

Occurrence and Mobilization of Uranium in Groundwater in Nova Scotia



John Drage

Nova Scotia Department of Natural Resources, Halifax, Nova Scotia, Canada

dragejo@gov.ns.ca

Gavin W. Kennedy

Nova Scotia Department of Natural Resources, Halifax, Nova Scotia, Canada

kennedgw@gov.ns.ca

ABSTRACT

Naturally occurring uranium exceeds drinking water guidelines in approximately 4% of water wells in Nova Scotia. This study looked at the association between calcium and uranium using the provincial 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 zero-valent calcium-uranyl-carbonate complex. 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 releasing calcium by ion exchange. Therefore, caution should be exercised when planning anthropogenic activities that can release calcium to the ground in areas with naturally occurring uranium.

RÉSUMÉ

L'uranium naturel dépasse les normes pour l'eau potable dans environ 4% des puits en Nouvelle-Écosse. Cette étude porte sur les liens entre le calcium et l'uranium, qui ont été étudiés en utilisant la base de données provinciale géochimique et de la modélisation de la spéciation chimique. Les résultats ont montré des corrélations positives fortes entre l'uranium, le calcium et les chlorures. La modélisation de la spéciation a indiqué que le calcium peut avoir une influence importante sur la mobilité de l'uranium dans les eaux souterraines due à la formation du complexe mobile à valence nulle de calcium-uranyle-carbonate. Les résultats indiquent également qu'ajouter du sel aux eaux souterraines (via intrusion d'eau de mer ou déglacage des routes) peut avoir un effet semblable sur la mobilisation de l'uranium, en ajoutant directement un peu de calcium, ou en libérant le calcium par échange ionique. Des précautions devraient être prises lors de la planification d'activités qui peuvent libérer du calcium dans les secteurs où de l'uranium naturel est présent.

1 INTRODUCTION

Naturally occurring uranium was first identified in groundwater in Nova Scotia in 1978 after elevated uranium levels were discovered in hair samples from a person who obtained drinking water from a drilled well. Subsequent province-wide investigations indicated that uranium commonly occurs in drilled wells above the Canadian Drinking Water Guideline of 20 µg/L (Health Canada, 2012) in certain geologic formations within the province, particularly plutonic and sedimentary rocks.

Roughly half the population of Nova Scotia relies on groundwater for its water supply and approximately 4% of water wells across the province exceed the uranium drinking water guideline. This makes uranium the second most common naturally occurring groundwater contaminant to exceed its guideline, after arsenic.

Since the 1980s, several studies have contributed to the understanding of the occurrence and mobilization of uranium and its daughter products in groundwater in Nova Scotia. This paper provides a brief summary of the occurrence of uranium in groundwater in Nova Scotia and findings from previous uranium studies, and then presents results from a recent investigation on how anthropogenic

activities that release calcium in aquifers appear to have mobilized uranium in groundwater in Nova Scotia.

1.1 Uranium Occurrence in Well Water in Nova Scotia

Figure 1 shows the distribution of uranium in well water samples within the five major bedrock groundwater regions in the province and Table 1 shows summary statistics for the province (Kennedy and Finlayson-Bourque 2011). It is estimated that approximately 4% of water wells in the province exceed the uranium drinking water guideline of 20 µg/L, and the majority of these wells are located in plutonic or sedimentary rocks (Figure 1). Figure 2 shows the estimated percentage of household wells that exceed the uranium drinking water guideline in each of the province's major groundwater regions by extrapolating the exceedance ratios observed in the provincial groundwater chemistry database. As indicated in Figure 2, 21% of well water samples from the plutonic groundwater region exceed the uranium guideline (representing approximately 5,300 households) and 3% of well water samples from the sedimentary groundwater region exceed the guideline (representing approximately 2,400 households).

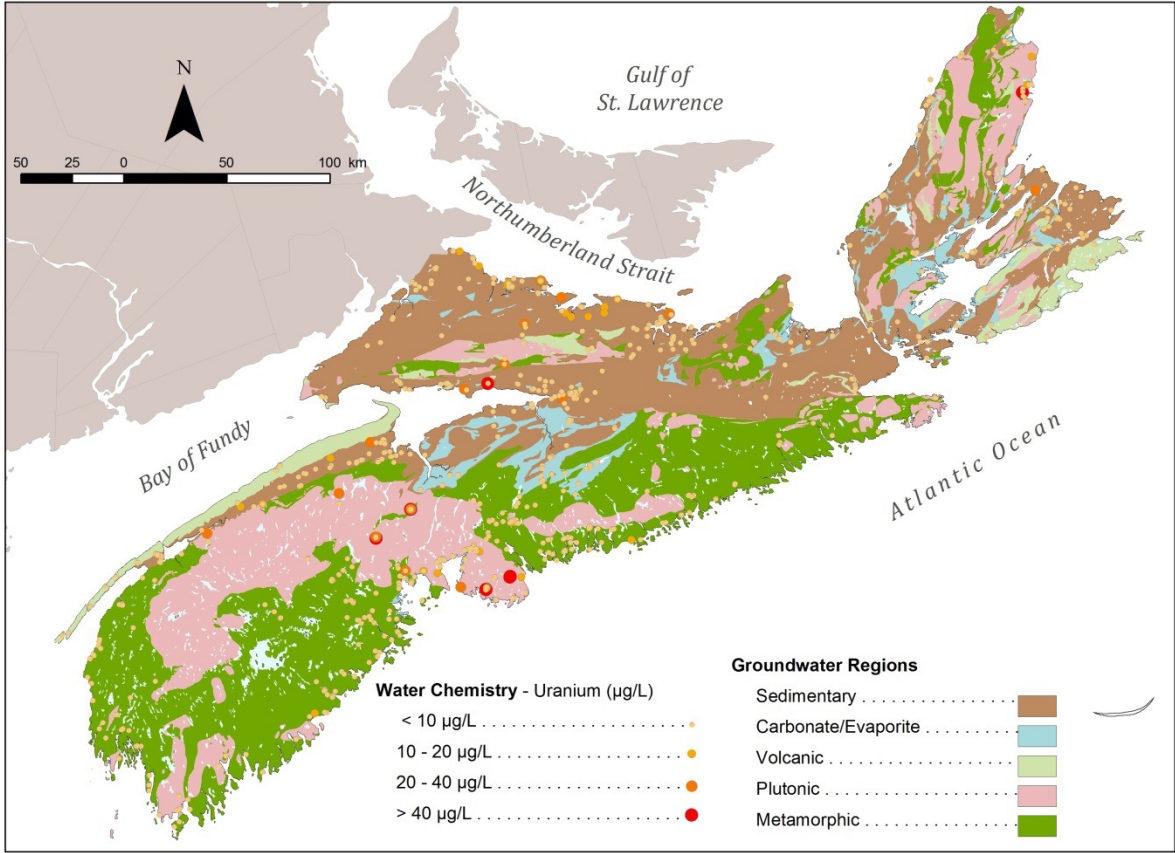


Figure 1. Uranium in bedrock aquifers in Nova Scotia

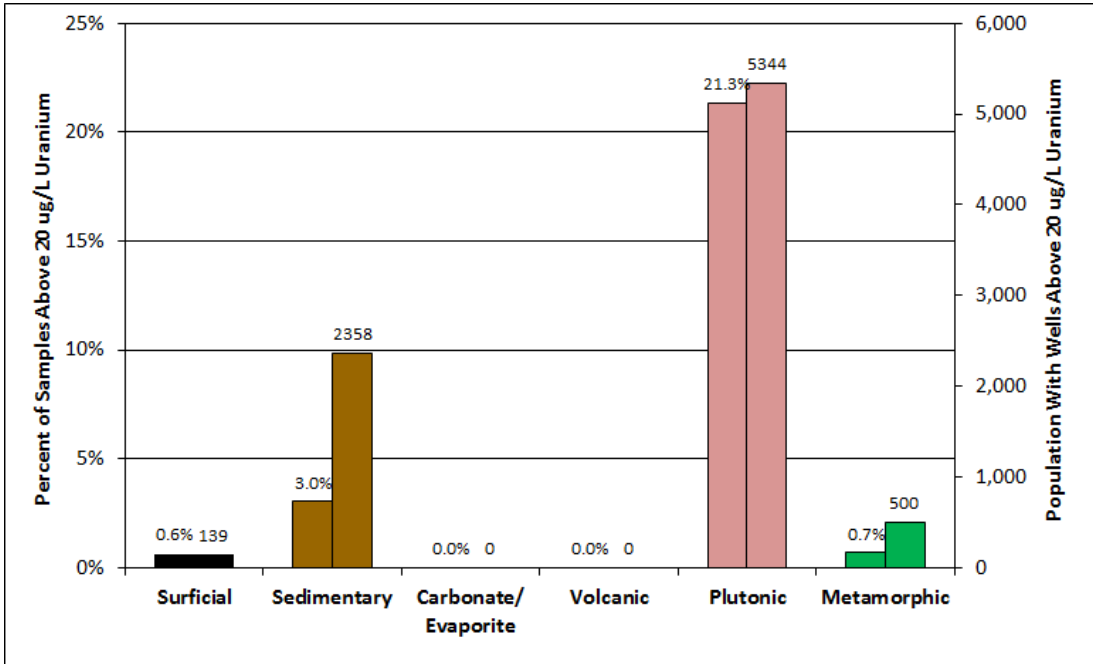


Figure 2. Uranium in well water by region in Nova Scotia

Table 1. Summary of uranium statistics in well water in Nova Scotia (from Kennedy and Finlayson-Bourque 2011)

Parameter	Value	Units
Number of samples	840	-
Maximum concentration	200	µg/L
Median concentration	0.7	µg/L
Mean concentration	4.3	µg/L
Percent above drinking water guideline ¹	4	%

¹The Maximum Acceptable Concentration for uranium in drinking water is 20 µg/L (Health Canada 2012)

1.2 Previous Studies in Nova Scotia

The first major investigation into uranium in drinking water in Nova Scotia was initiated in 1979 by the Provincial Uranium Task Force (Nova Scotia Department of Health 1986). The work involved sampling for uranium, radium and radon in public and private water supplies from across the province, although much of the sampling was focused on areas that were suspected to be high risk for uranium due to the geologic setting. Of the 784 wells that were sampled, 25% had uranium levels exceeding 20 µg/L and 99% of the exceedances occurred in drilled wells. Wells drilled into granite and Upper Carboniferous sandstone and shale were identified as most likely to have elevated uranium levels. Uranium was found to be strongly related to high pH values, and its mobility was related to oxidizing, alkaline conditions, as well as the formation of complexes of phosphate, carbonate, silicate and fluoride. Radium was commonly detected, but only 1% of the wells exceeded the drinking water guideline. Approximately 40% of the wells had radon levels above 370 Bq/L (10,000 pCi/L), although there is currently no Canadian drinking water guideline for radon.

In 2002, lead-210 was identified in well water at a public school in Nova Scotia. Lead-210 is a daughter product of uranium, which was not previously tested for in the province. As a result, another province-wide radionuclide testing program was initiated. Initial results indicated that both uranium and lead-210 commonly exceeded drinking water guidelines, but subsequent work revealed that the laboratory method did not provide a realistic indication of lead-210 levels because radon gas was rapidly decaying (i.e., radon's half-life is 3.8 days) to lead-210 while the samples were in transit and being analyzed at the laboratory. The sampling protocol was modified to eliminate radon decay effects and further testing indicated lead-210 was not a common problem in groundwater in Nova Scotia (Drage et al. 2005).

In 2006, moderate levels of uranium were identified in test wells during the development of a municipal wellfield in a fractured sandstone aquifer. Initial pumping tests and chemical tests on the open-hole test wells suggested uranium levels would remain below the drinking water guideline. Uranium levels increased above the drinking water guideline, however, after the test wells were converted to production wells using partial screens across the productive aquifer intervals. Subsequent investigations (involving leachate testing of rock samples, depth-specific groundwater sampling and discrete fracture

sampling) suggested that uranium may have been re-mobilized due to the development of oxidizing conditions during pumping (Finlayson-Bourque et al. 2010).

In 2006, a study was carried out in the Grand Pré region of Nova Scotia to investigate the influence of redox conditions and regional geology on the mobility of uranium in groundwater (Samolczyk et al. 2012). The work involved sampling 17 private water wells in fractured shale and sandstone aquifers, with nine of the wells selected for detailed chemistry analyses to determine redox conditions. The results indicated that redox conditions strongly influenced uranium levels. The higher uranium levels were primarily associated with oxidizing environments that contained low iron and manganese, while low uranium levels were typically associated with reducing environments that contained high iron and manganese.

1.3 Objectives

Elevated uranium levels were recently identified at a site in Nova Scotia where anthropogenic activities had introduced dissolved calcium into an aquifer. Calcium has been reported in the literature to influence the mobility of uranium in groundwater due to the formation of mobile, calcium-uranyl-carbonate complexes, $\text{Ca}_2\text{UO}_2(\text{CO}_3)_3^0$ and $\text{CaUO}_2(\text{CO}_3)_3^{2-}$. The objective of this study was to:

- review groundwater chemistry data from across Nova Scotia to determine if there is an association between dissolved calcium and elevated levels of uranium; and
- carry out aqueous speciation modelling to determine if the formation of calcium-uranyl-carbonate complexes is likely to influence uranium mobility in groundwater in Nova Scotia.

2 METHODS

Groundwater chemistry data for the study came from two main sources: (1) Nova Scotia Groundwater Chemistry Database (NSGCDB) (NSDNR 2013), and (2) Grand Pré dataset (Samolczyk et al. 2012).

The NSGCDB contains data from across the province and has been compiled from various federal, provincial and municipal groundwater chemistry data sources, including well water quality monitoring data from government buildings, and Nova Scotia Environment groundwater chemistry data from registered public drinking water supplies, pumping tests, municipal groundwater systems and provincial observation wells. The NSGCDB contains over 2,400 well water samples with uranium results, but only 2,100 of these also had calcium results available.

The Grand Pré dataset (Samolczyk et al. 2012), which was discussed in Section 1.2, contains data for 17 wells in the Grand Pré region of Nova Scotia. These data were used because they represent one of the few published datasets in Nova Scotia that contains redox data needed for speciation modelling.

Aqueous speciation modelling was carried out with PHREEQC, Version 3.0.1 (Parkhurst and Appelo 2013), using the Wateq4f.dat thermodynamic database within

PHREEQC. Although the Wateq4f.dat database includes thermodynamic data for uranium speciation, it does not include data for calcium-uranyl-carbonate complexes that might be expected to form in the presence of calcium. Figure 3 shows aqueous speciation calculations for uranium (VI) in the presence of calcium with respect to pH. The median pH value of the well water samples modeled in this study was 7.5, and Figure 3 indicates that at this pH value, the calcium-uranyl-carbonate complexes can be an important component of the total dissolved uranium. Therefore, data for calcium-uranyl-carbonate complexes were added to the Wateq4f.dat database. Thermodynamic data were added for magnesium and strontium-uranyl-carbonate complexes because these have also been reported to be potentially important uranium complexes (Nair and Merkel 2011). The thermodynamic data added to the Wateq4f.dat database are shown in Table 2.

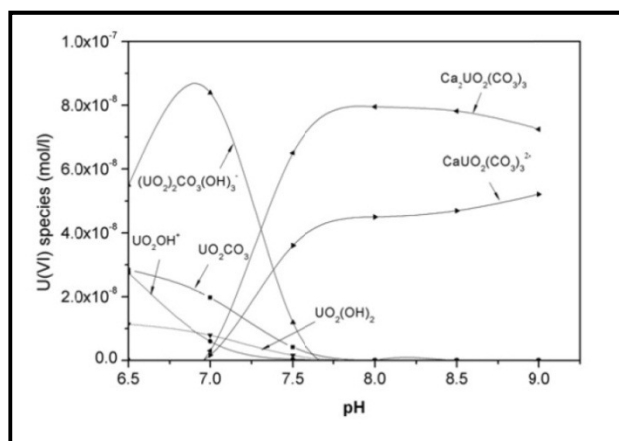


Figure 3. Aqueous speciation of calcium-uranyl-carbonate species in relation to pH ($U=0.126 \times 10^{-6} \text{M}$, $Ca=1 \times 10^{-3} \text{M}$, $NaCl=1.5 \times 10^{-3} \text{M}$, $NaHCO_3=1 \times 10^{-3} \text{M}$, $P_{CO_2}=10^{-3.5} \text{hPa}$, 23°C) (from Nair and Merkel 2011)

Table 2. Additional uranyl-carbonate species added to Wateq4f.dat database for aqueous speciation modelling

Aqueous Reaction	Log K^1
$2Ca^{+2} + UO_2^{+2} + 3HCO_3^- = Ca_2UO_2(CO_3)_3^0 + 3H^+$	-0.29 ^a
$Ca^{+2} + UO_2^{+2} + 3CO_3^{-2} = CaUO_2(CO_3)_3^{-2}$	27.18 ^b
$Mg^{+2} + UO_2^{+2} + 3CO_3^{-2} = MgUO_2(CO_3)_3^{-2}$	26.11 ^b
$Sr^{+2} + UO_2^{+2} + 3CO_3^{-2} = SrUO_2(CO_3)_3^{-2}$	26.86 ^b
$2Sr^{+2} + UO_2^{+2} + 3CO_3^{-2} = Sr_2UO_2(CO_3)_3^0$	29.73 ^b

¹K is the stability constant; ^afrom Jurgens et al. (2010); ^bfrom Nair and Merkel (2011)

3 RESULTS AND DISCUSSION

3.1 Aqueous Speciation

Aqueous speciation modelling indicated that all of the dissolved uranium present in the well water samples was present as uranium (VI). The predicted concentrations of

uranium (III), uranium (IV) and uranium (V) were negligible.

Table 3 summarizes the results from the speciation calculations for the Nova Scotia samples. The dominant uranium species present were calcium-uranyl-carbonate complexes ($Ca_2UO_2(CO_3)_3^0$ and $CaUO_2(CO_3)_3^{-2}$), and the majority of uranium was present as the zero-valent calcium-uranyl-carbonate complex. The data show that the median proportion of $Ca_2UO_2(CO_3)_3^0$ and $CaUO_2(CO_3)_3^{-2}$ was 89% and 10%, respectively. Furthermore, the dissolved uranium in 85% of the well water samples consisted of more than 80% of the zero-valent calcium-uranyl-carbonate complex.

Other complexes generally constituted minor amounts of the total uranium. In 25% of the samples, the carbonate complex $UO_2(CO_3)_3^{-2}$ constituted between 1% and 7% of the total uranium. Magnesium complexes constituted less than 1% of the total uranium. Strontium complexes were negligible. Although the ternary calcium-uranyl-carbonate complexes were the dominant uranium species in most cases, it should be noted that in samples that had elevated levels of dissolved phosphorous, the dominant species was $UO_2(HPO_4)_2^{-2}$. This was observed in only one of the well water samples.

Table 3. Aqueous speciation modelling results for Nova Scotia groundwater samples

Uranium Species ¹	Min	Median	Max
Total Uranium ($\mu\text{g/L}$)	1.4	73	200
$Ca_2UO_2(CO_3)_3^0$	6%	89%	100%
$CaUO_2(CO_3)_3^{-2}$	<1%	10%	17%
$UO_2(CO_3)_3^{-2}$	<1%	<1%	7%

¹Percentages show species proportion, relative to total uranium

The speciation results obtained in this study are similar to those reported by other authors. In a case study that looked at 350 well water samples from Central Valley, California, Jurgens et al. (2010) reported that the dissolved uranium in the majority of samples was mostly in the form of the zero-valent calcium-uranyl-carbonate complex. The presence of this zero-valent species in groundwater has important implications for the mobility of uranium because it is less likely to sorb to the aquifer matrix compared to other uranium complexes. Fox et al. (2006) reported that uranium (VI) adsorption to iron hydroxides can be reduced because of the formation of the zero-valent calcium-uranyl-carbonate complex. Furthermore, as shown in Figure 4, batch experiments on the sorption of uranium (VI) on quartz have shown that the addition of calcium can significantly reduce the amount of sorption, from 90% sorption when calcium is not present, to 10% sorption when calcium is present (Nair and Merkel 2011). This effect was also attributed to the formation of zero-valent and anionic calcium-uranyl carbonate complexes.

3.2 The Role of Calcium

Several authors (Dong et al. 2005, Fox et al. 2006, Nair and Merkel 2011) have investigated the influence of dissolved calcium on the speciation and sorption of

uranium (VI). The association between calcium and uranium in well water in Nova Scotia was investigated using groundwater data from the NSGCDB (as described in Section 2). Figure 5 shows the strong positive correlation between uranium and calcium concentrations for uranium samples in the Nova Scotia data that had greater than 40 µg/L uranium (correlation coefficient = 0.73). This suggests that calcium plays a role in the mobilization of uranium in Nova Scotia.

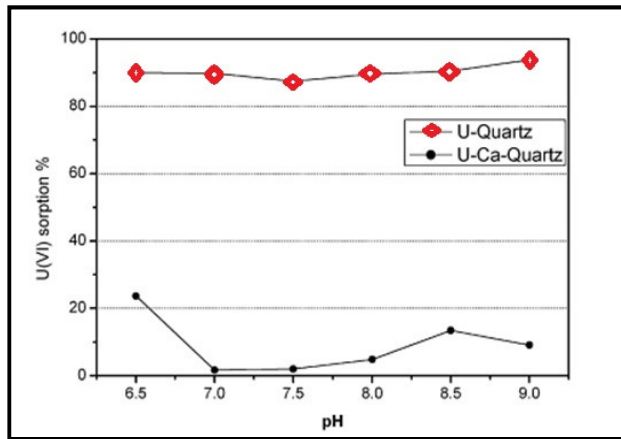


Figure 4. Batch experiment results showing the percent uranium (VI) sorption on quartz with and without dissolved calcium present ($U=0.126 \times 10^{-6} M$, $Ca=1 \times 10^{-3} M$, $NaCl=1.5 \times 10^{-3} M$, $NaHCO_3=1 \times 10^{-3} M$, $P_{CO_2}=10^{-3.5} hPa$, $23^\circ C$, 48hours) (from Nair and Merkel 2011)

The strong correlation was not evident in samples with lower uranium levels, possibly because not all wells are located in areas that have significant amounts of leachable uranium present. It should be noted that the geochemical modelling indicated that all of the groundwater samples were undersaturated with respect to uranium minerals.

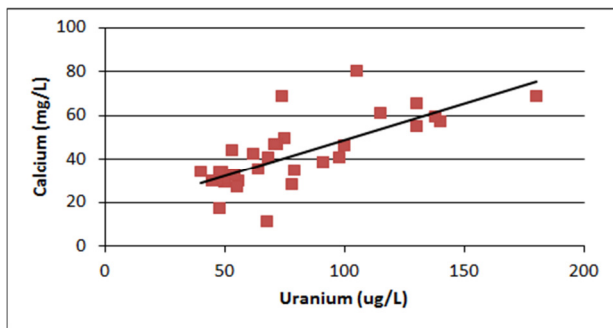


Figure 5. Uranium versus calcium in Nova Scotia well water, for samples greater than 40 µg/L uranium (correlation coefficient = 0.73)

A similar association was identified between chloride and uranium. Figure 6 shows a moderately strong positive correlation between uranium and chloride concentrations for uranium samples in the Nova Scotia data that had

greater than 40 µg/L uranium (correlation coefficient = 0.67). In fact, the majority of samples shown in Figures 5 and 6 (i.e., 69%) had chloride levels that exceeded the typical maximum background chloride level for water wells in coastal areas of Nova Scotia (i.e., 50 mg/L). The most common causes of elevated chloride levels in well water in Nova Scotia are road salt and seawater intrusion, although locally there may be other sources, such as natural formation salt or the discharge of backwash from water treatment units (e.g., water softeners). The sample with the highest uranium level in the Grand Pré dataset also had the highest chloride level and had a bromide/chloride ratio of 6.0 (i.e., $Br/Cl \times 10^4$), which is indicative of road salt impacts (Briggins and Cross 1995). The sample with the highest uranium level in the NSGCDB was associated with a site previously identified as impacted by seawater intrusion (NSDNR 2013).

Neither road salt impacts nor seawater intrusion are expected to introduce significant amounts of uranium, since uranium is not reported to be one of the trace metals present in road salt (Environment Canada and Health Canada 2001) and seawater is reported to contain only 3 µg/L uranium (Hem 1985).

Both of these types of impacts, however, may introduce calcium and, therefore, may promote the formation of calcium-urananyl-carbonate complexes. Road salt leachate in Nova Scotia has been reported to contain 50 mg/L of calcium (with 1,260 mg/L of sodium and 1,800 mg/L of chloride) (Briggins and Cross 1995) and seawater contains 410 mg/L of calcium (with 10,500 mg/L of sodium and 19,000 mg/L of chloride) (Hem 1985). In addition, both of these types of impacts introduce high levels of sodium to groundwater, which has been reported to release calcium due to sodium-calcium ion exchange (Bäckström et al. 2004).

The addition of chloride into groundwater has also been reported to lead to mobilization of metals because of the formation of metal-chloride complexes (Bäckström et al. 2004). The geochemical modelling carried out on the Nova Scotia groundwater samples, however, indicated that uranium-chloride complexes were not present and, therefore, it is more likely that calcium-urananyl-carbonate complexes are responsible for uranium mobilization in the Nova Scotia samples.

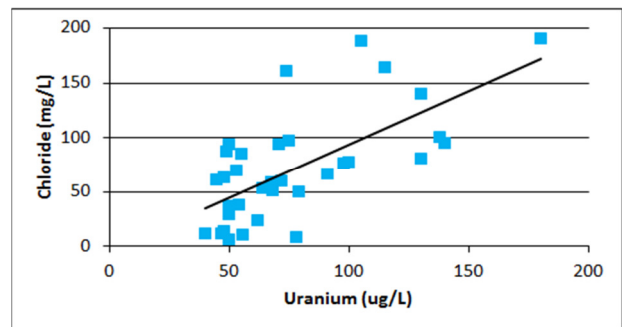


Figure 6. Uranium versus chloride in Nova Scotia well water, for samples greater than 40 µg/L uranium (correlation coefficient = 0.67)

The effect that dissolved calcium has on the formation of the zero-valent calcium-uranyl-carbonate complex was evaluated with PHREEQC by varying the amount of calcium present and holding other chemical concentrations constant. This was done using groundwater chemistry data from the NSGCDB. Chloride was adjusted proportionally with the calcium level to maintain ion balance. Figure 7 shows the modelling results, which indicate that the percentage of total uranium present as zero-valent calcium-uranyl-carbonate increases as the concentration of calcium in the sample increases. This suggests that adding calcium to the system can cause uranium to become more mobile in an aquifer.

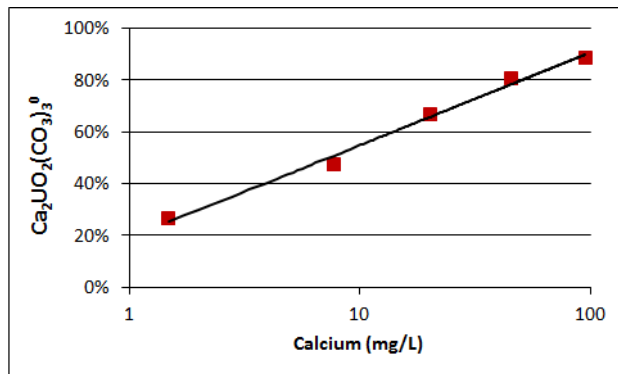


Figure 7. Percentage of zero-valent calcium-uranyl-carbonate ($\text{Ca}_2\text{UO}_2(\text{CO}_3)_3^0$) in groundwater as dissolved calcium level changes, as predicted by PHREEQC

3.3 Field Examples of Uranium Mobilization

Nova Scotia groundwater chemistry data from the NSGCDB were reviewed for examples where increasing levels of calcium were associated with increasing levels of uranium. Several sites were identified where uranium levels were strongly correlated with calcium. Figures 8 and 9 show examples of public water supply wells that have rising trends in chloride, calcium and uranium concentrations. The source of elevated chloride in both of these wells is suspected to be road salt, due to the distance of the sites from seawater (>1 km) and the absence of other potential sources of salt. In both of these examples the uranium levels show very strong positive correlations with both calcium and chloride (i.e., correlation coefficients >0.9, except for Figure 8 where the uranium-calcium correlation coefficient was 0.73).

These field examples of road salt contamination, and the saltwater intrusion case discussed in Section 3.2, suggest that anthropogenic activities can mobilize naturally occurring uranium in groundwater by either directly or indirectly increasing the amount of dissolved calcium in an aquifer. Elevated uranium levels have also been recently identified in water wells near a construction and demolition debris site in Nova Scotia where significant amounts of waste drywall are present.

Drywall is made of gypsum ($\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$), which is highly soluble in water and, therefore, can introduce

significant amounts of calcium into groundwater if allowed to leach into the ground. Drywall makes up approximately 20% of the construction and demolition waste stream in Nova Scotia. It has been proposed that it be diverted from the waste stream by using it as a compost feedstock (Nova Scotia Resource Recovery Fund Board Inc. 2007). Adding gypsum to soil and compost is considered beneficial because it reduces acidity and provides a source of calcium and sulphur. Precautions should be taken, however, so that these practices do not inadvertently cause uranium to be mobilized in groundwater.

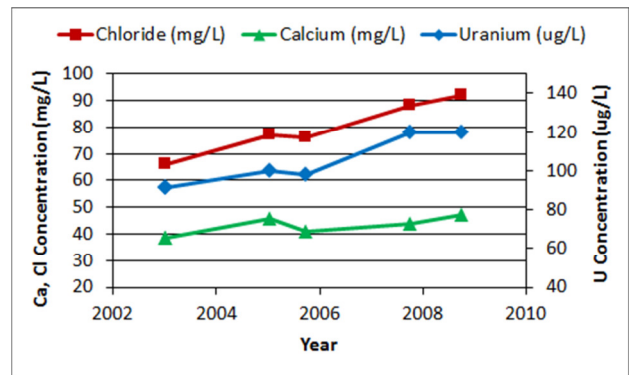


Figure 8. Site A: public water supply well with increasing road salt impacts and uranium concentrations

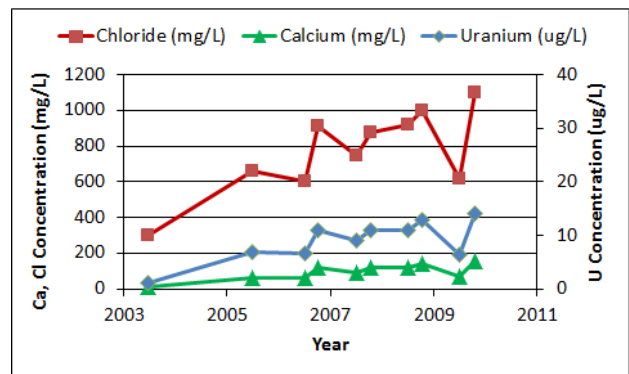


Figure 9. Site B: public water supply well with increasing road salt impacts and uranium concentrations

4 CONCLUSIONS

The results from this study indicate that calcium can be an important influence on the mobility of uranium in groundwater in Nova Scotia due to the formation of the mobile, zero-valent calcium-uranyl-carbonate complex. 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 because it can directly add small amounts of calcium, or because it can cause calcium to be released from the aquifer due to ion exchange.

Therefore, caution should be exercised whenever anthropogenic activities are being planned that can release dissolved calcium into the ground in areas with naturally occurring uranium. The highest risk areas of the province for uranium mobilization are the plutonic and sedimentary groundwater regions. Examples of anthropogenic activities that have potential to release calcium in aquifers include: road salt de-icing, seawater intrusion due to groundwater pumping near the coastline, the processing or disposal of construction and demolition debris, and the amending of soil or compost with gypsum or waste drywall.

Previous studies in Nova Scotia have determined that oxidizing conditions are also important to uranium mobilization in groundwater. Therefore, in areas with naturally occurring uranium, caution should also be exercised when activities are planned that can introduce oxygen into aquifers. This may occur when excessive drawdown is caused by groundwater pumping.

5. ACKNOWLEDGMENTS

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