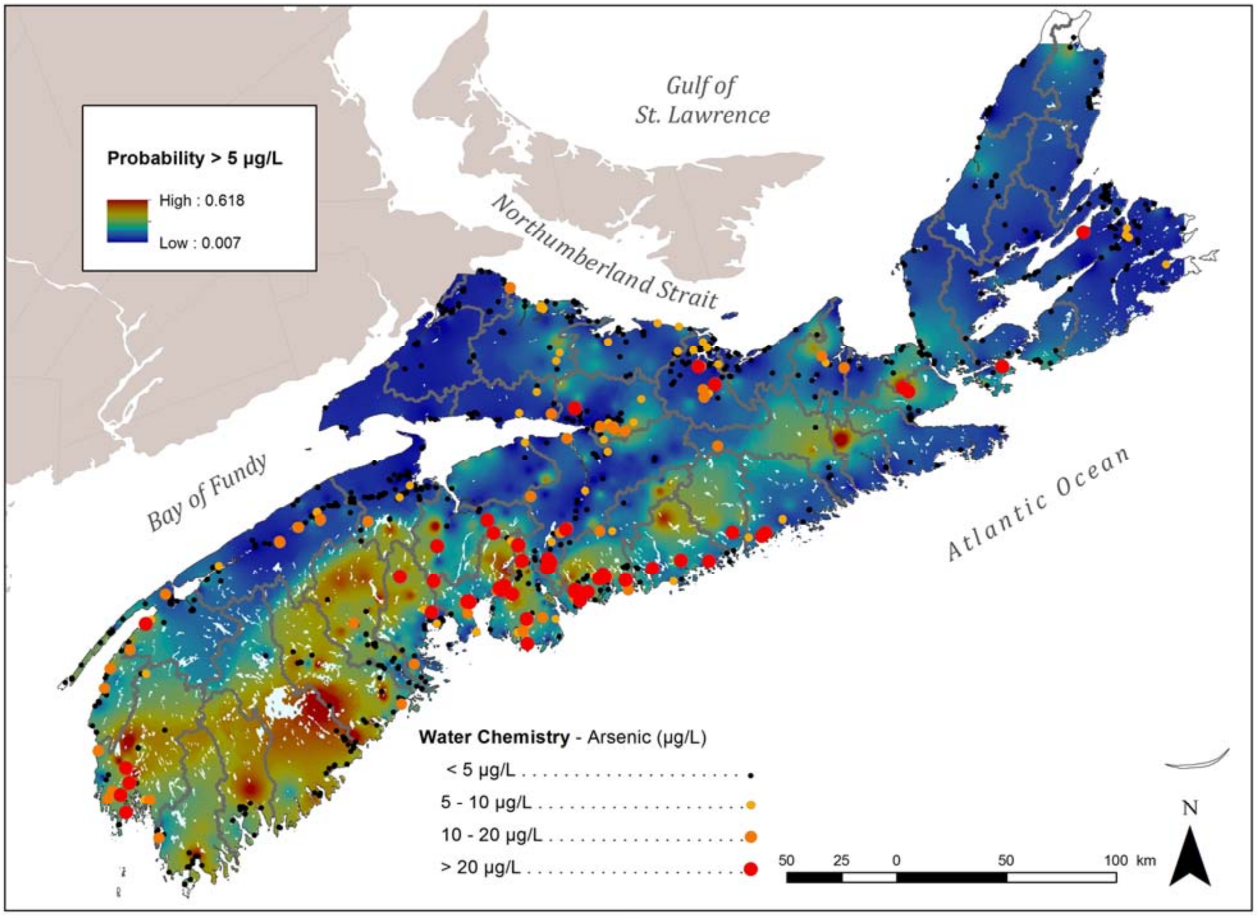

Figure 7. Interpolated probability of arsenic concentrations ≥5 µg/L in well water samples based on the QEII laboratory arsenic dataset (after Dummer *et al.*, 2015). The distribution of arsenic concentrations in well water from the Nova Scotia Groundwater Chemistry database is shown for comparison.

The Nova Scotia Department of Health (1983) developed the first generation of arsenic in well water risk mapping for the province (Fig. 5). The map identifies areas of the province that are more 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.

A provincial Arsenic Working Group was formed in 2004 to evaluate the impacts of lowering the arsenic maximum acceptable concentration from 25 µg/L to 10 µg/L, as proposed by the Federal-Provincial-Territorial Committee on Drinking Water. One of the objectives of the working group was to update the arsenic in well water risk map (Nova Scotia Environment, 2005a). The approach relied on using various existing digital data sets with arsenic geochemical data (stream sediment, biogeochemical surveys, till, rock, soil and lake sediments) (Fig. 6). The data were normalized using percentiles for comparison purposes so that multiple datasets representing various sample media could be combined and plotted. Using the QEII dataset, water well levels that exceeded the proposed arsenic in drinking water guidelines (i.e 10 µg/L) were assigned a percentile value of 100, as were arsenopyrite and gold occurrences and shafts (indicating gold-arsenic mineralization), and Halifax and Goldenville group bedrock. All data exceeding the 95th percentile were plotted and clusters were manually outlined by visual inspection after using contouring software. Areas with grouped percentile values >95 were considered to be 'more likely' to have elevated levels of arsenic in well water, and all other areas were considered 'less likely' (Nova Scotia Environment, 2005b).

More recently, as part of the Atlantic PATH project, two separate geostatistical process models using stepwise multivariable logistic regression were developed to predict areas of the province with high toenail arsenic concentrations (defined as toenail arsenic levels ≥0.12 µg/g), and areas of the province with high well water arsenic concentrations (defined as well water arsenic levels ≥5 µg/L) (Fig. 7) (Dummer *et al*., 2015). These predictive models can be useful where direct measurements of arsenic body burden or arsenic exposure are not available. Information on socio-demographic, lifestyle and health factors was obtained from Atlantic PATH study participants with a set of standardized questionnaires, while anthropometric indices and arsenic concentrations in well water and toenails were measured. The well water arsenic concentration process model, however, used the older, biased QEII arsenic dataset due to the greater spatial coverage of these data compared to the Atlantic PATH primary arsenic well water data.

The study found that the geological and environmental information used to predict well water arsenic concentrations could also be used to accurately predict toenail arsenic concentrations, and concluded that the geological and environmental factors causing arsenic contamination in well water are the major contributing influences on arsenic body burden among Nova Scotians. Regression of the QEII arsenic in well water data with various environmental and geological variables found that significant predictors of a lower probability of elevated arsenic in well water include the volcanic and sedimentary groundwater regions, and Halifax and Windsor group bedrock, whereas significant predictors of a higher probability of elevated arsenic in well water include Horton Group bedrock, proximity to gold districts (<5 km) and areas of stony till coverage. Surprisingly, a significant relationship between the Goldenville Group and the probability of elevated arsenic in well water was not found during multivariate regression, which may be due to a confounding spatial association of gold districts with Goldenville Group bedrock (Dummer *et al*., 2015).

To generate a province-wide map (Fig. 7), the probability of arsenic ≥5 µg/L in well water was predicted using the process model at each geographic location with available arsenic in well water data, and then these probabilities were interpolated across the province using the Inverse Distance Weighting technique at a cell resolution of 500 m x 500 m.

Public and Municipal Water Supplies with Arsenic Issues

There are presently no municipal water systems in the province specifically treating for arsenic, although a small system in the community of Fall River (Miller Lake subdivision) has an arsenic removal system servicing approximately 44 households. In the Village of Lawrencetown, located in the Annapolis Valley (Fig. 4), wellfield blending is used to lower arsenic to safe levels while meeting the community's water demands. Municipal servicing from surface water sources has also been extended to some areas (e.g. community of Waverley) to mitigate arsenic exposure in subdivisions, or small-scale surface water systems have been developed as an alternative to well water with unsafe levels of arsenic (e.g. Collins Park subdivision in the community of Wellington, Fig. 3). The cluster of private drilled wells with high levels of arsenic detected in Lower Sackville were all located within a central water servicing boundary, and residents complied with the Nova Scotia Department of Health's advice to discontinue the use of their wells for cooking and drinking and connect to the existing central water system.

Public water supplies are required to regularly test their water in accordance with the Guidelines for Monitoring Public Drinking Water Supplies (Nova Scotia Department of Environment, 2005) and if arsenic is detected above GCDWQ limits in well water, corrective action is required, which would likely involve water treatment, well modification or the development of an alternate potable water source.

Private Water Supplies with Arsenic Issues

Following the detection of arsenic in private wells in the 1970s, the viability of small-scale water treatment for arsenic removal in Nova Scotia was explored. Lutwick (1980) prepared a report for the Nova Scotia Department of Environment summarizing three previous reports by the Nova Scotia Research Foundation Corporation investigating arsenic removal from private wells. Laboratory and field testing programs were completed during these investigations. The field testing included the installation of treatment units in 19 homes in Waverley, Wellington and Lawrencetown (Halifax County). Treatment units were evaluated for both As(III) and As(V) removal, and the study recommended ion exchange as a practical and economical arsenic treatment method for private water wells in Nova Scotia, involving an initial step where As(III) is oxidized to As(V) using hypochlorite.

Today, point-of-use reverse osmosis (RO) systems are commonly used for the removal of arsenic from drinking water in small residential systems due to its relative affordability, effectiveness in treating multiple contaminants and low maintenance requirements (United States Environmental Protection Agency, 2000). Laboratory and field-testing results have reported that RO filters can reduce arsenic concentrations by up to 99% (United States Environmental Protection Agency, 2000), but this performance is only achievable for As(V) removal. Treatment for As(III) usually requires pre-treatment with an oxidation unit to convert As(III) to the more easily removable As(V). Since arsenic species are not typically analyzed, it is recommended that both raw and treated water are tested regularly to ensure that treatment equipment is effective. Reverse osmosis point-of-entry (i.e. all household water is treated) systems are not as widely used due to the cost and flow requirements of these systems, and because Health Canada's arsenic MAC applies only to the ingestion of water (Health Canada, 2014). New treatment technologies that are viable, robust and easy to implement for small treatment systems continue to be explored. For example, the re-use of municipal water treatment residual solids was recently investigated by Gibbons and Gagnon (2010) as a potential treatment media for small-scale arsenic treatment of Nova Scotia groundwater. The study found that the best adsorption results of arsenate in lab-scale testing were achieved with ferric and lime water treatment residual solids, due to their affinity for arsenic adsorption. Rahaman *et al.* (2008) tested the capacity of various locally available biomaterials and agro products to adsorb As(III) and As(V), and found that of the materials tested, Atlantic Cod fish scales had the maximum capacity for removing both species of arsenic.

As noted earlier, private water supplies are not regulated, but the province recommends that homeowners test their well water quality every two years, or if there are any noticeable changes in water quality. The recommendation for routine testing of private wells by homeowners has been promoted by the Nova Scotia government through various education and outreach initiatives since arsenic was first detected in the 1970s. For example, the province mailed form letters to individual homeowners to advise them of arsenic risks and to provide recommendations for appropriate mitigation (e.g. treatment options) following residential well water surveys led by the province, or following the submission of arsenic in well water results to the DOH between 1981 and 1994. An arsenic in drinking water webpage is presently maintained by NSE and a factsheet is available online, providing private well owners with recommendations on appropriate testing and treatment of well water with unsafe levels of arsenic (Nova Scotia Environment, 2008).

In addition to the 2004 provincial Arsenic Working Group noted earlier, an inter-departmental provincial-federal 'Historic Gold Mines Advisory Committee' was established in 2005 to evaluate the potential ecological and human health risks associated with tailings in historic gold producing areas found in the Meguma terrane, including the risk of arsenic contamination to private water supplies. The activities of the committee have focused mainly on the characterization of tailings in these gold producing areas to support risk assessments and are summarized in Drage (2015).

Despite these efforts, research has shown that many private well owners in Nova Scotia do not test their well water. Chappells *et al*. (2014) recently evaluated the testing behaviours and knowledge of arsenic risk exposure among a demographic cross-section of well users residing in five distinct geographic areas of the province assessed to be at varying risk levels with respect to arsenic occurrence in groundwater based on water sample analyses. An integrated knowledge-to-action methodological approach was used to assess the personal, social and local factors that influence the perception of well water contaminant risks and the translation of knowledge into routine water testing behaviors. Analysis of the survey data found that only 12% of respondents (n = 420) tested their well water as per the provincial recommendation. Survey respondents had a high level of confidence in their well water quality (74% describing water quality as 'good' or 'very good'), which was unrelated to the relative risk of arsenic exposure or to routine testing behaviours. Based on the survey responses and in-depth well user interviews (n = 32), the study concluded that the assessment of risk among well users was mainly influenced by personal experience, local knowledge, social networks and convenience of infrastructure rather than the communication of risk information through formal channels. To improve public risk knowledge, the authors recommend various government and community level interventions, combined with improved knowledge and communication of the local occurrence of arsenic to support individual risk assessment and water testing.

Arsenic Hazard Assessment in New Subdivisions

Starting in the 1970s, applications for subdivisions using private wells for water supply were reviewed by NSE staff, who provided comment on proposed water supplies such as the type of well most suitable for the development, the availability of groundwater, and potential water quality problems. The recommendations were not enforceable and the reviews were discontinued in the late-1980s. In 2006, Halifax Regional Municipality (HRM) implemented guidelines for groundwater assessment for new subdivisions as part of open space development projects, which is a form of development designed to conserve open space and typically uses private wells for water supply. The guidelines include a requirement to install test wells, collect water chemistry information and recommend mitigation measures if water quality issues, such as arsenic, are detected. A provincial version of the guidelines for groundwater assessment for subdivision development was released in 2011 (Nova Scotia Environment, 2011) and adopted by HRM, although it is not known how widely this process is used for subdivision planning in the rest of the province.

Summary and Perspectives

Considerable research and risk management activities have been conducted in the province with respect to arsenic exposure in water wells since its initial detection in the community of Waverley in the 1970s. The province has conducted extensive surveys of arsenic in various media (till, sediment, bedrock, groundwater), primarily in areas underlain by bedrock related to the Goldenville and Halifax groups or South Mountain Batholith, and has compiled arsenic concentration data from water wells across Nova Scotia. Drilled wells are associated with a significantly higher probability of elevated levels of arsenic compared to dug wells. Dug wells in past-producing gold districts, however, are associated with higher exceedance rates of the Health Canada MAC for arsenic in drinking water compared to dug wells in areas outside of gold districts. Exceedance rates are generally around 10% for drilled wells (bedrock aquifers) across the province, although much higher exceedance rates (>50%) have been reported in selected communities, especially those underlain by Goldenville Group bedrock between Dartmouth and Lake Echo (Fig. 3). Limited arsenic speciation measurements suggest that the dominant species of arsenic (>80%) present in Nova Scotia well water is inorganic As(V).

The main anthropogenic source of arsenic in Nova Scotia has been identified as tailings and waste rock in past-producing gold districts in the Goldenville and Halifax groups of the Meguma terrane. Geogenic sources of arsenic are widespread, consisting primarily of arsenopyrite and arsenian pyrite. Arsenic concentrations in well water, which have been found to range over four orders of magnitude (up to 5000 µg/L), mainly depend on the availability of arsenic in the geologic materials in contact with groundwater, the aquifer geochemistry (e.g. redox and pH environment, presence of iron oxides and competitive ions), and the aquifer structure (e.g. fracture flow dynamics). A conceptual model for arsenic occurrence in water wells involves the oxidation of arsenic-bearing sulphide minerals in the oxic portion of the groundwater flow path, releasing iron and arsenic, followed by iron oxidation to form iron oxyhydroxide particulates, which are transported in fractures and sorb arsenic along the flow path until intercepted by boreholes (Yang *et al.*, 2014). In the anoxic portions of the flow path, reductive dissolution of hydrous ferric oxides could lead to enhanced arsenic mobility. The complexity of Nova Scotia's fractured bedrock aquifers with respect to fracture flow and host mineralogy contributes to the spatial and temporal heterogeneity of arsenic in groundwater at local and regional scales, and additional research is warranted to advance our understanding of these relationships.

Epidemiological studies have shown that arsenic in well water is the main contributing factor to arsenic body burden among Nova Scotians. Given that arsenic ingestion is associated with a range of adverse health effects, including skin and internal cancers, a significant risk persists due largely to the behaviours and risk perception of private well users. Arsenic can usually be removed from well water using conventional point of use reverse osmosis treatment systems, although the mitigation of arsenic in private wells is the responsibility of individual homeowners. Effective public awareness programs, including risk maps, are therefore essential to manage the risk of arsenic exposure and to reduce the associated adverse health effects and long-term medical costs. Future efforts by the Nova Scotia Department of Natural Resources will be directed toward the development of a refined risk map and web mapping application to better communicate the risk of arsenic in drinking water to private well users.

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Appendix A
Compiled Exceedance Rates for Arsenic in Drinking Water (>10 µg/L) for Selected Communities Based on Data from the Nova Scotia Department of Health and the Queen Elizabeth II Hospital Datasets

		All Data					***Detects Only***					
Community	Info Source	Number of Observations	Number of Detects	Per cent of Detects	Per cent Exceedance[2]	K-M[1] Mean (µg/L)	Min (µg/L)	Median (µg/L)	95%ile (µg/L)	Max (µg/L)	Mean (µg/L)	Median (µg/L)
Amiraults Hill	QEII	43	32	74%	65%	55.9	2	28	188.0	400	74.4	38
Black Point	QEII	26	9	35%	31%	25.0	2	2	86.8	185	68.6	62
Brookside	DOH	33	31	94%	48%	25.0	5	10	79.0	110	26.3	12
	QEII	31	15	48%	19%	7.4	2	2	23.0	60	13.1	10
Chester Basin	QEII	66	37	56%	32%	30.2	2	2	115.8	340	52.3	20
Cole Harbour	DOH	41	37	90%	71%	56.3	5	23	150.0	520	61.8	30
	QEII	20	11	55%	35%	22.4	2	2	99.3	180	39.1	17
Country Harbour Mines	QEII	16	11	69%	56%	41.5	2	16	134.5	430	59.5	30
Cow Bay	DOH	15	15	100%	87%	52.2	6	26	140.0	140	52.2	26
	QEII	62	29	47%	26%	16.0	2	2	86.3	240	31.8	11
Crousetown	QEII	17	13	76%	59%	32.1	2	13	106.6	129	41.3	28
Dartmouth	DOH	61	52	85%	54%	44.1	5	20	160.0	360	50.9	30
	QEII	67	29	43%	21%	11.4	2	2	56.6	124	23.7	10
East Chezzetcook	DOH	3	3	100%	100%	96.3	20	31	217.3	238	96.3	31
	QEII	63	21	33%	21%	35.8	2	2	239.8	250	103.4	21
East Dover	QEII	20	13	65%	45%	30.1	2	8.5	86.6	116	45.2	26
East Petpeswick	DOH	5	5	100%	80%	75.4	5	70	163.2	180	75.4	70
	QEII	50	26	52%	32%	22.7	2	3	102.3	210	41.8	16
East Preston	DOH	20	20	100%	95%	111.1	7	44.5	188.5	1300	111.1	44.5
	QEII	49	33	67%	47%	25.8	2	7	81.6	181	37.3	29
Eastern Passage	DOH	22	22	100%	82%	82.8	5	35	295.5	700	82.8	35
	QEII	53	21	40%	15%	8.2	2	2	29.4	131	17.6	7
Fall River	DOH	191	126	66%	45%	21.2	5	10	65.0	300	29.6	20
	QEII	449	183	41%	19%	10.33	2	2	45.8	280	22.45	10
Glen Margaret	QEII	53	30	57%	32%	35.4	2	3	155.0	260	61.0	30
Gold River	QEII	70	36	51%	29%	25.0	2	2	138.4	350	46.7	16.5
Halifax	DOH	14	13	93%	36%	20.4	5	9.5	57.7	70	21.5	10
	QEII	9	1	11%	0%	2.3	2	2	3.8	5	5.0	5
Head of Chezzetcook	DOH	4	4	100%	75%	62.8	10	78	84.4	85	62.8	78
	QEII	108	49	45%	32%	45.6	2	2	220.0	1500	98.0	26
Head Of St Margarets Bay	DOH	30	30	100%	63%	57.1	5	29.5	185.5	210	57.1	29.5
	QEII	57	19	33%	16%	15.9	2	2	65.6	380	43.6	10
Herring Cove	DOH	13	13	100%	62%	54.7	6	20	204.0	420	54.7	20
	QEII	54	27	50%	28%	25.1	2	2	71.0	420	48.1	11
Hubbards	DOH	5	4	80%	60%	85.4	5	20	301.2	370	105.5	23
	QEII	314	82	26%	12%	18.2	2	2	37.7	1380	64.2	10
Hubley	DOH	72	71	99%	67%	37.5	5	28.5	110.0	250	38.0	30
	QEII	62	43	69%	42%	14.7	2	8	54.2	69	20.3	14

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Appendix A. continued

Community	Info Source	Number of Observa-tions	Number of Detects	Per cent of Detects	Per cent Exceedance[2]	K-M[1] Mean (µg/L)	Min (µg/L)	Median (µg/L)	95%ile (µg/L)	Max (µg/L)	Mean (µg/L)	Median (µg/L)
Lake Charlotte	QEII	14	5	36%	21%	36.2	2	2	176.4	320	97.8	62
Lake Echo	DOH	26	25	96%	85%	38.1	5	27	82.3	130	39.4	30
	QEII	352	142	40%	24%	13.8	2	2	63.9	510	31.3	13
Lakelands	DOH	10	10	100%	80%	39.6	6	20	101.0	110	39.6	20
	QEII	8	5	63%	38%	9.4	2	6	23.2	27	13.8	14
Lawrencetown	DOH	109	98	90%	77%	74.9	5	37	306.0	650	82.7	40
	QEII	117	73	62%	41%	32.0	2	6	129.2	640	50.0	22
Lower Sackville	DOH	52	37	71%	42%	26.5	5	10	83.5	300	35.2	20
	QEII	33	13	39%	12%	6.5	2	2	13.4	97	13.3	6
Middle New Cornwall	QEII	12	8	67%	42%	27.1	2	4.5	120.0	120	39.6	18
Mineville	DOH	39	38	97%	85%	53.6	5	30	162.0	216	54.8	30
Molega	QEII	17	10	59%	59%	63.7	2	12	256.4	362	106.8	45
Montague Gold Mines	DOH	11	9	82%	73%	59.3	5	30	215.0	350	71.3	30
	QEII	9	8	89%	89%	515.1	2	82	2420.0	3900	579.3	96
Mooseland	QEII	10	7	70%	40%	31.5	2	4	115.0	119	44.1	14
Porters Lake	DOH	20	16	80%	60%	28.5	5	24.5	49.4	151	34.4	30.5
	QEII	474	165	35%	18%	14.2	2	2	79.1	400	37.1	12
Prospect	DOH	30	30	100%	77%	284.4	6	40	1814.0	2300	284.4	40
	QEII	89	59	66%	42%	66.4	2	6	181.6	1800	99.2	16
Rhodes Corner	QEII	30	22	73%	43%	20.3	2	7	65.1	85	26.9	16.5
Seabright	QEII	68	41	60%	40%	30.22	2	4	148.8	300	48.8	19
Ship Harbour	QEII	25	13	52%	32%	42.2	2	2	276.0	300	79.2	24
South Brookfield	QEII	41	21	51%	37%	23.5	2	4	97.0	116	44.0	24
Spry Harbour	QEII	18	8	44%	39%	70.2	2	2	386.5	650	155.4	55
Tantallon	DOH	7	4	57%	0%	6.3	5	5	9.4	10	7.3	7
	QEII	155	85	55%	30%	11.8	2	2	47.6	112	19.9	12
Tusket	QEII	10	7	70%	70%	22.5	2	28	40.0	40	31.3	34
Upper Lakeville	QEII	11	7	64%	27%	43.2	2	4	175.0	250	66.7	9
Upper Lawrencetown	QEII	16	12	75%	50%	29.4	2	11.5	101.5	115	38.5	31.5
Upper Sackville	DOH	70	57	81%	63%	25.1	5	20	70.0	120	29.7	20
	QEII	60	37	62%	20%	7.7	2	2.5	36.2	60	12.3	6
Waverley	DOH	73	50	68%	37%	16.3	5	9	44.0	116	21.5	20
	QEII	85	37	44%	22%	10.7	2	2	59.6	89	21.9	12
Wellington	DOH	35	19	54%	37%	28.6	5	5	115.3	200	48.4	30
	QEII	132	48	36%	12%	9.4	2	2	33.5	120	22.3	7.5
Westphal	DOH	16	12	75%	38%	29.1	5	9.5	105.0	300	37.2	10.5
	QEII	14	9	64%	50%	16.6	2	9	42.9	65	24.7	26
Whites Lake	DOH	14	14	100%	50%	22.0	5	15	53.5	60	22.0	15
	QEII	83	46	55%	27%	7.6	2	3	25.8	49	12.2	9
Wine Harbour	QEII	13	6	46%	31%	35.6	2	2	174.6	345	74.8	17
TOTAL		**4696**	**2529**	**54%**	**33%**							

1. Kaplan-Meier

2. Exceedance of Health Canada's MAC for arsenic in drinking water (10 µg/L)

QEII: Queen Elizabeth II Hospital laboratory dataset, typical detection limit was 2 µg/L

DOH: Nova Scotia Department of Health dataset, typical detection limit was 5 µg/L