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G. W. Kennedy and J. Drage

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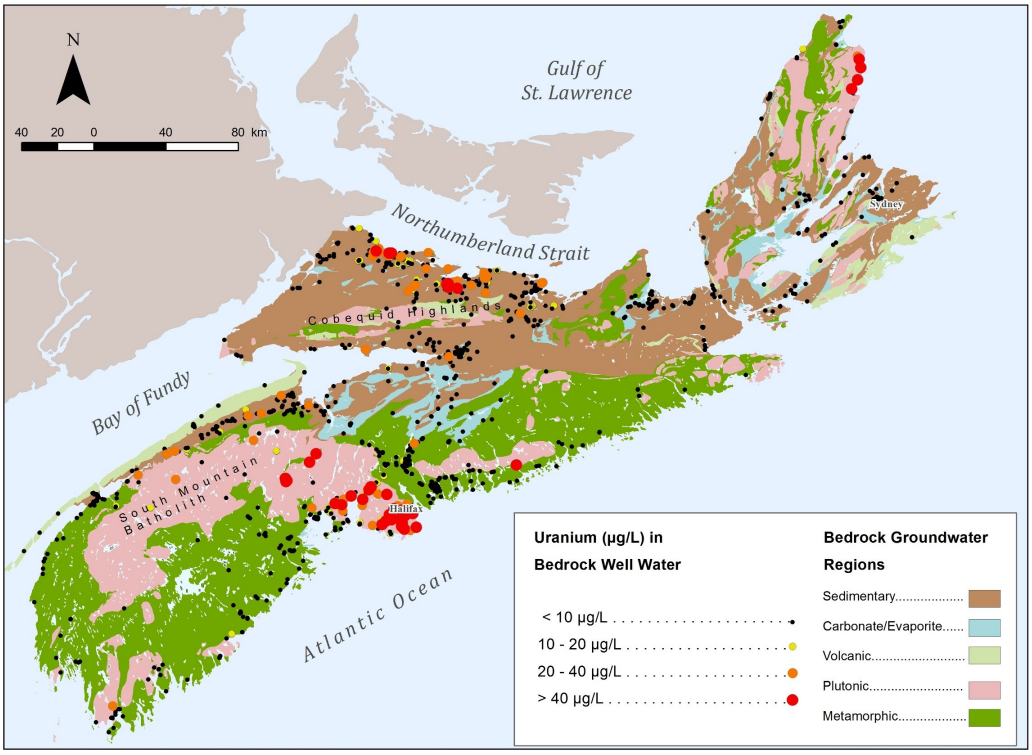

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Introduction

Human health concerns relating to the presence of naturally occurring uranium in Nova Scotia well water first emerged in the late 1970s based on epidemiological testing, similar to the discovery of the issue of arsenic in Nova Scotia well water. During a 1977 study by Dalhousie University on the concentrations of various heavy metals in the general population, elevated levels of uranium were found in random hair samples of a particular subject relative to levels found in other subjects (Moss et al., 1983). The elevated uranium was linked to exposure in well water in a mobile home park in the community of Harrietsfield, Halifax County (Fig. 1), where the concentration of uranium in well water was 70 µg/L. At the time of the Dalhousie University study, the guideline for drinking water was 5000 µg/L based on aesthetic considerations (water discolouration) (Joint Committee on Drinking Water Standards, 1969). Shortly after the discovery of uranium in Harrietsfield well water, the Federal-Provincial Working Group on Drinking Water (1979) released a health-based guideline of 20 µg/L.

During the 1970s there was strong interest in uranium mineral exploration, mainly to meet the demand for uranium as the raw material used in the production of fuel for nuclear reactors. Emerging genetic models of uranium enrichment led to extensive exploration activity in Nova Scotia, with the total area of Nova Scotia under uranium exploration licence peaking in 1977 (Interdepartmental Uranium Committee, 1994). Mineral exploration activities often included well water surveys as a reconnaissance-scale geochemical-mapping technique to identify zones of anomalous uranium and radon concentrations, which could potentially indicate the presence of a uranium deposit. All uranium exploration, however, ceased in 1981 following the announcement of a moratorium on uranium mineral exploration and mining in response to mounting public concern regarding the health and environmental impacts of these activities (Commission of Inquiry on Uranium, 1985). An inquiry was commissioned in 1982 to explore the issues surrounding uranium exploration and mining, which involved a broad consultation with Nova Scotians consisting of 44 public meetings across the province (Commission of Inquiry on Uranium, 1985) and a technical submission by the Nova Scotia Department of Mines and Energy (1982). An interdepartmental committee was formed pursuant to a recommendation from the Uranium Inquiry to investigate some of the issues raised during the consultation and a report was produced to address these issues (Interdepartmental Uranium Committee, 1994). In 2009, the moratorium became a legislated ban under the *Uranium Exploration and Mining Prohibition Act*, prohibiting all uranium mining and uranium exploration activity in Nova Scotia.

In addition to mounting public concern regarding uranium exploration and mining, there was growing concern in the late 1970s regarding naturally occurring uranium in well water in the Harrietsfield area (Grantham, 1986). An inter-departmental task force was appointed in 1979 to investigate the occurrence of uranium in drinking water supplies in Nova Scotia (Grantham, 1986). The goal of the Uranium Task Force was to ensure that the health of Nova Scotians was not being adversely affected by the presence of uranium and radionuclides, such as radium and radon, in their drinking water. The task force employed a multifaceted approach, which included surveys of uranium in private-well drinking water. The survey work was initially focused in the communities of Harrietsfield and Williamswood in Halifax County (Fig. 1), but was later expanded to include a limited number of communities with similar granite aquifers, and sedimentary aquifers in northern Nova Scotia where mineral exploration work had

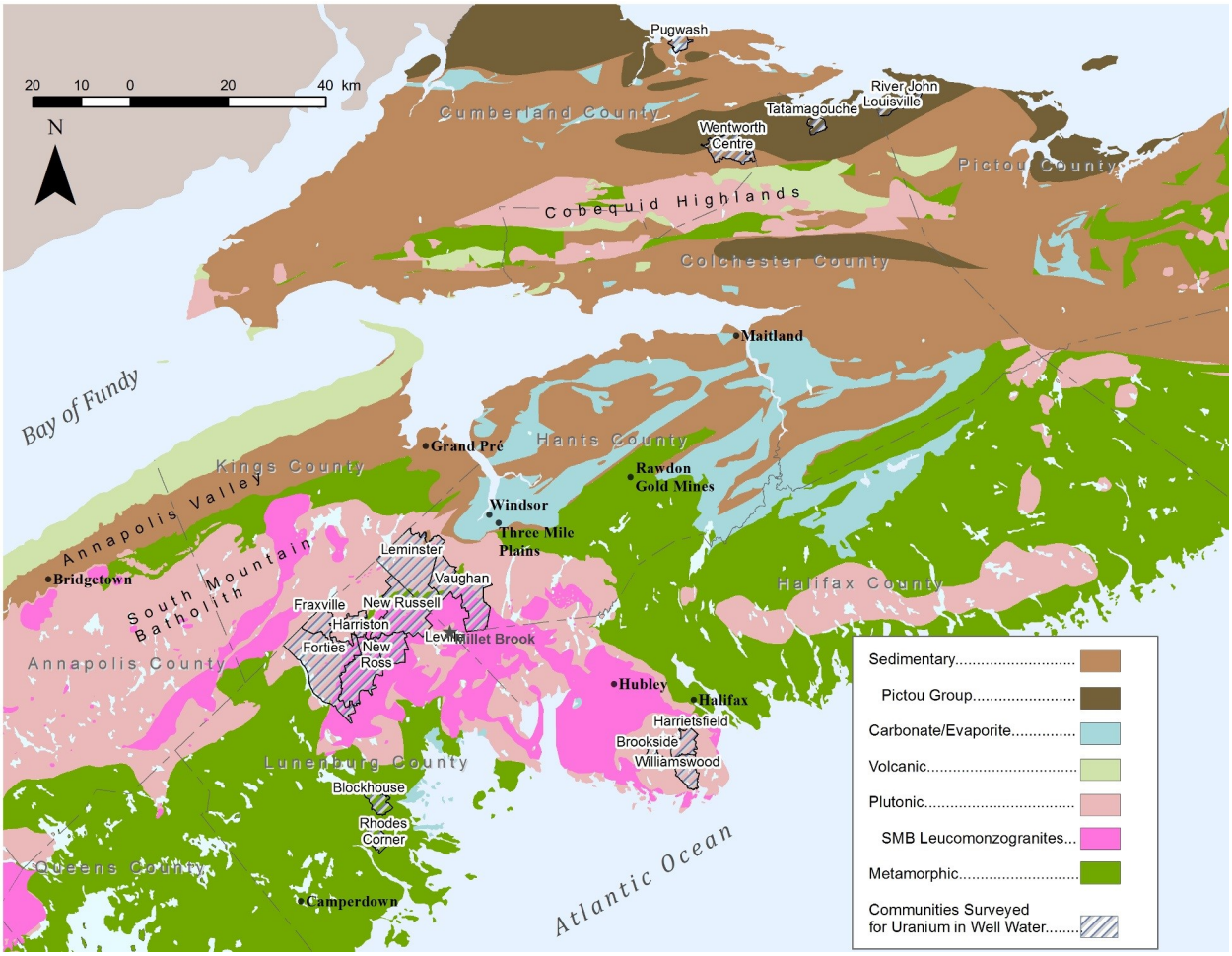

Figure 1. Locations of uranium in well water surveys conducted by the Province of Nova Scotia compared to the province's five major bedrock groundwater regions. The Pictou Group in the sedimentary groundwater region and South Mountain Batholith (SMB) leucomonzogranites in the plutonic groundwater region are highlighted darker shades of brown and pink, respectively.

previously identified uranium concentrations greater than 20 µg/L in water wells (Fig. 1) (e.g. Brummer, 1958; Dyck et al., 1976; Guardia and MacNabb, 1977).

The Task Force findings were documented in a report titled *The Occurrence and Significance of Uranium, Radium, and Radon in Water Supplies in Nova Scotia* (Grantham, 1986), which included a separate report on *The Hydrogeology and Distribution of Naturally-Occurring Uranium in Well Water in Nova Scotia* (MacFarlane, 1983) as an appendix. Key findings of the Uranium Task Force included confirmation that the source of uranium in well water was geogenic and was mainly associated with bedrock aquifers (99% of the surveyed wells with uranium concentrations greater than 20 µg/L were drilled wells). The task force also found that elevated levels of uranium in well water were most likely to occur in granite batholiths and in alkaline sandstone and shale rocks in the upper Carboniferous basin in northwestern Nova Scotia, down-gradient of the Cobequid Highlands. The Task Force investigated radium and radon in well water, but because few exceedances of the drinking water quality guideline for radium were detected, and radon in well water is not considered a health hazard, these radionuclides were not considered a significant concern with respect to drinking water exposure.

Since the initial discovery over 40 years ago of uranium in water wells in Nova Scotia at concentrations that could adversely affect human health (Moss et al., 1983), various studies have been conducted into

the occurrence and controls of uranium in the province's aquifers (e.g. MacFarlane, 1983; Kronfeld et al., 2004; Samolcyk et al., 2012; Drage and Kennedy, 2013). Naturally occurring uranium in well water remains a significant concern, especially for private well owners in regions of the province where uranium mineralization is present. The purpose of this report is to summarize the available research related to uranium occurrence in water wells in Nova Scotia, including the following: (1) human health effects and exposure pathways of uranium; (2) sources of uranium in well water data and the distribution of uranium in Nova Scotia groundwater; (3) controls on the occurrence and mobility of uranium in well water; and (4) strategies for managing the risk of uranium exposure in private wells in Nova Scotia.

Health Effects and Exposure

Common pathways for human exposure to uranium include air, soil, and food, but drinking water is considered to have the most significant impact on human health (World Health Organization, 2012). Uranium, a weakly radioactive heavy metal, has the potential to be both chemically and radiologically toxic, but of principal concern with respect to the ingestion of uranium in well water are the toxic effects of uranium on kidneys (chemical toxicity) (Health Canada, 2017a). Long-term ingestion of uranium by humans can adversely affect kidney function at the levels of uranium found in some groundwater supplies. Due to the element's radioactivity, there has also been concern that the ingestion of uranium in drinking water may be associated with an increased risk of cancer; however, the epidemiological research to date has not demonstrated a clear linkage (Health Canada, 2009). Health Canada (2009) has calculated that there may an elevated risk of cancer when uranium in drinking water exceeds 119 µg/L.

Various animal (e.g. Gilman et al., 1998) and human (e.g. Limson Zamora et al., 1998) epidemiological studies have correlated kidney bioeffects (nephrotoxicity) with uranium exposure, and the current Health Canada (2017b) maximum acceptable concentration (MAC) of 20 µg/L uranium in drinking water is based on studies of kidney effects in male rats. These studies suggest that the primary site for interference with kidney function is the proximal tubule based on observed increases in urinary glucose. Bones have a high affinity for uranium and there is also some evidence that uranium intake may interfere with normal bone functions (Arzuaga et al., 2015; Kurttio et al., 2005).

The published maximum acceptable concentration of uranium has varied since 1968 (Fig. 2) according to the understanding of the element's toxicity. Uranium was originally treated as an aesthetic issue, with health authorities recommending that drinking water supplies meet the aesthetic objective of 5000 µg/L (Joint Committee on Drinking Water Standards, 1969). A much lower health-based limit of 20 µg/L was first established in 1978 (Federal-Provincial Working Group on Drinking Water, 1979) based on emerging epidemiological evidence, but was then raised to 100 µg/L in 1987 (Federal-Provincial Subcommittee on Drinking Water, 1988), until it was lowered again to 20 µg/L in 1999 (Health Canada, 2001). Health Canada (2017a) is currently reviewing the MAC for total uranium in drinking water and has proposed that the MAC of 20 µg/L be reaffirmed.

Public health messaging has changed in response to the fluctuations in the MAC over the last 40 years, which has created some confusion regarding the health risk of uranium in private wells. When the MAC was raised to 100 µg/L, it is estimated that only a very small percentage (<1%) of private wells in Nova Scotia would have exceeded the guideline, compared to an estimated 4% of private wells exceeding the current MAC of 20 µg/L (Drage and Kennedy, 2013). Risk assessments of uranium in well water (e.g. Grantham, 1986; O'Reilly et al., 2009) have historically been combined with other radiological parameters, such as radium and radon, which may also contribute to public confusion regarding the human health effects of uranium in well water, because the uranium MAC is based on

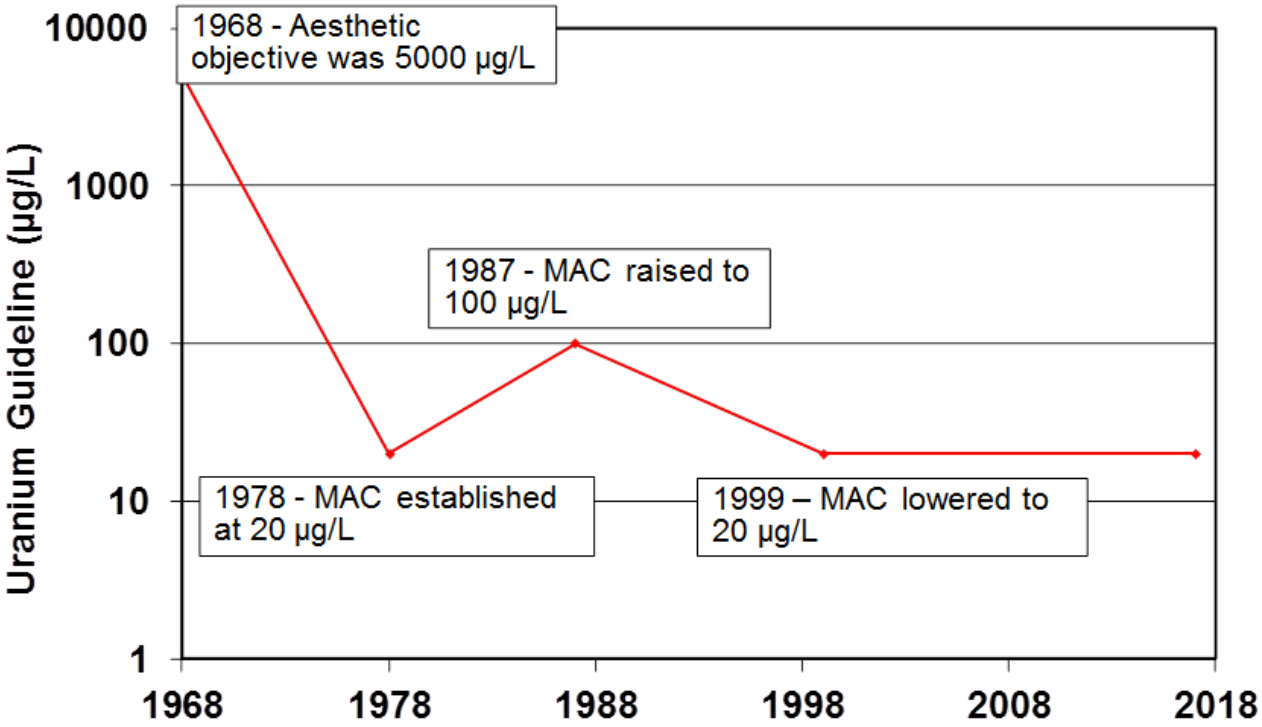

Figure 2. Change in Health Canada maximum acceptable concentration (MAC) for uranium in drinking water over time from 5000 µg/L to 20 µg/L (Joint Committee on Drinking Water Standards, 1969; Federal-Provincial Working Group on Drinking Water, 1979; Federal-Provincial Subcommittee on Drinking Water, 1988; Health Canada, 2001).

chemical toxicity whereas the MACs for radium and other naturally occurring radionuclides are based on radiological toxicity.

The Province of Nova Scotia has had an important role in establishing the toxicity of uranium in drinking water. The community of New Ross (Fig. 1) was known to have elevated levels of uranium in well water, and therefore members of the community were recruited for an initial study on drinking water exposure and urinary concentrations of the metal (Moss et al., 1983; Moss, 1985), and two subsequent studies on the toxicological effects of uranium ingestion from drinking water (Limson Zamora et al., 1998) and gastrointestinal uranium uptake from water and food (Limson Zamora et al., 2002). The latter two studies contributed important evidence towards Health Canada's decision to lower the uranium drinking water MAC from 100 to 20 µg/L in 1999 (Health Canada, 2017a).

Well Surveys of Uranium in Groundwater

Various regional- and local-scale surveys of private wells have been conducted by the Province of Nova Scotia over the past 40 years, contributing to our understanding of the distribution of uranium in well water (e.g. MacFarlane, 1983; Johannessen, 2000; Drage et al., 2005; Samolczyk et al., 2012). Most of the private well surveys conducted by the Province were completed during the Uranium Task Force study and were concentrated in granitoid rocks of the South Mountain Batholith and sedimentary rocks associated with uranium mineralization (e.g. Carboniferous basin) (MacFarlane, 1983). Other relevant private well surveys that included uranium in well water analyses were conducted by the federal government (e.g. Dyck et al., 1976), academic institutions (e.g. Samolczyk et al., 2012), environmental consultants (e.g. CBCL Limited, 2012[1]) and non-governmental organizations (e.g. Clean Annapolis River Project, 2014).

[1]CBCL Limited, 2012. Pugwash Water Supply Investigation – Final Report. CBCL Limited, Halifax. 19 p. (unpublished)

Well Water Surveys for Uranium Exploration

Prior to the formation of the Uranium Task Force in 1979, the focus of well water surveys of uranium was on the detection of uranium-bearing mineral deposits (e.g Dyck et al., 1976). The roll-front deposit model for uranium mineralization in Nova Scotia was first proposed by Brummer (1958). To investigate depositional models of uranium occurrence, the Geological Survey of Canada (GSC) conducted a regional-scale survey in 1975 of over 2000 domestic well-water samples (725 samples in Nova Scotia) collected from 1721 well sites in the Carboniferous to early Permian sedimentary rocks underlying the north shore of Nova Scotia, all of Prince Edward Island, and southeast New Brunswick (Dyck et al., 1976). Dyck et al. (1976) showed that uranium, radon, helium, fluoride, conductivity, and alkalinity in well water exhibited systematic regional trends, indicating regional belts of element enrichment. Following the GSC work, various smaller scale surveys were conducted between 1976 and 1981 in Nova Scotia by mineral exploration companies (e.g. Guardia and MacNabb, 1977) as part of an intensive effort to identify potential uranium deposits. These types of surveys ceased in 1981 when the moratorium on uranium exploration and mining was imposed.

Uranium Task Force

From 1980 to 1982 the Uranium Task Force investigation tested over 700 private wells across four counties, all 72 of the province's municipal water supplies, and the water systems of eight Canadian Armed Forces bases (Grantham, 1986). The communities surveyed are shown in Figure 1, and selected data from these surveys were compiled from Nova Scotia Environment (NSE) files and are presented in Table 1. The dataset compiled for the present study is more comprehensive and has better geographic resolution than the dataset summarized in the Uranium Task Force Study report (MacFarlane, 1983), and so some minor differences between the summary tables will be noted. Approximately 24% of the wells surveyed exceed the Health Canada MAC of 20 µg/L for uranium in drinking water; however, the exceedance rate increases to 35% for drilled wells (Table 1). Less than 1% of the dug wells surveyed exceed the MAC, whereas in the communities of Brookside, Fraxville, New Ross, Wentworth Centre, and Williamswood, more than 40% of the drilled wells in these communities exceed the MAC for uranium in drinking water. The reported exceedance rates are relatively high because the surveys targeted areas where uranium mineralization was interpreted to be present and uranium in well water was a concern.

Special Waters Advisory Group

A limited province-wide testing program of uranium was conducted in 2002 when the radionuclide lead-210 was identified in well water at a public school in the community of Hubley, Halifax County (Fig. 1) (Drage et al., 2005). Lead-210 is a daughter product of uranium and had not previously been tested for in the province. As a result, an intergovernmental Special Well-Water Advisory Group (SWAG) was formed, including representatives of various provincial government departments and Health Canada, and a province-wide water-supply testing program was implemented. The program initially tested the well water supplies of 52 schools for 14 uranium daughter isotopes, including lead-210 (Drage et al., 2005). The program was later expanded to include the collection of drinking water samples from municipal supplies (n=82), and representative point-of-use locations in self-supplied schools (n=178) and other select registered public drinking-water supplies (n=99), and testing the samples for uranium and lead-210 (Drage et al., 2005).

The initial results suggested that lead-210 commonly exceeds the Health Canada MAC of 0.1 Bq/L in Nova Scotia well water, but subsequent testing using a different protocol revealed that the laboratory

Table 1. Summary of exceedance rates (>MAC) compiled from uranium in well water surveys conducted by the Province of Nova Scotia.

Area	County	Count of wells sampled		>20 µg/L uranium		Exceedance rate (>20 µg/L uranium)		Groundwater region	Source
		Dug	Drilled	Dug	Drilled	Dug	Drilled		
Blockhouse*	Lunenburg	0	7	-	0	-	0%	Metamorphic	a
Brookside	Halifax	8	34	0	17	0%	50%	Plutonic	b
Forties	Lunenburg	62	43	1	8	2%	19%	Plutonic	b
Fraxville	Lunenburg	1	4	0	2	0%	50%	Plutonic	
Harrietsfield	Halifax	27	82	0	25	0%	30%	Plutonic	b
Harriston	Lunenburg	4	1	0	0	0%	0%	Plutonic	b
Lake Ramsay	Lunenburg	6	7	0	1	0%	14%		
Leiminster	Hants	16	4	0	1	0%	25%	Plutonic & Metamorphic	b
Louisville area	Colchester	2	19	0	3	0%	16%	Sedimentary	b
New Ross	Lunenburg	77	47	1	23	1%	49%	Plutonic	b
New Russell	Lunenburg	7	0	0	-	0%	-	Plutonic	b
Pugwash	Cumberland	2	36	0	11	0%	31%	Carbonate/ Evaporite & Sedimentary	b
Rhodes Corner*	Lunenburg	1	16	0	0	0%	0%	Metamorphic	a
River John area	Pictou	5	36	0	10	0%	28%	Sedimentary	b
Tatamagouche area	Colchester	0	18	-	5	-	28%	Sedimentary	b
Vaughan	Hants	17	0	0	-	0%	-	Plutonic & Metamorphic	b
Wentworth Centre	Cumberland	2	21	0	9	0%	43%	Sedimentary	b
Williamswood	Halifax	21	172	0	79	0%	46%	Plutonic	b
		Totals		**Per cent of totals**		**Overall per cent exceeding**			
		258	**547**	**0.8%**	**35.5%**	**24.3%**			

* These surveys were not part of the Uranium Task Force work.
a. Arsenic and Uranium Concentrations Well Water Supplies Rhodes Corner and Blockhouse Areas (Nova Scotia Department of Health, 1989)
b. Uranium Task Force (MacFarlane, 1983)

method did not provide a representative indication of lead-210 concentrations because radon gas was rapidly decaying (i.e. radon's half-life is 3.8 days) to lead-210 in the time interval between sample collection and analysis. The sampling protocol was modified to eliminate the effects of radon decay, and additional testing confirmed that lead-210 was not a concern in Nova Scotia well water (Drage at al. 2005). A low overall exceedance rate of the uranium MAC, which had been recently lowered to 20 µg/L, in school well-water supplies was recorded (2%) during the project (Drage et al., 2005), although it should be noted that since point-of-use (i.e. tap where water is consumed) samples were tested, many of the samples may have been collected post-treatment (i.e. after the water had been treated by a water treatment system).

Other Relevant Surveys of Uranium in Private Well Water

As part of an investigation into the use of uranium series isotopes to characterize watershed-scale groundwater flow processes, Johannessen (2000) sampled a total of 20 water wells in 1998 and 1999 for total uranium, ^{234}U, and ^{238}U in the Avon River watershed, near Windsor, Nova Scotia (Fig. 1). Well construction was not consistently recorded during the study; however, interpretation of the dataset suggests that 16 dug wells and 4 drilled wells were sampled. One of the four drilled wells had uranium levels exceeding the Health Canada MAC for uranium (41 µg/L) while the rest of the samples had well water concentrations of uranium below 3 µg/L.

In 2006, a study was carried out by Acadia University in the community of Grand Pré, Annapolis County (Fig. 1), to investigate the influence of redox conditions and regional geology on the mobility of uranium in groundwater (Samolczyk et al. 2012). Seventeen private drilled water-wells in fractured shale and sandstone aquifers were sampled, and nine of the wells were selected for detailed chemical analyses to estimate in-situ redox conditions. Five of the 17 samples (29%) exceeded the MAC for uranium and the results showed that redox conditions strongly influenced levels of uranium in private water wells.

The Clean Annapolis River Project (2014) surveyed rural private wells in the Annapolis Valley and found that approximately 4% of the drilled wells tested (n=124), most of which intercepted fractured shale and sandstone aquifers of the Wolfville and Blomidon formations, exceeded the Health Canada MAC for uranium in drinking water. It is likely, however, that point-of-use water samples were collected during the survey, and therefore some samples may have been collected post-treatment.

Various community-scale surveys have also been conducted in recent years by the Province (e.g. Blockhouse and Rhodes Corner, Table 1) and by municipalities to assess private well-water supplies. Of particular interest is a survey conducted in the Village of Pugwash (CBCL Limited, 2012), Cumberland County (Fig. 1), where uranium was one of the water quality issues that eventually led to the development of a municipal groundwater supply system in 2017.

Sources, Management, and Limitations of Uranium in Well Water Data

During the work of the Uranium Task Force, the Province used a centralized inventory (a ledger book) for tracking uranium results. Some of these data were then entered into a digital database called NAQNSWELL by NSE around the mid-1980s. For the well water surveys conducted in Lunenburg, Pictou, Colchester, and Cumberland counties, property-level location information was not available in NSE files.

The well water chemistry data collected by uranium exploration companies in the 1970s is available from the Nova Scotia Department of Energy and Mines in mineral assessment reports, and was digitized by Dalhousie University in 1999 (O'Beirne-Ryan, 2006) as part of a preliminary risk assessment of uranium and radionuclides in well water. In total the digitized uranium exploration dataset comprises 2041 well and spring water samples, although in most cases the well type and sample type (treated vs. untreated) was not recorded for each sample. O'Beirne-Ryan (2006) also compiled the Uranium Task Force results, although as noted above, property-level location information was not available for parts of the survey.

The GSC dataset, which included water chemistry data for 725 water samples collected from springs and water wells from northern mainland Nova Scotia in 1975, was released in a digital format (Dyck, 1980).

The sampling protocol did not record well type and sample type, although one of four categories of well depth was assigned to each sample, and given the objectives of the sampling program, it is assumed that the GSC collected only untreated water samples.

Well water samples (n=276) were collected from schools and a limited number of registered water supplies between 2002 and 2003 for uranium and other radionuclides as part of the Special Well-Water Advisory Group provincial survey work (Drage et al., 2005). These data were compiled in a digital format during the SWAG work and have been previously published as part of the Nova Scotia Groundwater Chemistry Database (Nova Scotia Department of Natural Resources, 2018). Point-of-use water samples were collected, and so in many cases the sample type (treated vs. untreated) is not known.

The Nova Scotia Groundwater Chemistry Database consists mostly of non-domestic well-water sample results that are assumed to reflect ambient groundwater chemistry (i.e. treated well water samples and data from contaminant investigations are excluded) (Nova Scotia Department of Natural Resources, 2018). This database was compiled from various federal, provincial (e.g. SWAG survey), and municipal groundwater chemistry data sources, including water-quality-monitoring data from government buildings with well water supplies, and NSE groundwater chemistry data from registered public drinking-water supplies, pumping tests, municipal groundwater systems, and provincial observation wells. Where adequate information regarding the well type, well location, and water type (raw vs. untreated) was available, the Uranium Task Force, mineral exploration, and GSC datasets were assigned co-ordinates (if required) and added to the provincial groundwater chemistry database to expand the coverage of uranium in well water samples across the province.

The Nova Scotia Department of the Environment requested available uranium data from the Queen Elizabeth II hospital (QEII) Environmental Services laboratory in Halifax, which performs many of the residential well-water analyses in the province, for analysis in a *State of the Nova Scotia Environment 1998* report (Nova Scotia Department of the Environment, 1998). An update to this dataset was provided in 2000, and the available data contains more than 4000 records of uranium in water analyses completed between 1991 and 2000, although owner and site location data were removed from the dataset by the QEII to protect the water-supply owner's privacy. The dataset mostly contains data for communities in Halifax County because the QEII Environmental Services laboratory is located in Halifax and development activity, including well construction and household water sampling, is concentrated in suburban areas of the County. In addition to poor location information there are a significant number of duplicate, treated water, and surface water samples, and wells sampled multiple times, that cannot be filtered from the dataset.

Despite the data quality issues noted above, the QEII dataset contains a large number of samples, and community-level exceedance rates (i.e. compared to Health Canada MAC of 20 µg/L) can provide useful information regarding the spatial distribution of uranium in well water. Uranium concentrations in communities with some of the highest rates of exceedances based on the QEII dataset are presented in Figure 3 and a statistical summary of the data is provided in Appendix A. The compiled results show that uranium can range over three orders of magnitude (<5 µg/L to 3500 µg/L) in Nova Scotia groundwater under natural conditions, with uranium levels exceeding 200 µg/L in the communities of Glen Margaret, Hacketts Cove, Hammonds Plains, Hubbards, Seabright, and Whites Lake in Halifax County, and New Ross in Lunenburg County. The highest Kaplan-Meier mean uranium levels for communities with at least 15 well water samples include Hacketts Cove, Glen Margaret, Harrietsfield, Whites Lake and New Ross. The overall percentage of samples exceeding the uranium MAC in the communities listed in Appendix A is 8%, with exceedance rates surpassing 40% in the communities of New Ross and Harrietsfield (Fig. 3 and Appendix A).

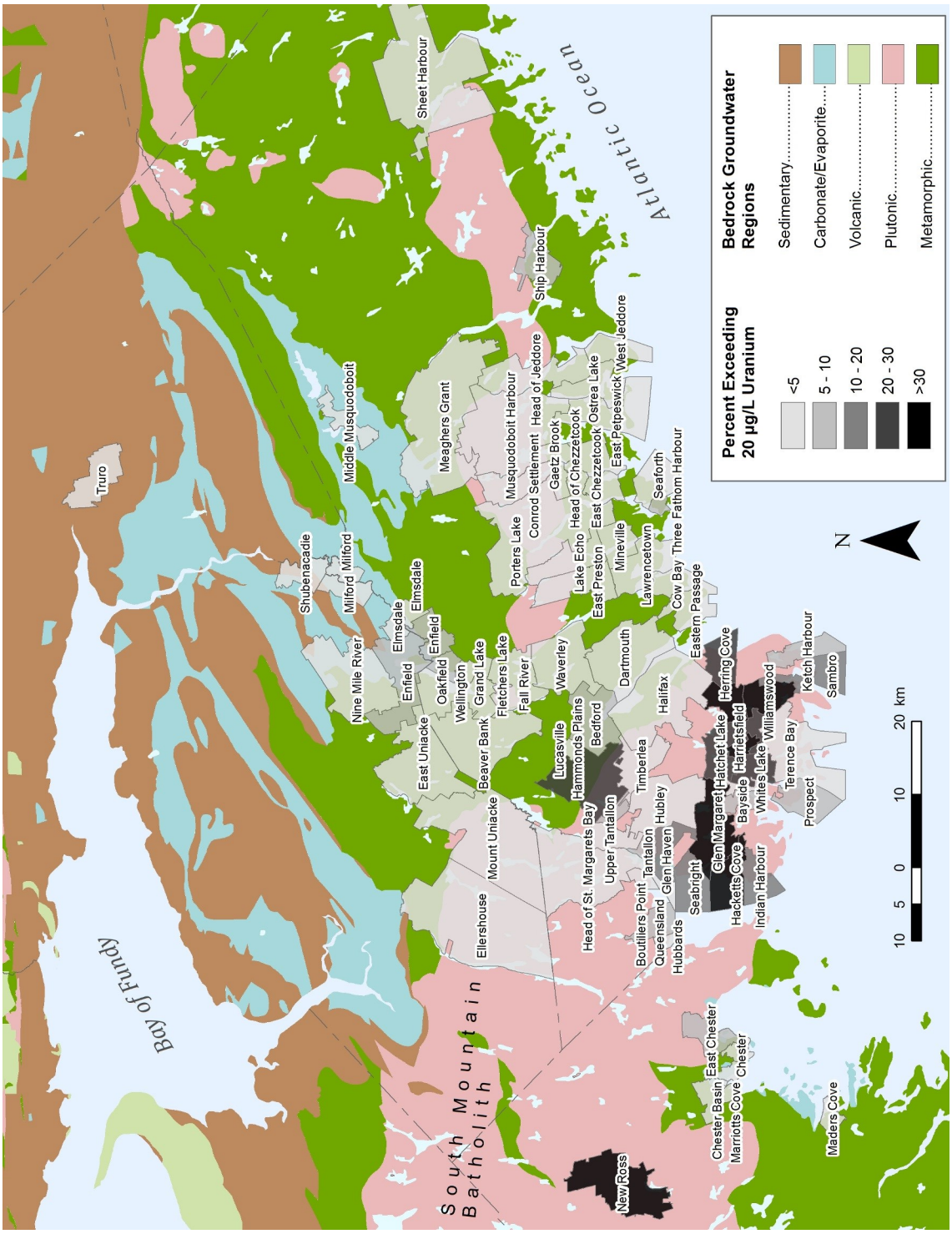

Figure 3. Percentage of samples exceeding 20 µg/L uranium in well water for various communities reported in the Queen Elizabeth II hospital (QEII) Environmental Services laboratory dataset (minimum 10 samples) compared to the province's five major bedrock groundwater regions.

Distribution of Uranium in Nova Scotia's Bedrock Groundwater Regions

The Groundwater Regions Map of Nova Scotia (Kennedy and Drage, 2008) broadly classifies the groundwater regions of Nova Scotia into five major categories based on the dominant rock type of the

various bedrock units shown on the provincial bedrock geology compilation map (Keppie, 2000). The distribution of uranium 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, 2011a).

An updated version of the map showing the distribution of uranium in Nova Scotia's five bedrock groundwater regions was developed using data from the Nova Scotia Groundwater Chemistry Database (Fig. 4; Nova Scotia Department of Natural Resources, 2018). Uranium levels in well water exceeding the Health Canada MAC are present in all of Nova Scotia's groundwater regions, except for the volcanic groundwater region. Uranium concentrations in bedrock aquifers range from less than 1 µg/L to 700 µg/L and approximately 10% of raw water samples (n=2277) exceed the uranium MAC of 20 µg/L. In comparison only 0.7% of raw water samples exceed the uranium MAC in the province's surficial aquifers (n=420).

The exceedance rate for bedrock aquifers is higher than previously reported in Kennedy and Finlayson-Bourque (2011a) because the distribution of the data is spatially heterogenous and the updated dataset presented here includes data recently compiled from intensive well-water uranium surveys in known areas of concern. Previous estimates of the percentage of private wells that may exceed the uranium MAC, accounting for the distribution of private well users relative to the province's major aquifer types, have reported that about 4% or 7000 households may exceed safe limits for uranium in drinking water (Drage and Kennedy, 2013). Most of these exceedances are located in the plutonic groundwater region, especially in granites of the South Mountain Batholith and northern Cape Breton Island, and the sedimentary groundwater region, especially along the Northumberland Strait (Fig. 4). The spatial association of uranium in well water with the granitoid rocks of the South Mountain Batholith and the sandstone and shale units of the Upper Carboniferous formations, especially down-gradient or in contact with plutonic rocks, is well established from previous studies of the distribution of uranium in well water in Nova Scotia (e.g. MacFarlane, 1983; Drage et al., 2005; O'Reilly et al., 2009).

In comparison, various regional surveys of uranium in well water in the northeastern USA (e.g. Ayotte et al., 2007; Colman, 2011; Yang et al., 2014; Flanagan et al., 2014; Flanagan and Brown, 2017) have found that around 3 to 5% of wells have uranium concentrations exceeding the United States Environmental Protection Agency uranium Maximum Contaminant Level of 30 µg/L. These surveys tended to focus on wells located in metamorphic and plutonic rock aquifers because elevated levels of uranium in well water in the northeastern USA have been associated with granite plutons and metasedimentary rocks near plutons (e.g. Yang et al, 2014).

Origin and Controls of Uranium in Well Water

Although there is broad correspondence between groundwater concentrations of uranium and bedrock composition at regional scales, significant heterogeneity has been consistently observed at local scales. Generally, uranium concentrations in well water are heterogeneous and the result of complex hydrogeochemical processes. Both a mineralogical source of uranium and favourable geochemical conditions for uranium to be mobile in the groundwater system are needed for uranium to be present in well water. The distribution of uranium in wells intercepting fractured bedrock aquifers is therefore controlled by local variations in uranium mineralization and rock composition, fracture flow, groundwater pH, and redox potential. The latter two are affected by regional groundwater flow and the geochemical evolution of the groundwater. Well water concentrations of uranium commonly exceed the Health Canada MAC for uranium in drinking water in Nova Scotia because uranium is relatively 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 (MacFarlane, 1983).

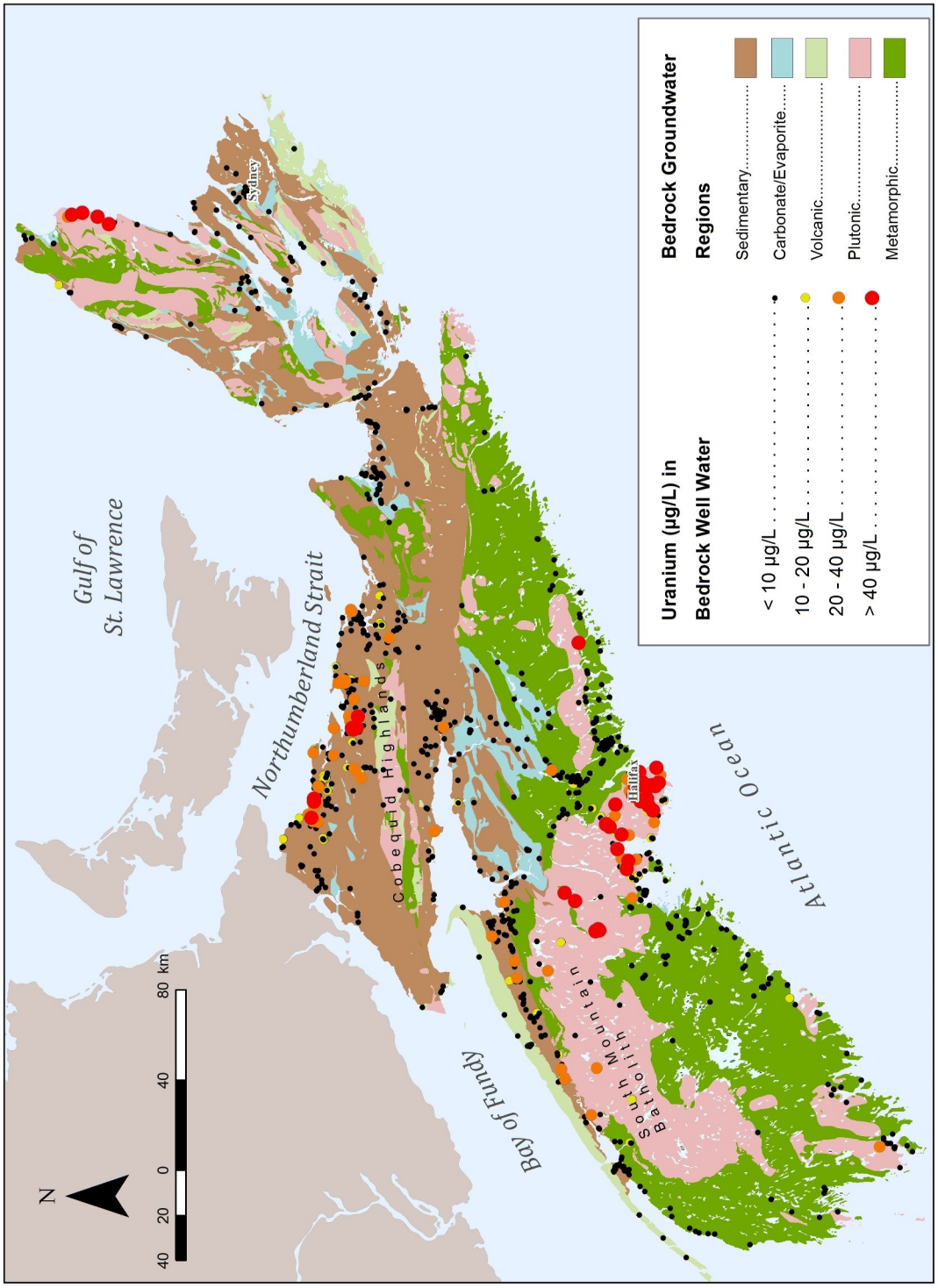

Figure 4. Distribution map of uranium concentrations in bedrock well water from the Nova Scotia Groundwater Chemistry database (n=2277) (Nova Scotia Department of Natural Resources, 2018) compared to Nova Scotia's bedrock groundwater regions (Kennedy and Drage, 2008). Where multiple sample results were available for a given well, the most recent sample concentration is shown.

Geogenic Sources of Uranium

Uranium typically occurs in the upper 100 metres of the earth's crust and has an average continental crustal abundance of 2.7 ppm (Rudnick and Gao, 2003). Uranium is a multivalent heavy metal consisting of a mixture of three radionuclides (^{234}U, ^{235}U and ^{238}U) and is generally found in the environment in either its tetravalent (U(IV) or U^{4+}) or hexavalent (U(VI) or U^{6+}) form. In most uranium-

bearing minerals, uranium occurs as U(IV). The highest concentrations of uranium tend to occur in granites, phosphates, and organic-rich sedimentary rocks (Yang et al., 2014). The most abundant (and most commonly mined) uranium-bearing mineral in Canada is uraninite (UO_2) and its oxidized forms (e.g. U_3O_8). Other common minerals associated with uranium include coffinite and uranium-phosphate minerals, such as torbernite and autunite.

The geogenic occurrence of uranium in Nova Scotia has been known since the late 1950s, when Brummer (1958) noted the presence of uranium mineralization in northern mainland Nova Scotia. The two principal models of uranium concentration in rocks in Nova Scotia include (1) enrichment during igneous differentiation and hydrothermal activity, typically occurring as magmatic and vein type deposits in granitoid rocks, and (2) enrichment during the conversion of the mobile U(VI) to the immobile U(IV) at a reducing barrier, typically occurring as roll-front type deposits in sedimentary rocks. A review of the available literature indicates that anthropogenic sources of uranium in Nova Scotia have not been investigated, although potential sources include the combustion of coal and the use of phosphate fertilizers because these materials may contain uranium in the host rock (Health Canada, 2017a).

Uranium in Plutonic Rocks

The South Mountain Batholith (Fig. 1), a large peraluminous granitoid body comprising multiple intrusions, is associated with the highest rock-concentrations of uranium detected in the province. Radiometric surveys indicate that greater concentrations of uranium and thorium occur in the South Mountain Batholith compared to granites occurring in the Cobequid Highlands and Cape Breton Island (Carson et al., 2003). The overall background average concentration of uranium in the South Mountain Batholith is 6 ppm, with average concentrations ranging from 3 to 8.7 ppm (n=475) for various types of granite (MacDonald et al., 1992) and mineralized zone concentrations as high as 3180 ppm (Chatterjee et al., 1982). The average uranium concentrations in eight granitoid plutons sampled from Cape Breton Island were reported to range from 1 to 9 ppm (n=61) (Barr et al., 1982), and the average concentration in rocks from the Cobequid highlands was 3.5 ppm (n=2105) based on XRF analyses by MacHattie (unpublished data, 2018).

Magmatic-type deposits of uranium in granitoid rocks of the South Mountain Batholith were interpreted to have formed by fluid migration in late stages of granitoid emplacement (Chatterjee and Muecke, 1982; MacDonald et al., 1992; Ryan and O'Beirne-Ryan, 2009). Magmas that are the last to crystallize contain more incompatible elements that do not easily fit into the crystal structure of rock forming minerals. As the magma progressively crystallizes, the volume of molten magma decreases whereas the concentrations of incompatible elements, such as uranium, increases. Because uranium tends to be more enriched in the later phases of igneous differentiation, its concentration has been related to the granite type, increasing from the more mafic rock types, such as diorite and granodiorite, to the more felsic and silica-rich rock types, such as monzogranites and leucogranites (e.g. Chatterjee and Muecke, 1982). Similarly, Ayotte et al. (2007) associated the highest concentrations of uranium in granitoid rocks in New England with two-mica granites (i.e. peraluminous leucogranites containing biotite and muscovite) similar to those found in Nova Scotia.

The most significant documented uranium prospect in the province is a hydrothermal-vein-type deposit that was discovered in the early 1980s near Millet brook (Fig. 1, Chatterjee et al., 1982). The deposit is associated with northeast-trending shear zones, which are hosted in biotite granodiorite of the Salmontail Lake Pluton in the South Mountain Batholith. Bedrock concentrations of uranium in the mineralized zone were reported to range from 938 to 3180 ppm (n=8) (Chatterjee et al., 1982). The interpreted depositional model at Millet Brook involves upward convective movement of solutions enriched with incompatible elements, such as uranium, into surrounding granites, where these elements precipitate in regions of lower pressure and temperature, such as veins, fractures, and shear zones.

Chatterjee and Muecke (1982) proposed further uranium enrichment in South Mountain Batholith granitoid rocks where glaciation and erosion of the roof of the batholith produced fractures and lineaments that could act as channels for fluid migration and zones of mineralization in or near the most differentiated zones of the batholith. MacFarlane (1983) noted that these mineralized fracture zones could also serve as preferential conduits for uranium-bearing groundwater flow to water wells. Bedrock structures, such as late-stage fractures and fault structures, are therefore also associated with uranium enrichment.

MacFarlane (1983) suggested that where granite aquifers in the South Mountain Batholith were overlain by thick, overlying, low-permeability till, reducing conditions may develop because the aquifer is receiving less recharge of younger, oxygenated water compared to surrounding groundwater. Uranium may accumulate in these zones by a roll-front-type mechanism, where more evolved, oxygenated groundwater containing uranium migrates through the reduced zone, and U(VI) is converted to the insoluble U(IV) and redeposited in aquifer materials underlying the till deposit. Ryan and O'Beirne-Ryan (2009) observed strongly weathered horizons (i.e. saprolites) of the South Mountain Batholith that indicated that uranium may have been eroded from the rock matrix and redeposited by the groundwater flow system, possibly in reducing zones of an aquifer.

Uranium in Sedimentary and Carbonate/Evaporite Rocks

The average whole-rock concentrations of uranium in the Sydney coal field of Cape Breton Island were reported to range from 2.9 to 3.8 ppm (n=48) for various sedimentary rock units and 0.06 to 4.8 ppm (n=59) for coal units (Zodrow and Zentilli, 1979). Low-grade uranium anomalies (10 -100 ppm) have been identified in black shale horizons to the west of the coal field in Bourinot Group Cambrian sedimentary rocks. Average uranium concentrations in sedimentary bedrock on mainland Nova Scotia were reported to be 3 ppm in various sedimentary units in central and western Nova Scotia (MacFarlane, 1983), 2.1 ppm (n=187) in sedimentary units in the Cobequid Highlands based on XRF analyses (MacHattie, unpublished data, 2018), and 2 ppm in carbonate and evaporite (i.e. Windsor Group) rocks (MacFarlane, 1983).

Weathering of plutonic rocks, including granitoid rocks containing uranium mineralization, provides source material for the formation of sedimentary layers in the province's adjacent Carboniferous basins. Uranium enrichment in sedimentary bedrock, however, is generally interpreted to have formed by the roll-front mechanism described earlier (e.g Dyck et al., 1976; Ryan and O'Beirne-Ryan, 2009). Broad belts of uranium enrichment can occur when U(VI), which is soluble and mobile as the uranyl ion and its complexes, is leached from rock within a zone of uranium enrichment, such as the Cobequid Highlands or South Mountain Batholith, and is transported by groundwater under oxidized, alkaline conditions until it is redeposited when it encounters a reductive front (Dyck et al., 1976). Under reducing conditions, U(VI) is converted to U(IV), which is typically insoluble and readily sorbs to aquifer material (Langmuir, 1978; O'Reilly, 1982). Reducing zones in sedimentary bedrock tend to be associated with pyrite mineralization (reductant) (Ryan and O'Beirne-Ryan, 2009) or with organic black shale or clay-rich sedimentary units (Smedley et al., 2006).

Dyck et al. (1976) attributed regional patterns of elevated uranium in well water in the province's sedimentary rocks, along with patterns of radon, helium, fluoride, alkalinity and conductance, to roll-front-type deposition. Uranium anomalies were generally associated with lower Pictou Group rocks, which are the youngest sedimentary strata in Nova Scotia and mostly consist of alternating sequences of red and grey sandstone and siltstone. Although only a weak relationship was found by Dyck et al. (1976) between copper and uranium occurrence in water wells, copper minerals are often found in association with uranium since they also precipitate in reductive zones (Brummer, 1958). Ryan and O'Beirne-Ryan (2009) also ascribed uranium mineralization in Horton Group sandstone to roll-front-type mineralization; however, the source of the uranium in the system was interpreted to include weathered

horizons of basement rocks (i.e. saprolites) beneath the Horton Group. The most significant roll-front uranium deposit in Nova Scotia occurs near the community of Three Mile Plains (Fig. 1), where concentrations as high as 953 ppm were recorded in a mineralized zone of Horton Group sandstones (Nankamba, 2011).

There are fewer examples of uranium mineralization observed in carbonate and evaporite rocks, although uranium mineralization was encountered near the community of Maitland (Fig. 1) at the base of the Windsor Group in Macumber Formation limestones and overlying solution collapse breccia (Charbonneau and Ford, 1978).

Uranium in Metamorphic and Volcanic Rocks

Background average concentrations of uranium in metamorphic rocks, measured by XRF, were reported to be 2.4 ppm (n=445) in southwestern Nova Scotia (Meguma terrane) (White, 2010) and 3.3 ppm (n=149) in the Cobequid Highlands (MacHattie, unpublished data, 2018). The Uranium Task Force noted that most occurrences of elevated uranium in metamorphic rock aquifers were located near the geological contact with granites, where intrusions in metamorphic rock are common (MacFarlane, 1983). Anomalously high concentrations of uranium in well water and surface water at Camperdown (Lunenburg County) and Rawdon Gold Mines (Hants County) (Fig. 1) could not be readily associated with a geogenic source, but were proposed to have originated from granitic intrusions at shallow depth below Halifax Group rocks (MacFarlane, 1983; Page, 1999).

Some studies have suggested that uranium content in metamorphic rocks may be associated with the metamorphic grade (Hall et al., 1987; Ayotte et al., 2007). Because metamorphism releases uranium-bearing fluids from rock, depletion of uranium is common in metamorphic rocks with a high level of metamorphism (O'Reilly, 1982). In the state of Maine, USA, Yang et al. (2014) found elevated concentrations of uranium and radon in metamorphic rocks within a 5 to 10 km distance of granitic intrusions, but did not find evidence of correlation between whole-rock concentrations of uranium and metamorphism at intermediate scales.

Dostal et al. (1983) reported average concentrations of uranium of 2.6 to 5.4 ppm (n=61) in various volcanic rocks across Nova Scotia, while MacHattie (unpublished data, 2018) reported an average concentration of 4.4 ppm (n=954) based on XRF analyses of volcanic rocks in the Cobequid highlands. O'Reilly (1982) attributed anomalous levels of uranium in rhyolitic rocks of the Byers Brook Formation to hydrothermal alteration, whereas Dostal et al. (1983) attributed the distribution of uranium in Paleozoic rhyolites in Nova Scotia to secondary processes, such as low-grade metamorphism, which could lead to either enrichment or depletion of uranium. Elevated levels of uranium in volcanic rocks were also found to be related to enrichment with K_2O (Dostal et al., 1983).

Uranium in Tills and Sediment

Regional till surveys were conducted between 1977 and 1982 over the Meguma Terrane, and between 1982 and 1985 in northern mainland Nova Scotia (Nova Scotia Department of Natural Resources, 2006a). The average uranium concentration in tills (including granite tills) reported by the regional till surveys was 2.7 ppm (n=1587). In comparison, the average uranium concentration in tills over the South Mountain Batholith was 6.5 ppm (n=2071) based on a regional till survey conducted between 1984 and 1989 (Nova Scotia Department of Natural Resources, 2006b). Granite tills are generally associated with higher concentrations of uranium compared to slate or clay tills in the province (Stea and O'Reilly, 1982; Boner et al., 1990).

A regional lake sediment survey conducted in eastern Cape Breton Island (1983-1985) reported an average uranium concentration of 3.8 ppm (n=354) (Nova Scotia Department of Natural Resources, 2006c), and a regional lake sediment survey conducted on mainland Nova Scotia (1977-1978) reported an average uranium concentration of 4.6 ppm (n=3399) (Nova Scotia Department of Natural Resources, 2006d). The average uranium concentration for stream sediment in Nova Scotia was 8.2 ppm (n=11,120) based on a compilation of data from surveys conducted from 1982 to 1983 (Nova Scotia Department of Natural Resources, 2006e) and from 1986 to 1987 (Nova Scotia Department of Natural Resources, 2006f) in northern mainland Nova Scotia and Cape Breton Island. In comparison, Page (1999) summarized uranium exploration data collected by Saarberg Interplan Canada Ltd. in areas underlain by sedimentary and metasedimentary rocks in Hants and Kings counties and found uranium contents ranging from 0 to 32 ppm in stream sediment, with an average concentration of 0.7 ppm (n=1309).

Geochemical Controls of Uranium in Well Water

The fate and transport of uranium in the subsurface is governed by various processes including oxidation/reduction, dissolution/precipitation, and adsorption/desorption. These processes are mediated by various geochemical controls, many of which have been studied in Nova Scotia, including pH, redox, the weathering of host mineralogy, the availability of soluble complexes, and ion exchange (e.g. MacFarlane, 1983; Finlayson-Bourque et al., 2010; Samolcyk et al., 2012; Drage and Kennedy, 2013 Letman et al., 2018). Uranium has variable oxidation states (U^{+4}, U^{+5}, U^{+6}), of which the more soluble, oxidized form U^{+6}, or U(VI), is the most important in terms of uranium in well water (Langmuir, 1978). The U(VI) form of uranium is found in the uranyl ion, UO_2^{2+}, which readily forms stable complexes with bicarbonate and carbonate, and is mobile under the range of typical redox and pH conditions observed in Nova Scotia's aquifers.

pH and Redox Controls on Uranium Mobility

Because the concentration of U(VI) increases with pH when pH levels are greater than 4 (Goodwin, 1980), several studies have highlighted the importance of pH as a control on uranium concentrations in the province's well water (e.g. MacFarlane, 1983; Drage and Kennedy, 2013; Letman et al. 2018). For example, elevated uranium in well water was found to be associated with pH levels between 7.5 to 8.5 in both plutonic and sedimentary aquifers during the Uranium Task Force study (MacFarlane, 1983). MacFarlane (1983) also found a relationship between overburden thickness and uranium concentrations in granite aquifers, which was partially attributed to the more evolved groundwater (i.e. more alkaline and higher pH) occurring under thicker glacial till deposits (e.g. drumlins) compared to surrounding groundwater. In the Harrietsfield-Williamswood survey areas, approximately 70% of the wells with uranium exceeding 20 µg/L had at least 12 m of overburden, despite these wells accounting for only 43% of the wells surveyed in the two communities (with available stratigraphic information). A similar relationship between overburden thickness and uranium concentrations, however, was not observed in the province's sedimentary aquifers (MacFarlane, 1983).

Uranium is present in tills derived from the South Mountain Batholith (e.g. Stea and O'Reilly, 1982; Boner et al, 1990); however, few surface waters or dug wells show elevated levels of uranium. Despite surface water and shallow groundwater being oxygenated, low uranium levels are typically observed due to the low solubility of uranium at lower pH values. All 166 raw surface water samples collected from municipal and institutional water supplies by the Uranium Task Force reported uranium levels below 5 µg/L (MacFarlane, 1983), whereas stream sample concentrations of uranium ranging from 0 to 24 µg/L (average 0.78 µg/L, n=115) were reported by Page (1999) in Hants and Kings counties and concentrations ranging from 0.15 to 0.29 µg/L (n=9) were recorded by Johannessen (2000) in the Avon

River watershed. Elevated uranium levels, however, can occur in dug wells where tills are highly alkaline and the pH favours uranium mobilization. Laboratory leachate experiments by Parsons (2007), which involved the placement of sandstone samples from Horton Group outcrop in rain water, confirmed that surface weathering of mineralized bedrock outcrop can produce aqueous concentrations of uranium above acceptable limits.

During the Uranium Task Force well water surveys, MacFarlane (1983) did not measure redox potential or redox indicator parameters, such as dissolved oxygen, iron and manganese. However, a relationship between the redox environment and uranium levels in well water was proposed to partially explain the association between till thickness and elevated concentrations of uranium. MacFarlane (1983) attributed higher uranium concentrations in granite aquifers overlain by thick till deposits to a greater accumulation, and hence availability, of uranium in these aquifers because the presence of thick, overlying, low-permeability till promotes the development of reducing conditions (e.g. lesser input of younger oxygenated groundwater recharge) and the deposition of uranium minerals. It is inferred that groundwater pumping increases the redox potential (i.e. more oxic) around the well, remobilizing the stored uranium, which is then transported to the water well. The Uranium Task Force findings indicated that short- (e.g. pumping cycles) (Grantham, 1986) and long-term (e.g. seasonal) (Feldman, 1986) variation in uranium levels was negligible if water usage (and hence pumping rates) remained consistent; however, significant increases (~60%) were observed when a well was sampled following a period of inactivity of several months.

Although no studies in Nova Scotia have directly measured redox potential in-situ, several studies have used indicators of the redox environment to show a relationship between uranium and redox potential. For example, in a municipal wellfield located in the community of Bridgetown, Annapolis County (Fig. 1), a relationship between aquifer drawdown and uranium concentrations was observed for the community's new production wells (Finalyson-Bourque et al., 2010). Following the conversion of open borehole test wells to larger diameter, partially screened production wells, uranium concentrations in the wells increased by 80%, and approximately 25 m of additional drawdown was observed in the wells at the required production rates. A series of investigations, including aqueous leachate tests of drill cuttings, depth discrete sampling, discrete fracture sampling, and pumping tests, concluded that uranium may have been remobilized due to the development of more oxidizing conditions in the aquifer. The screened production wells were associated with increased redox potential in the aquifer due to greater turbulence and entrance velocity through wells screens during pumping, and lower well-efficiencies resulting in greater drawdown (dewatering) of the aquifer around the production wells.

Another indicator of redox controls on uranium concentration in well water is the relationship of uranium with iron and manganese. Finalyson-Bourque et al. (2010) showed that low iron and manganese concentrations, indicative of an oxidizing environment, and higher uranium concentrations were measured in the Bridgetown production wells during pumping, whereas high iron and manganese concentrations, indicative of a reducing environment, and lower uranium concentrations were measured in the wells under non-pumping conditions. Pothier (2009) also observed an inverse relationship between uranium and both iron and manganese in the Bridgetown wellfield during depth-discrete sampling of an open borehole test hole. Similarly, inverse relationships were found between dissolved uranium and dissolved iron and manganese by Samolczyk et al. (2012) during an investigation into the influence of redox conditions and regional geology on the mobility of uranium in groundwater in the community of Grand Pré, Kings County (Fig. 1). Elevated concentrations of uranium in Grand Pré well water were associated with oxidizing environments and low groundwater concentrations of iron and manganese. The wells with uranium exceeding 10 µg/L had low to moderate levels of iron (<10 to 32 µg/L), whereas four of the five wells with low concentrations of uranium (<2.5 µg/L) had iron concentrations exceeding 100 µg/L (102 to 605 µg/L).

Other Geochemical Controls on Uranium Mobility

The mobility of uranium is also related to the mineral with which it is associated. For example, uranium in minerals such as biotite is typically present in cleavages and is therefore readily mobilized when the biotite is physically or chemically weathered to clay minerals (Ryan and O'Beirne-Ryan, 2009). Synthetic leach experiments by Blume (2016) provided evidence that mineralogy was a key control in the release of uranium, with South Mountain Batholith granites leaching more than three times the amount of uranium than the Horton Group siltstones, despite the granites having less than half the concentration of uranium in the rock samples. Blume (2016) suggested that the uranium present in the siltstone rock samples are associated with organic-rich layers that hold the uranium more strongly compared to the uranium contained in the granite rock samples. In addition to the host mineralogy, the adsorption of uranium to mineral surfaces, such as iron and manganese oxides and clay minerals, can have an important role in governing uranium mobility in the groundwater-flow system (Giblin et al., 1981, Fox et al., 2006).

Another key control on the mobility and distribution of uranium is the availability of dissolved or suspended anions such as carbonate, phosphate, sulphate, silicate, and fluoride to form complexes with uranium. Hexavalent uranium readily forms complexes with these anions that are mobile in groundwater-flow systems. Based on historical well chemistry data, MacFarlane (1983) generally found elevated levels of fluoride, silicate, phosphate, and carbonate in well water samples compared to provincial averages in areas with elevated uranium in well water. A comparison of the spatial patterns of elevated fluoride (Kennedy and Finlayson-Bourque, 2011b) and uranium in Nova Scotia bedrock water wells shows good correspondence between the two elements. In sedimentary rock, MacFarlane (1983) also found a positive association between well water concentrations of uranium and concentrations of total dissolved solids (TDS) and hardness. Generally, the presence of uranium in well water tends to be associated with the presence of other incompatible elements (e.g. S, As, Mo, F, and Cs) that become enriched during late stage magmatic cooling in granites (MacFarlane, 1983, Yang et al. 2014) or with elements that are also soluble and mobile under oxidizing conditions, such as vanadium (Pothier, 2009) and arsenic (MacFarlane, 1983).

Although radon is a daughter product of uranium, clear evidence of a strong correlation between uranium and radon in Nova Scotia well water has not been shown. A direct correlation between uranium and radon was observed by MacFarlane (1983) in the River John area and by Kronfeld et al. (2004) in the Avon Valley and Windsor areas (Page, 1999) of Nova Scotia (mostly Carboniferous basin rocks); however, in most other areas of the province, including granitoid rocks that are associated with higher concentrations of uranium, only a weak correlation has been observed (Dyck et al., 1976; MacFarlane, 1983). The decoupling of uranium and radon may be due to the short half-life of radon and to differences in their mobility in the groundwater-flow system in response to local hydrogeochemical conditions (e.g. Yang et al., 2014).

Anthropogenic Influences on Uranium Mobility

Drage and Kennedy (2013) investigated the association between calcium and uranium using data from the Nova Scotia Groundwater Chemistry Database and aqueous speciation modelling. The results showed strong positive correlations between uranium, calcium, and chloride. Speciation modelling indicated that calcium can be an important influence on the mobility of uranium in groundwater in Nova Scotia due to the formation of highly mobile zero-valent calcium-uranyl-carbonate complexes, which can inhibit adsorption to iron oxyhydroxides (Fox et al., 2006). The results also indicate that adding salt to groundwater (i.e. by road salt or seawater intrusion) can have a similar effect on uranium mobilization, either by directly adding small amounts of calcium or by releasing calcium by ion

exchange. Letman et al. (2018) developed a new uranium leach procedure using core samples from six sites in Nova Scotia to provide a more conservative tool for identifying high-risk areas of uranium mobilization. The procedure was designed to estimate the maximum concentrations of uranium that may be mobilized from rock under alkaline pH and oxidizing conditions, along with higher than background concentrations of calcium, sodium, chloride, carbonate/bicarbonate, and sulphate.

Because anthropogenic land-use activities can introduce complexing agents, uranium mobility can be sensitive to land use. For example, Letman et al. (2018) reported that seven water wells downgradient of a construction and demolition waste disposal facility in the community of Harrietsfield showed anomalously high uranium concentrations (>1200 µg/L). The elevated uranium may have been related to the storage of gyprock at the facility, which contributed calcium, sulphate, and alkalinity to the groundwater, thus mobilizing geogenic uranium. Other examples of anthropogenic activities that may influence uranium mobility in Nova Scotia groundwater include agricultural land-use, road de-icing, and seawater intrusion induced by groundwater extraction. In the San Joaquin Valley (California, USA), Jurgens et al. (2010) found that increasing bicarbonate concentrations in groundwater related to agricultural land-use was associated with increasing concentrations of uranium in groundwater. In northern Germany (van Berk and Fu, 2016) and in two major aquifers in the USA (Nolan and Weber, 2015), agricultural inputs of nitrates in fertilizers caused a redox shift toward oxidizing environments, mobilizing uranium in groundwater. Bäckström et al. (2004) found that the use of de-icing agents such as sodium chloride (NaCl) and calcium magnesium acetate (CMA) in Sweden increased the concentrations of heavy-metal concentrations in soil water. Other studies have linked uranium mobilization at regional scales to groundwater extraction and consequent changes in groundwater-flow patterns and oxidation state (e.g. Ayotte et al., 2011; Coyte et al., 2018).

Risk Management

Over the last 40 years, various activities have been conducted by the Province of Nova Scotia to manage the risk of uranium in well water. These activities include well water surveys and risk mapping, participation in the remedial design of a wellfield impacted by naturally occurring uranium, assessment of small-scale uranium treatment technology, an assessment of the potential for uranium in well water during subdivision planning, and outreach and education.

Risk Mapping

Geohazard risk maps are important tools for land-use planning and for the communication of the relative risk of various geohazards to homeowners. Uranium in well water risk maps are used to communicate to well owners the relative risk of unacceptable levels of uranium, and to inform land development, groundwater supply development, and risk communication and mitigation strategies. It should be noted that residential development activity in Nova Scotia tends to be focused in suburban Halifax, which includes areas of the province known to have an elevated risk of uranium in well water, such as areas underlain by the South Mountain Batholith.

Although the Uranium Task Force well water survey captured only a small part of the province, the task force produced the first generation of uranium risk map, which outlined two major types of bedrock where well water concentrations of uranium exceeding 10 µg/L had been detected (igneous and Carboniferous basin rocks). The map was titled *Uranium Occurrence in Groundwater in Nova Scotia* (Grantham, 1986). The Nova Scotia Department of Health (no date) subsequently republished a simplified version of this map titled *Naturally Occurring Uranium in Groundwater in Nova Scotia,* although the ‘occurrence’ zones were not changed from the original task force version (Fig. 5).

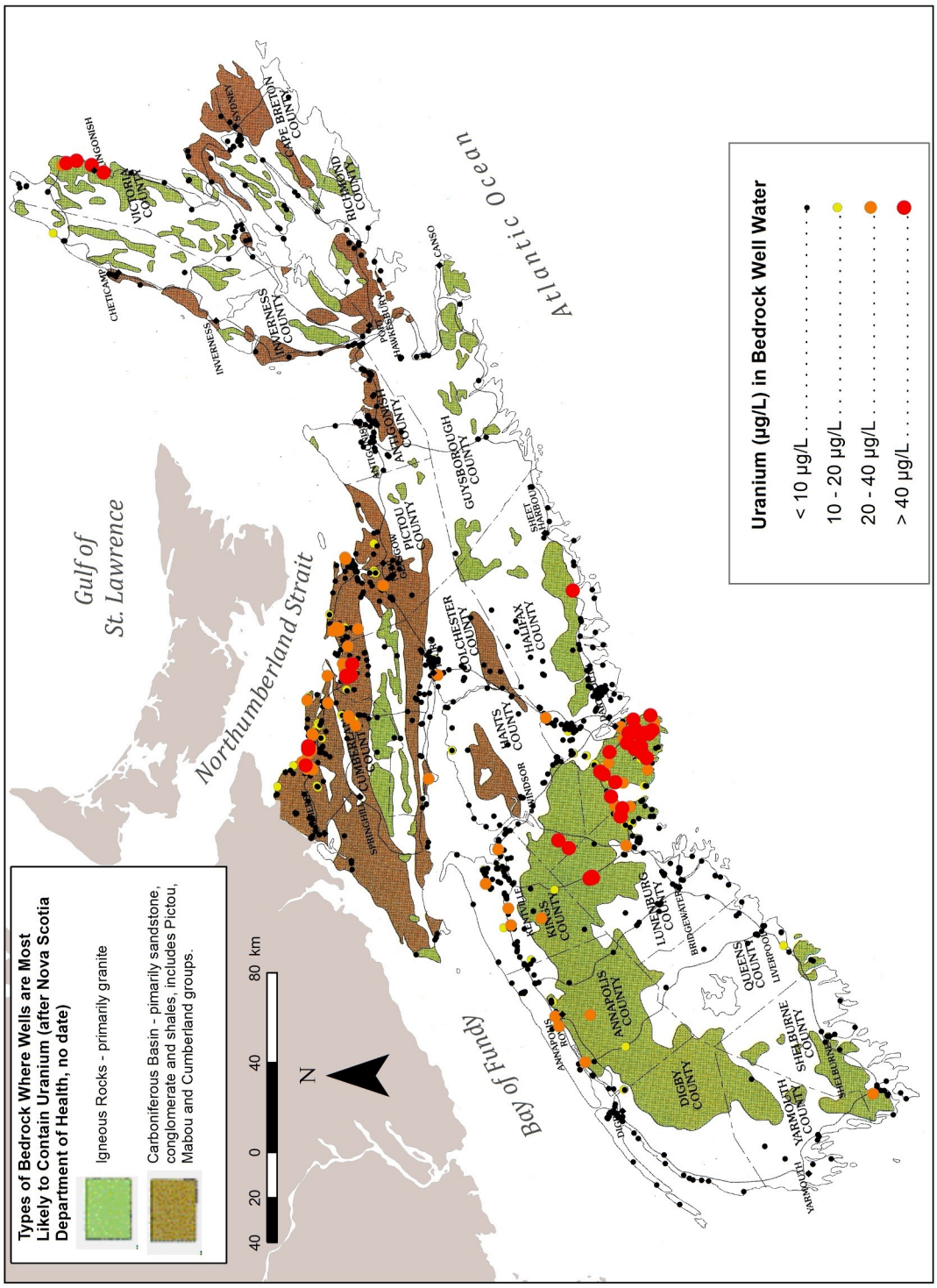

Figure 5. First-generation uranium in well water risk map for the province developed by the Nova Scotia Department of Health (no date). The distribution of uranium concentrations in well water from the Nova Scotia Groundwater Chemistry database (Nova Scotia Department of Natural Resources, 2018) is shown on the map for comparison to the risk zones.

A second-generation uranium in well water risk map, titled *Potential for Uranium and Related Radionuclides in Groundwater in Nova Scotia*, was published in 2009 (O'Reilly et al., 2009) and includes the risk of related radionuclides occurring in well water (Fig. 6). The risk map divides the province into two risk levels. Level 1 areas are more likely to have groundwater containing uranium and related radionuclides above the Health Canada MAC whereas Level 2 areas are less likely to have groundwater containing uranium and radionuclides exceeding the MAC.

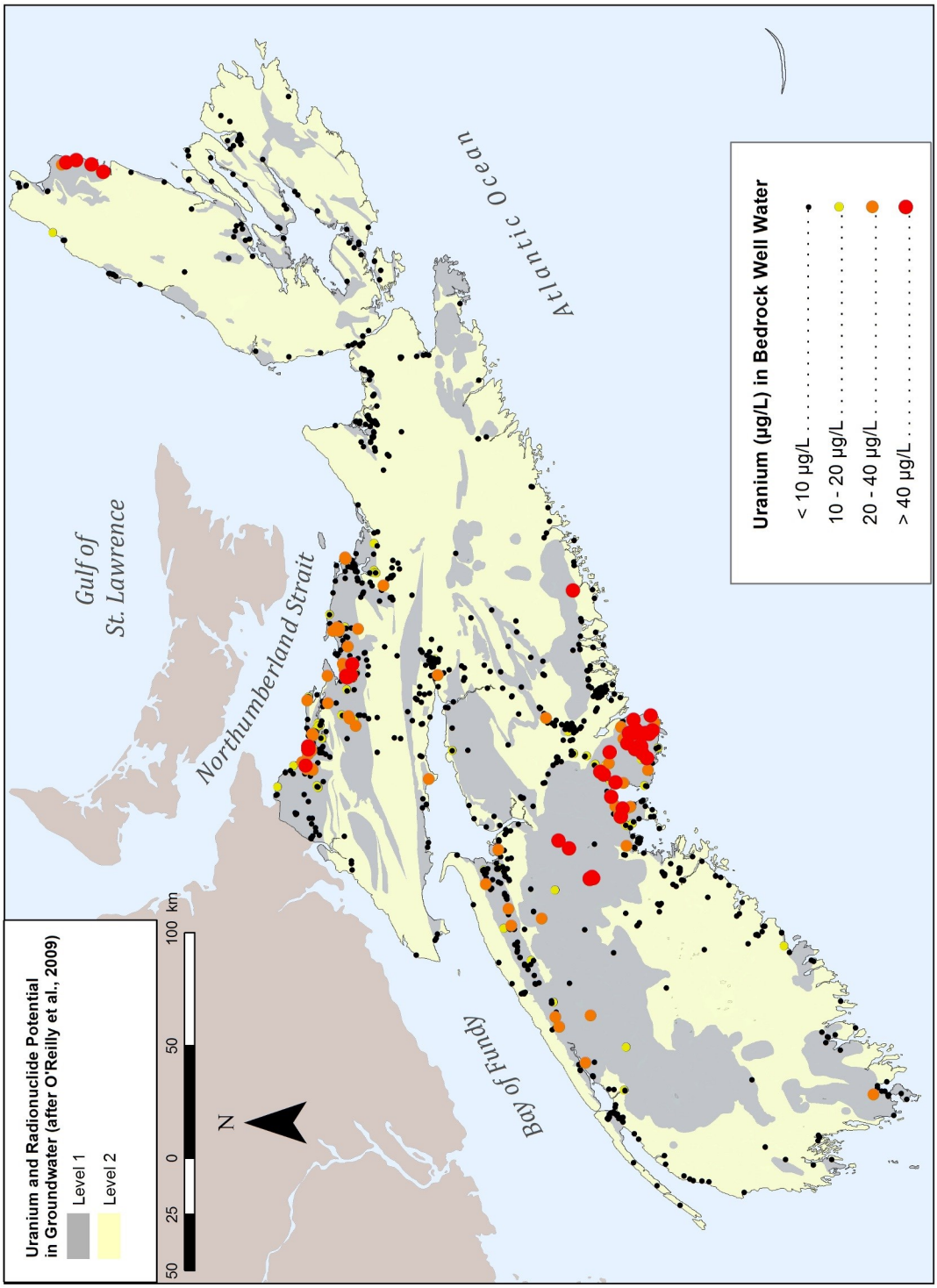

Figure 6. Second-generation uranium in well water risk map for the province (O'Reilly et al., 2009). The distribution of uranium concentrations in well water from the Nova Scotia Groundwater Chemistry database (Nova Scotia Department of Natural Resources, 2018) is shown on the map for comparison to the risk zones.

The two main geological terrains that constitute the Level 1 zone include (1) granitoid rocks in the southern and eastern mainland, and (2) Carboniferous and Jurassic sedimentary rocks in which both chemically reduced and oxidized rock units are common. These sedimentary units underlie much of the Cumberland Basin in northern Nova Scotia, the Annapolis Valley, and areas between the Town of Windsor and the community of Maitland in Hants County. The Level 2 zone comprises areas underlain by geological units in which uranium concentrations may exceed the Health Canada MAC, but are rare and tend to be more isolated compared to the Level 1 zone.

O'Reilly et al. (2009) reduced the extent of high-risk areas for elevated uranium in well water compared to the original uranium occurrence map in various sedimentary basins and granitoid rocks that are not part of the South Mountain Batholith but expanded the extent of the high-risk zone to include sandstones and shales in Annapolis, Kings, and Hants counties. The second-generation risk map (O'Reilly et al., 2008) was developed based on a more comprehensive dataset (n=3717) derived from various well water surveys, including provincial surveys (e.g. SWAG and Uranium Task Force), uranium exploration projects (e.g. Guardia and MacNabb, 1977), and federal government surveys (e.g. Dyck, 1980), in addition to compilation mapping of airborne gamma surveys previously conducted across Nova Scotia (Carson et al., 2003).

Public Water Supplies with Uranium Issues

There are presently no municipal water systems in Nova Scotia that are treating for uranium, although a wellfield-blending strategy is being used in the community of Bridgetown to lower the overall uranium concentration in the distribution system to acceptable levels (CBCL Limited, 2018[2]). As discussed previously, elevated levels of uranium were detected in production wells in the Bridgetown wellfield (Finlayson-Bourque et al., 2010). The initial mitigation strategy to lower uranium concentrations in the wellfield involved the modification of production wells (e.g. conversion of the partially screened wells to fully screened wells and sealing off deeper uranium bearing fractures) and changes to the wellfield configuration (e.g. adding a third production well and reducing the pumping rate from the well with the highest uranium levels) to minimize aquifer drawdown and the development of oxic conditions in the aquifer. More recently, a new production well has been constructed with lower concentrations of uranium and will be used to reduce the overall concentration of uranium in the distribution system (CBCL Limited, 2018).

Smaller public (non-municipal) water supplies are required to regularly test their water in accordance with the *Guidelines for Monitoring Public Drinking Water Supplies* (Nova Scotia Environment and Labour, 2005), and if the Health Canada MAC of 20 µg/L for uranium is exceeded, corrective action is required, which would likely involve water treatment, well modification, or the development of an alternate potable water source. Several smaller public water systems have had issues with uranium in well water, including a trailer park in Harrietsfield, schools in New Ross and Harrietsfield, and a subdivision in Prospect Bay. Mitigation strategies that have been employed at these public water supplies include ion exchange and point-of-use and point-of-entry reverse osmosis systems.

Private Water Supplies with Uranium Issues

In response to concerns regarding the viability of household treatment of uranium, bottled water was provided to residents in Harrietsfield and an investigation into available household treatment technologies was conducted under the Uranium Task Force study (Grantham, 1986). The study showed that point-of-use distillation and reverse osmosis systems were effective and practical. Point-of-use reverse osmosis systems are now commonly employed in Nova Scotia for the removal of uranium from drinking water in small residential systems due to their relative affordability, effectiveness in treating multiple contaminants, and low maintenance requirements (United States Environmental Protection Agency, 2000). At the residential level, Health Canada (2017a) advises that treatment devices are not expected to contain sufficient radioactivity to warrant special precautions by homeowners with respect to the disposal of reject water and spent reverse osmosis cartridges.

Approximately 42% of Nova Scotians use private water wells for their domestic water supply (Kennedy and Polegato, 2017). These 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

[2]CBCL Limited, 2018. Bridgetown Water Supply Water Withdrawal Application. CBCL Limited, Halifax, 22 p. (unpublished)

recommendation for routine testing of private wells by homeowners has been promoted by the Nova Scotia government through various outreach and education initiatives, including risk maps and factsheets, since uranium was first detected in well water in the late 1970s. Despite these efforts, Chappells et al. (2014) discovered poor compliance with the recommended testing frequency for chemical water quality in a study that surveyed risk behaviours amongst private well owners in five separate areas of Nova Scotia that were considered to represent a demographic cross-section of the province. Analysis of well testing and mitigation behaviours in New Hampshire, USA, did not show a correlation between elevated uranium in water wells and the likelihood of a well owner implementing a water treatment mitigation strategy (Flanagan et al., 2014).

Uranium Hazard Assessment in New Subdivisions

Starting in the 1970s, applications for subdivisions using private wells for water supply were reviewed by NSE staff, which provided comment on proposed water supplies including the potential for water quality problems. If uranium exceeded acceptable limits, mitigation was recommended. The recommendations were not enforceable and the reviews were discontinued in the late 1980s. In 2006, Halifax Regional Municipality implemented guidelines for groundwater assessment for new subdivisions using 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 uranium, are detected.

Summary and Perspectives

Since the issue of uranium in well water emerged in Nova Scotia in the late 1970s, extensive research and risk management activities relating to uranium hazards in well water supplies have been conducted in the province. Uranium in well water data, combined with analyses of the uranium content of various types of geological media, have helped to identify patterns of elevated well water concentrations of uranium across Nova Scotia. Research has shown that drilled wells are associated with a significantly higher probability of elevated levels of uranium compared to dug wells and that the source of uranium in water wells in Nova Scotia is geogenic.

The province-wide exceedance rate for drilled wells in bedrock aquifers is approximately 5%, although much higher exceedance rates (>40%) have been reported in selected communities, especially those underlain by the South Mountain Batholith. Water wells located in granitoid rocks and upper Carboniferous and Triassic sandstone and shale aquifers are more likely to contain elevated uranium levels than other aquifer types.

The fate and transport of uranium in the subsurface is governed by various processes including oxidation/reduction, dissolution/precipitation, and adsorption/desorption. Anthropogenic land uses can introduce changes to groundwater-flow patterns and groundwater chemistry which can mobilize uranium stored in the aquifer matrix. The complexity of Nova Scotia's fractured bedrock aquifers with respect to fracture flow and host mineralogy, and hence the evolution of groundwater along its flowpath, contributes to the spatial heterogeneity of uranium in groundwater at various scales. Additional research is warranted to advance our understanding of these relationships.

Uranium, along with other contaminants of concern, such as arsenic, can usually be removed from well water using conventional point-of-use reverse osmosis treatment systems. It is the responsibility of private well owners to mitigate uranium hazards in their well water, and therefore effective public awareness programs are critical for managing uranium exposure risks and associated adverse health

effects. The Nova Scotia Department of Energy and Mines plans to develop a refined risk map and web mapping application to communicate the risk of uranium in drinking water to private well users.

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Appendix A. Compiled Exceedance Rates for Uranium in Drinking Water (>20 µg/L) for Selected Communities Based on Data from the Queen Elizabeth II Hospital Environmental Services Laboratory Dataset (1991-2000).

		All Data						Detects Only			
Community	County	Number of observations	Number of detects	Per cent of detects	Per cent exceedance[1]	K-M[2] Mean (µg/L)	95th percentile (µg/L)	Minimum (µg/L)	Median (µg/L)	Mean (µg/L)	Maximum (µg/L)
Bayside	Halifax	10	3	30%	10.0%	9.3	25.5	8	20	19.3	30
Beaver Bank	Halifax	102	15	15%	0.0%	5.2	6.0	5	6	6.3	10
Bedford	Halifax	16	2	13%	6.3%	9.9	26.0	8	44	44.0	80
Boutiliers Point	Halifax	30	6	20%	3.3%	6.0	6.6	5	6	9.8	30
Brookside	Halifax	35	22	63%	34.3%	26.9	92.0	5	30	39.9	130
Chester	Lunenburg	41	7	17%	0.0%	5.9	7.0	5	7	10.1	20
Chester Basin	Lunenburg	18	1	6%	0.0%	5.0	5.0	5	5	5.0	5
Chezzetcook	Halifax	12	0	0%	0.0%	-	-	-	-	-	-
Conrod Settlement	Halifax	22	0	0%	0.0%	-	-	-	-	-	-
Cow Bay	Halifax	28	0	0%	0.0%	-	-	-	-	-	-
Dartmouth	Halifax	14	2	14%	0.0%	5.3	6.7	6	7	7.0	8
East Chester	Hants	15	4	27%	6.7%	7.2	16.0	5	9	13.3	30
East Chezzetcook	Halifax	31	3	10%	3.2%	8.3	8.5	7	10	39.0	100
East Petpeswick	Halifax	33	8	24%	3.0%	6.4	10.0	6	8.5	10.9	30
East Preston	Halifax	13	1	8%	0.0%	5.2	5.8	7	7	7.0	7
East Uniacke	Halifax	10	2	20%	0.0%	5.1	5.6	5	5.5	5.5	6
Eastern Passage	Halifax	22	1	5%	0.0%	5.1	5.0	7	7	7.0	7

Appendix A. Continued.

		All Data						Detects Only			
Community	County	Number of observations	Number of detects	Per cent of detects	Per cent exceedance[1]	K-M[2] Mean (µg/L)	95th percentile (µg/L)	Minimum (µg/L)	Median (µg/L)	Mean (µg/L)	Maximum (µg/L)
Ellershouse	Hants	10	0	0%	0.0%	-	-	-	-	-	-
Elmsdale	Hants	20	5	25%	0.0%	5.8	10.0	5	9	8.2	10
Enfield	Hants	29	8	28%	6.9%	8.8	26.0	5	15	18.8	46
English Corner	Halifax	18	3	17%	0.0%	5.0	5.0	5	5	5.0	5
Fall River	Halifax	194	34	18%	0.5%	5.8	10.0	5	8	9.3	29
Fletchers Lake	Halifax	13	0	0%	0.0%	-	-	-	-	-	-
Gaetz Brook	Halifax	24	2	8%	0.0%	5.3	5.9	6	8	8.0	10
Glen Haven	Halifax	12	5	42%	16.7%	19.5	82.8	5	9	39.8	140
Glen Margaret	Halifax	19	9	47%	31.6%	43.0	141.0	6	60	85.1	240
Grand Lake	Halifax	18	1	6%	0.0%	5.2	5.6	9	9	9.0	9
Hacketts Cove	Halifax	26	15	58%	30.8%	167.3	225.0	7	30	286.3	3500
Halifax	Halifax	22	3	14%	0.0%	5.5	9.9	7	10	9.0	10
Hammonds Plains	Halifax	387	224	58%	20.7%	19.5	70.0	5	20	30.0	260
Harrietsfield	Halifax	15	13	87%	53.3%	36.4	78.0	7	40	41.2	120
Hatchet Lake	Halifax	73	33	45%	21.9%	21.3	92.0	6	20	41.0	160
Head of Chezzetcook	Halifax	35	4	11%	0.0%	5.1	6.0	5	6	6.0	7
Head of Jeddore	Halifax	19	2	11%	0.0%	5.1	5.1	5	5.5	5.5	6
Head of St. Margarets Bay	Halifax	55	8	15%	3.6%	6.6	10.0	5	8.75	15.7	50
Herring Cove	Halifax	29	17	59%	20.7%	17.2	60.0	5	20	25.9	90
Hubbards	Halifax	45	15	33%	4.4%	15.2	18.6	5	10	35.5	380

Appendix A. Continued.

		All Data						Detects Only			
Community	County	Number of observations	Number of detects	Per cent of detects	Per cent exceedance[1]	K-M[2] Mean (µg/L)	95th percentile (µg/L)	Minimum (µg/L)	Median (µg/L)	Mean (µg/L)	Maximum (µg/L)
Hubley	Halifax	53	13	25%	0.0%	5.8	10.0	5	8	8.3	20
Indian Harbour	Halifax	14	5	36%	14.3%	15.9	60.5	8	20	35.4	80
Ketch Harbour	Halifax	17	6	35%	5.9%	10.2	24.0	5	9.5	19.8	80
Kinsac	Halifax	10	3	30%	0.0%	5.5	7.8	5	5	6.7	10
Lake Echo	Halifax	139	10	7%	0.0%	5.1	6.0	5	6	6.4	11
Lawrencetown	Halifax	65	5	8%	0.0%	5.1	5.0	5	5	6.2	10
Lower Sackville	Halifax	16	1	6%	0.0%	5.1	5.5	7	7	7.0	7
Lucasville	Halifax	11	0	0%	0.0%	-	-	-	-	-	-
Maders Cove	Lunenburg	10	0	0%	0.0%	-	-	-	-	-	-
Marriotts Cove	Lunenburg	11	1	9%	0.0%	5.4	7.0	9	9	9.0	9
Meaghers Grant	Halifax	17	3	18%	0.0%	5.1	5.2	5	5	5.3	6
Middle Musquodoboit	Halifax	12	1	8%	0.0%	5.0	5.0	5	5	5.0	5
Middle Sackville	Halifax	41	6	15%	2.4%	7.7	6.0	5	5.5	23.3	110
Milford	Hants	13	4	31%	0.0%	8.0	20.0	9	15	14.8	20
Mineville	Halifax	47	3	6%	0.0%	5.1	5.7	6	8	7.3	8
Mount Uniacke	Halifax	47	8	17%	0.0%	6.0	10.0	5	9.5	10.8	20
Musquodoboit Harbour	Halifax	59	6	10%	1.7%	5.5	5.0	5	5	10.0	30
New Ross	Lunenburg	27	16	59%	44.4%	63.3	124.0	5	50	103.3	800

Appendix A. Continued.

		All Data						Detects Only			
Community	County	Number of observations	Number of detects	Per cent of detects	Per cent exceedance[1]	K-M[2] Mean (µg/L)	95th percentile (µg/L)	Minimum (µg/L)	Median (µg/L)	Mean (µg/L)	Maximum (µg/L)
Nine Mile River	Hants	13	4	31%	0.0%	7.5	18.8	6	13.5	13.3	20
Oakfield	Halifax	10	8	80%	10.0%	8.8	21.0	5	6.5	9.8	30
Ostrea Lake	Halifax	10	2	20%	0.0%	5.5	7.8	5	7.5	7.5	10
Porters Lake	Halifax	164	9	5%	0.6%	5.8	5.0	5	6	19.6	100
Prospect	Halifax	52	21	40%	9.6%	13.5	80.0	5	10	26.1	100
Queensland	Halifax	11	2	18%	9.1%	11.4	40.0	20	40	40.0	60
Sambro	Halifax	14	6	43%	14.3%	12.7	33.5	8	20	23.0	40
Seabright	Halifax	23	8	35%	17.4%	26.2	58.0	5	25	66.0	360
Seaforth	Halifax	13	2	15%	0.0%	5.3	6.6	5	7	7.0	9
Shad Bay	Halifax	35	8	23%	2.9%	6.8	20.0	5	8	13.0	30
Sheet Harbour	Halifax	15	0	0%	0.0%	-	-	-	-	-	-
Ship Harbour	Halifax	11	1	9%	9.1%	14.6	57.5	110	110	110.0	110
Shubenacadie	Hants	12	3	25%	0.0%	5.6	7.8	6	6	7.3	10
Stillwater Lake	Halifax	205	75	37%	5.4%	8.7	28.0	5	8	15.1	120
Tantallon	Halifax	84	26	31%	6.0%	8.1	25.1	5	9.5	15.0	60
Terence Bay	Halifax	22	9	41%	0.0%	7.0	19.5	5	7	9.8	20
Three Fathom Harbour	Halifax	10	3	30%	10.0%	12.6	46.7	5	6	30.3	80
Timberlea	Halifax	27	6	22%	0.0%	5.6	9.7	5	8.5	7.8	10
Truro	Colchester	29	5	17%	0.0%	5.9	9.6	5	9	10.2	20
Upper Sackville	Halifax	33	4	12%	0.0%	5.1	5.4	5	5.5	5.8	7

Appendix A. Concluded.

		All Data						Detects Only			
Community	County	Number of observations	Number of detects	Per cent of detects	Per cent exceedance[1]	K-M[2] Mean (µg/L)	95th percentile (µg/L)	Minimum (µg/L)	Median (µg/L)	Mean (µg/L)	Maximum (µg/L)
Upper Tantallon	Halifax	13	4	31%	0.0%	6.9	14.0	6	9.5	11.3	20
Waverley	Halifax	23	5	22%	0.0%	5.2	5.9	5	5	6.0	9
Wellington	Halifax	53	6	11%	0.0%	5.2	5.8	5	6	7.0	10
West Chezzetcook	Halifax	12	2	17%	0.0%	5.1	5.5	5	5.5	5.5	6
West Jeddore	Halifax	15	1	7%	0.0%	5.2	5.9	8	8	8.0	8
Whites Lake	Halifax	63	34	54%	27.0%	35.4	170.0	5	30	61.3	210
Williamswood	Halifax	61	47	77%	37.7%	28.0	72.0	5	20	34.9	130

* These surveys were not part of the Uranium Task Force work.

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

b. Uranium Task Force (MacFarlane, 1983)