Potential for Uranium Mobilization from Weathered Outcrops of Uranium-Bearing Sedimentary Strata, Southern Nova Scotia

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Abstract

Nova Scotia has a number of uranium occurrences within sedimentary sequences of the Carboniferous Horton Group. These occurrences pose a potential risk to human health if the uranium enters drinking water supplies. This research focuses on a uranium bearing occurrence of the Horton Group strata to establish both the nature of the uranium distribution in the Horton Group and to determine whether uranium in the strata may become chemically mobile under present day weathering conditions. The main outcrop section of interest is currently undergoing active weathering, and is located upslope from a small stream. Samples were taken from two sandstone and two siltstone layers, as well as from unconsolidated outwash material located at the base of the outcrop. The samples were analyzed to determine major, trace, and rare earth element geochemistry, and also underwent artificial weathering to determine what elements may be liberated from the rocks during weathering.

Geochemical data indicate that there are differences in uranium levels in the samples, but does not indicate that levels are linked to lithology within the Formation. The highest uranium concentration is from one of the siltstones (212 ppm) and the second highest value is from one of the sandstones (109 ppm). Geochemical data also indicate that higher uranium values correspond to higher values of copper, vanadium and titanium. Positive correlations exist between uranium and copper, lead, and vanadium for the samples analysed. Values for uranium, copper, vanadium and titanium are all lowest in the sample taken from the outwash material near the stream, suggesting liberation of uranium during weathering.

Water samples from artificial weathering experiments indicate that a number of elements, including uranium, are mobilized during weathering. Values for total uranium in water samples were as high as 24µg/L, exceeding the recommended guideline for Canadian drinking water.

1.0 Introduction

1.1 Statement of Purpose

Uranium is a naturally occurring element that can be found in rocks, soils, and waters. In Nova Scotia, many areas have naturally elevated uranium levels, often concentrated in specific geologic formations, such as the granitoids of the South Mountain Batholith, and the sedimentary strata of the Horton Group (O'Reilly, 1982)(Table 1.1). This is of concern to many residents of these and surrounding areas, because of the radioactive nature and toxicity of uranium and its daughter products, and the evidence that these elements may be linked to various cancers and birth defects in humans (US Environmental Protection Agency, 2006). Radon, a daughter product of uranium, has been linked to lung cancer, especially when present in elevated levels in indoor air (Ball et al., 1991). This thesis attempts to (1) determine the levels of uranium in Horton Group sandstones and shales, and their weathered equivalents, (2) to investigate elemental associations with the uranium in these rocks, and (3) to determine under what present day conditions the uranium in these rocks is chemically mobile during weathering. I will also make recommendations regarding water source contamination.

1.2 Study Area

For this thesis, units of interest lie within the Cheverie Formation of the Horton Group of Lower Carboniferous age (Bell, 1929). The area studied is located in the vicinity of Green Street, near Windsor, Nova Scotia (Fig. 1.1). This area was chosen because of a known uranium anomaly (Ryan, 1998), as well as the close proximity to

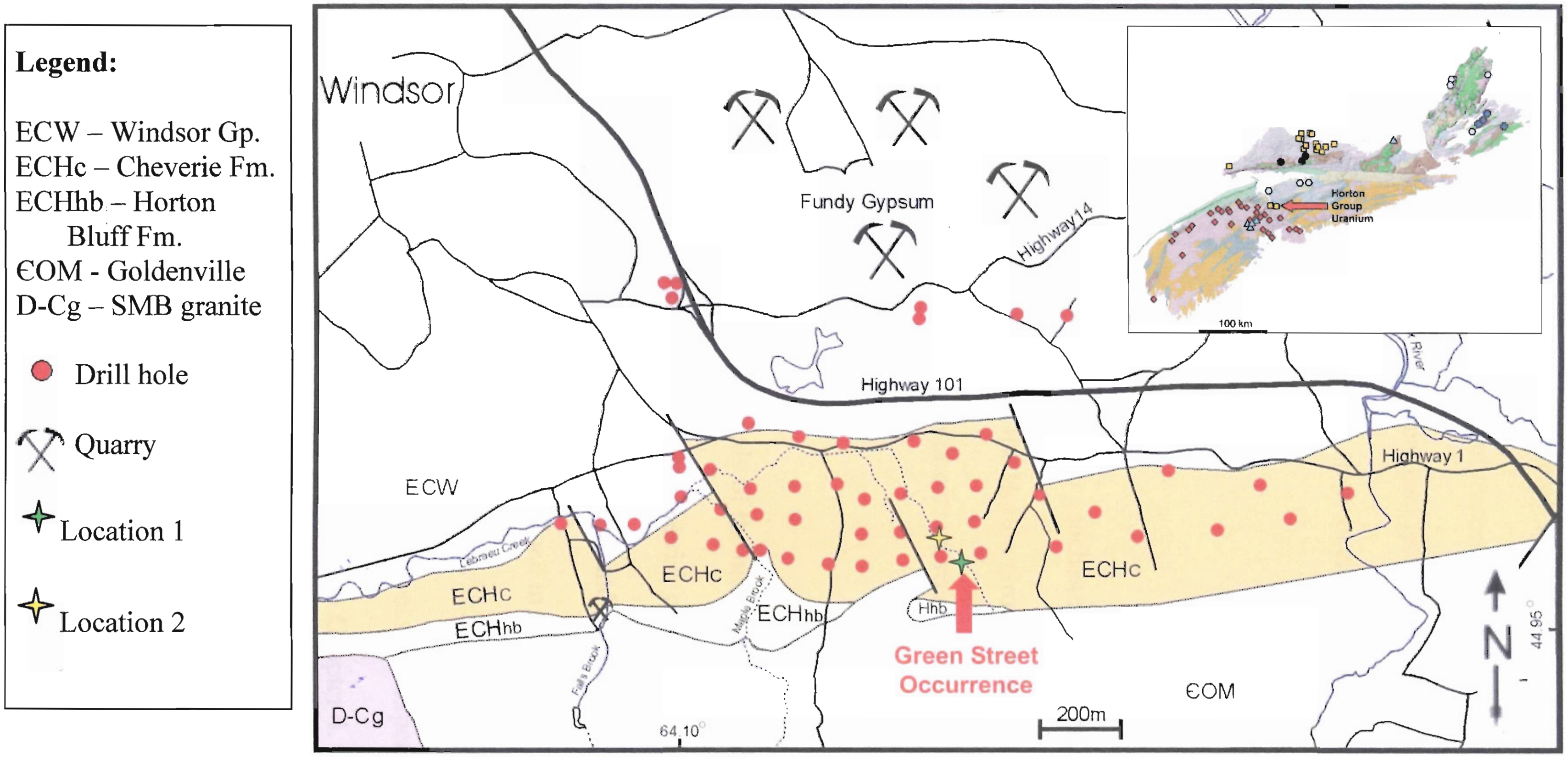


Figure 1.1 – Location of Green Street Uranium Occurrence, Near Windsor, NS (After Ryan and O'Beirne-Ryan, 2007)

surrounding surface water. The main outcrop studied is located on a roadside, where it extends over a distance of several tens of meters, within which are localized pockets of uranium values several times the normal background, as measured with a handheld scintillometer. The outcrop is also on a slope in close proximity to a river, and drains into farmland located downslope.

1.3 Project Objectives

The objectives of this project are to geochemically characterize the uranium-enriched clastic sedimentary rocks of the Horton Group, and to determine if there is potential for uranium mobilization under current weathering conditions.

1.4 Format of Thesis

This thesis is a geochemical study of the uranium-enriched sedimentary strata of the Cheverie Formation that will determine the risk of uranium migration from exposed outcrops of these strata to surface waters and ground waters. The geology of the study area and the geochemistry of uranium are outlined in Chapter 2. The methods used in determining the levels and possible mobility of the uranium under natural conditions are considered in Chapter 3, and include field work, laboratory experiments, and analytical procedures. Results are presented and summarized in Chapter 4. Chapter 5 is a discussion and interpretation of the data and a consideration of the results as they relate to surface waters and ground waters in Nova Scotia. Chapter 6 summarizes the conclusions, and proposes recommendations for future study.

Table 1.1 – Average uranium values in rocks of the South Mountain Batholith and the Horton Group, compared to world averages (Modified from O'Reilly, 1982) (*approximate value, personal communication, Bob Ryan)

Area	**Rock Type**	**U** (ppm)
South Mountain Batholith	Granodiorite	3.9
	Monzogranite	6.1
	Leuco-monzogranite	8.3
Horton Group	Arkosic Sandstone	6*
World Averages	Dioritic	2
	Granitic	4

2.0 Geology

2.1 Introductory Statement

There are many areas in Nova Scotia in which the local rocks have natural elevated Uranium levels. For this thesis, I will be focusing on the sandstones and siltstones of the Horton Group, specifically those of the Cheverie Formation. The geochemistry of these rocks has not been undertaken previously, nor has consideration of the weathering potential been examined. The following section describes the detailed geology of the Horton Group, as well as the South Mountain Batholith, a possible source of Horton Sedimentary material.

2.2 The Horton Group

The Horton Group is a unit of sedimentary rocks of late Devonian to Early Carboniferous age (Van de Poll et al.1995). It is composed of red-grey green polymictic conglomerate, arkosic sandstone, mudstone, oil shales, and minor non-marine evaporites. The Horton Group was deposited in a series of Northeast-East trending basins separated by uplifted blocks of basement material (Van de Poll et al. 1995). This occurred during an extensional phase following the Acadian Orogeny (Martel and Gibling, 1995).

Commonly, the Horton Group overlies Pre-Acadian metamorphosed basement rocks with an angular unconformity, and in places is overlain conformably to unconformably by limestone from the Windsor Group (Van de Poll et al. 1995). The Horton Group also lies non-conformably on the South Mountain Batholith of Devonian age in many areas, and for this reason, the granites of the South Mountain Batholith are a likely source of sediment during the formation of much of the Horton Group (Figure 2.1).

The Horton Group is composed of the lower Horton Bluff Formation and the upper Cheverie Formation (Bell, 1929). The Cheverie Formation consists of coarse grained braided channel deposits and red floodplain sediments and paleosols (Ryan, 1998) (Figure 2.2). It lies disconformably on the lower Horton Bluff Formation, but this unconformity does not represent a large time period. The Cheverie Formation consists of sandstones and siltstones which are known to have natural elevated levels of uranium (Ryan, 1998). The sandstones are coarse grained and variably weathered. The siltstone appears as thin lenses (usually less than 5 cm thick), and are also variably weathered.

2.3 Geologic History

The South Mountain Batholith is late Devonian in age and consists of a series of granitic plutons formed in two stages (McDonald, 2001). The plutons of the first stage are dominated by biotite granodiorite to leucomonzogranites, the plutons of the second stage consist of mica rich monzogranite to coarse and fine grained leucogranite. Sediments weathered from these primarily granitic rocks were deposited into basins throughout eastern Canada to form members of the Horton Group (Bell, 1929). During the late Devonian, the lowermost portion of the Horton Bluff Formation formed in an alluvial environment (Martel and Gibling, 1995). Subsequent regional flooding resulted in a change in depositional environment from dominantly alluvial to dominantly lacustrine environments. The sediments deposited in these lakes form the middle and top of the Horton Bluff Formation (Martel and Gibling, 1995, Bell, 1929).

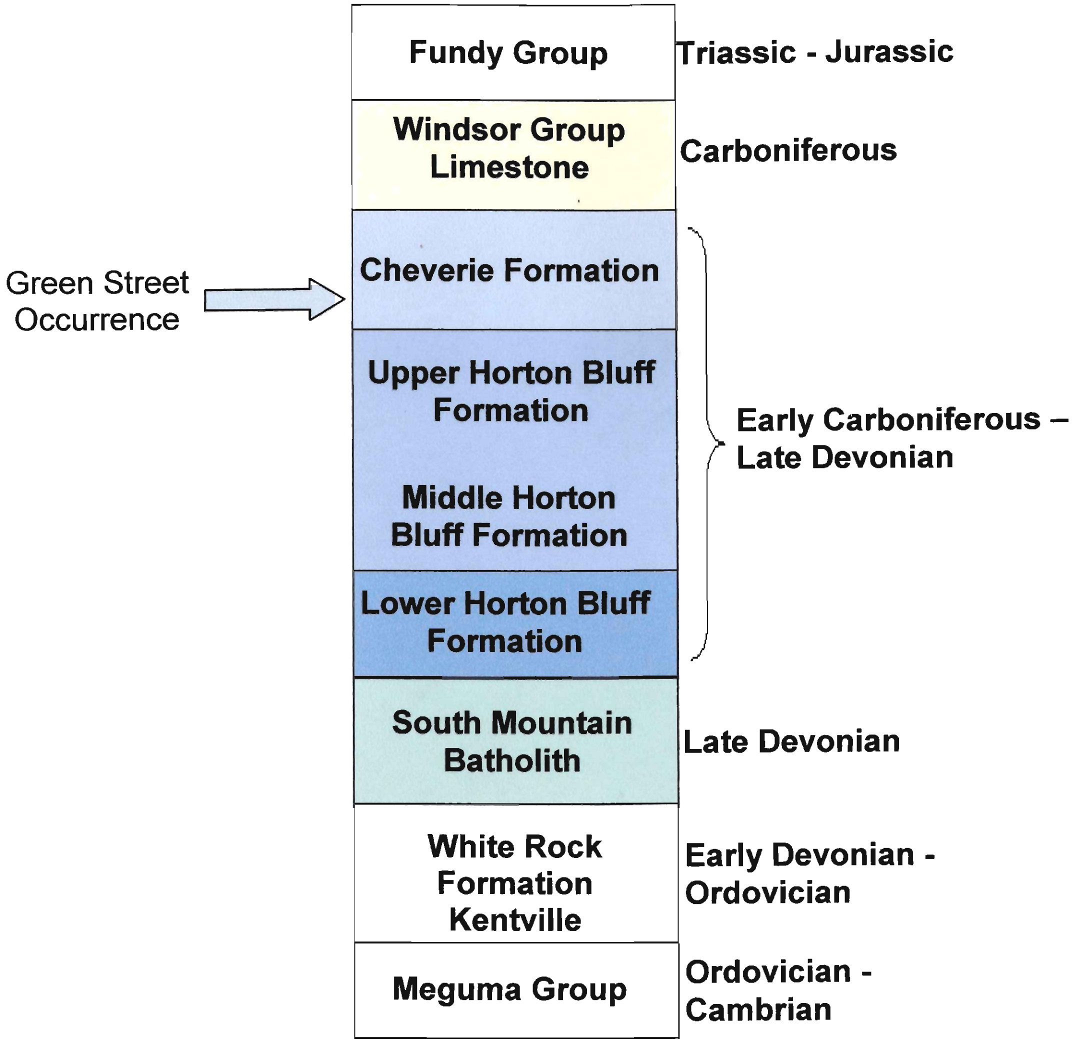


Figure 2.1 – Simplified stratigraphic column of the study area, near Windsor, NS (Modified from Ryan, 1998)

Figure 2.2 – Southwest facing exposed outcrop of Cheverie Formation near Windsor, NS, arrow indicates 3m (photo by A. Parsons, September, 2006)

There is an unconformity between the Horton Bluff Formation and the overlying Cheverie Formation (Figure 2.1), during which time there was some erosion of Horton Bluff sediments. The presence of similar spore assemblages in both Formations suggest that very little time passed between periods of deposition (Martel and Gibling, 1995). A return to an alluvial environment marks the base of the Cheverie Formation, as material derived from the South Mountain Batholith continued to be deposited (Martel and Gibling, 1995).

Marine limestone of the Windsor Group was deposited on top of the Horton Group during the Carboniferous. Subsequent deformation including folding, faulting, and uplift, resulted in widespread erosion of the strata (Martel and Gibling, 1995, Van de Poll et al., 1995).

The recently exposed outcrops of Cheverie Formation sandstones show evidence of modern day weathering, as can be seen in the exposed outcrops in the Windsor area (Figure 2.2). Storm and flooding events weather and erode sediment and it is deposited throughout stream beds and ditches in the area.

2.4 Uranium Occurrences in Nova Scotia

To understand the problems associated with uranium in the environment, it is important to understand some of the general properties of uranium. The following sections deal with both the chemical and physical properties of uranium, as well as the significance of uranium occurrences in Nova Scotia.

2.4.1 Physical and Chemical Properties of Uranium

Uranium is a naturally occurring radioactive element. It has an atomic number of 92, and occurs in three different isotopes, ^{238}U, ^{235}U, and ^{234}U. These isotopes are found in different proportions, with an average distribution of 99.3%, 0.71% and 0.006%, respectively. The radioactive nature of uranium isotopes is the result of decay as the unstable isotopes move toward more stable configurations through a series of radioactive daughter products. ^{206}Pb is a stable isotope resulting from the decay of ^{238}U.

The solubility of an element is a property that greatly affects its chemical mobility. In minerals, uranium is most commonly found as U^{4+}, and is insoluble. However, when U^{4+} is oxidized to U^{6+}, it becomes soluble and will form soluble complexes with carbonate, phosphate, sulphate, fluoride, and silicate ions (Figure 2.3). U^{6+} will also be adsorbed onto organic compounds (Ivanovich and Harmon, 1992). The chemical mobility of uranium is therefore largely determined by the environment in which it is found, and the amount of weathering that occurs in the environment.

2.4.2 Significance in Nova Scotia

In Nova Scotia, the presence of naturally occurring elevated uranium has been known since the beginning of the 20th century (O'Reilly, 1982). There has been much interest in uranium, due to both its potential as a resource, and the potential negative health effects caused by uranium and its daughter products in rocks, soil, water, and air. There have been a number of surveys carried out

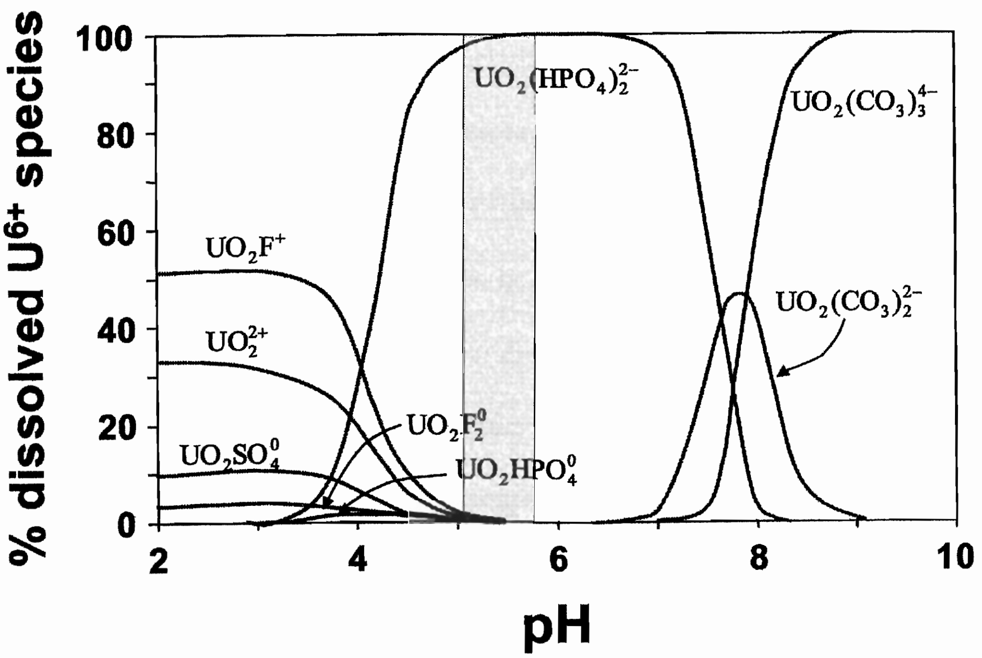


Figure 2.3 – Distribution of uranyl complexes (uranyl, UO_2^{2+} is the oxidized form of dissolved uranium) as a function of pH for some typical ligand concentrations in groundwater (Langmuir, 1978). Shading indicates range of pH values for water samples in this thesis.

by various agencies and organizations in the province, to determine background levels of Uranium as well as target areas for exploration (O'Reilly, 1982). From these surveys, occurrences were mapped extensively, and a summary of these occurrences is given in Table 2-1.

Uranium levels are commonly high in granites as uranium is preferentially incorporated into crustal rocks because of the incompatible nature of the element. Those rock types that are last to crystallize from a melt, monzogranites, leuco-monzogranites, granites, alaskites, and pegmatites, are commonly higher in incompatible elements than those that crystallize earlier. For this reason, there is often a higher amount of uranium in these rocks. Sedimentary rocks, such as those of the Cheverie Formation, which derive their sediments from these uranium enriched granites, will also have elevated Uranium levels.

There have been several maps published regarding the distribution of uranium occurrences in Nova Scotia. A map by the Nova Scotia Department of Health (n.d.), shows areas with potential for elevated uranium in groundwater (Fig. 2.4). This map is based solely on occurrences of granitic and clastic sedimentary rocks, and other minor occurrences, and does not actually provide specific data on elevated uranium levels. Figure 2.5 is more comprehensive and shows various locations with elevated uranium, as well as the rock types in which they occur (after O'Reilly, 1982).

Table 2.1 – Types and features of uranium deposits in Nova Scotia (Adapted from G. O'Reilly, 1982)

Deposit Type	Elemental Association	Geological Features
Magmatic	U, Sn, W, Mo, Be, and Cu	Radioactive mineral occurs in pegmatite dykes and veins in intrusive bodies of alaskite and leuco-monzogranite.
Vein	U, Th, and Cu	Veins and shear zones altered and mineralized by hot hydrothermal fluids in the interval 280-340 million years ago. The hydrothermal fluids are at least in past derived from specialized granitic intrusions enriched in incompatible elements. Occur predominantly in southern mainland granitic rocks.
Sandstone	U and Cu	Low grade copper and uranium mineralization associated with sandstone layers containing coal fragments and fossil plant detritus in the large Carboniferous sedimentary basins of northern Nova Scotia.
Basal Windsor Group	U	Uranium bearing minerals occur disseminated in the solution-collapse Pembroke breccia and underlying Macumber Formation limestone at the base of the Carboniferous Windsor Group.
Black Shales	U and Ba	Black shale horizons containing anomalous uranium content occur within the Cambrian sedimentary rocks of southeastern Cape Breton Island.
Devonian Volcanics	U and Th	Uranium and thorium bearing minerals in veins and shear zones occur in hydrothermally altered volcanic rocks of the Lower Devonian Byers Brook Formation. The mineralization is confined to rhyolitic volcanic units which have been intruded by granitic rocks.

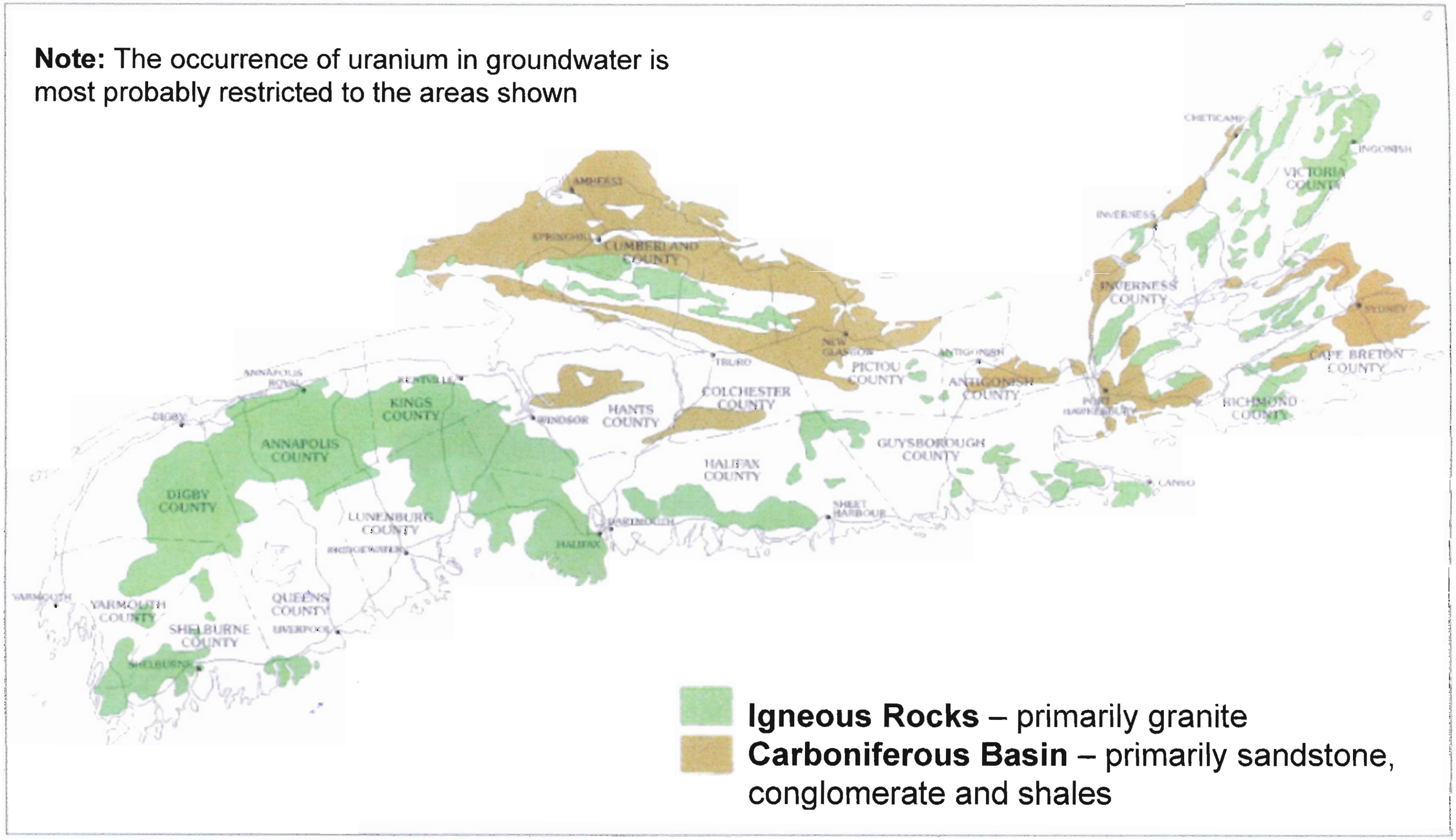


Figure 2.4 – Naturally Occurring Uranium in Groundwater in Nova Scotia – Showing the types of Bedrock where wells are most likely to contain Uranium (Nova Scotia Department of Health, n.d.)

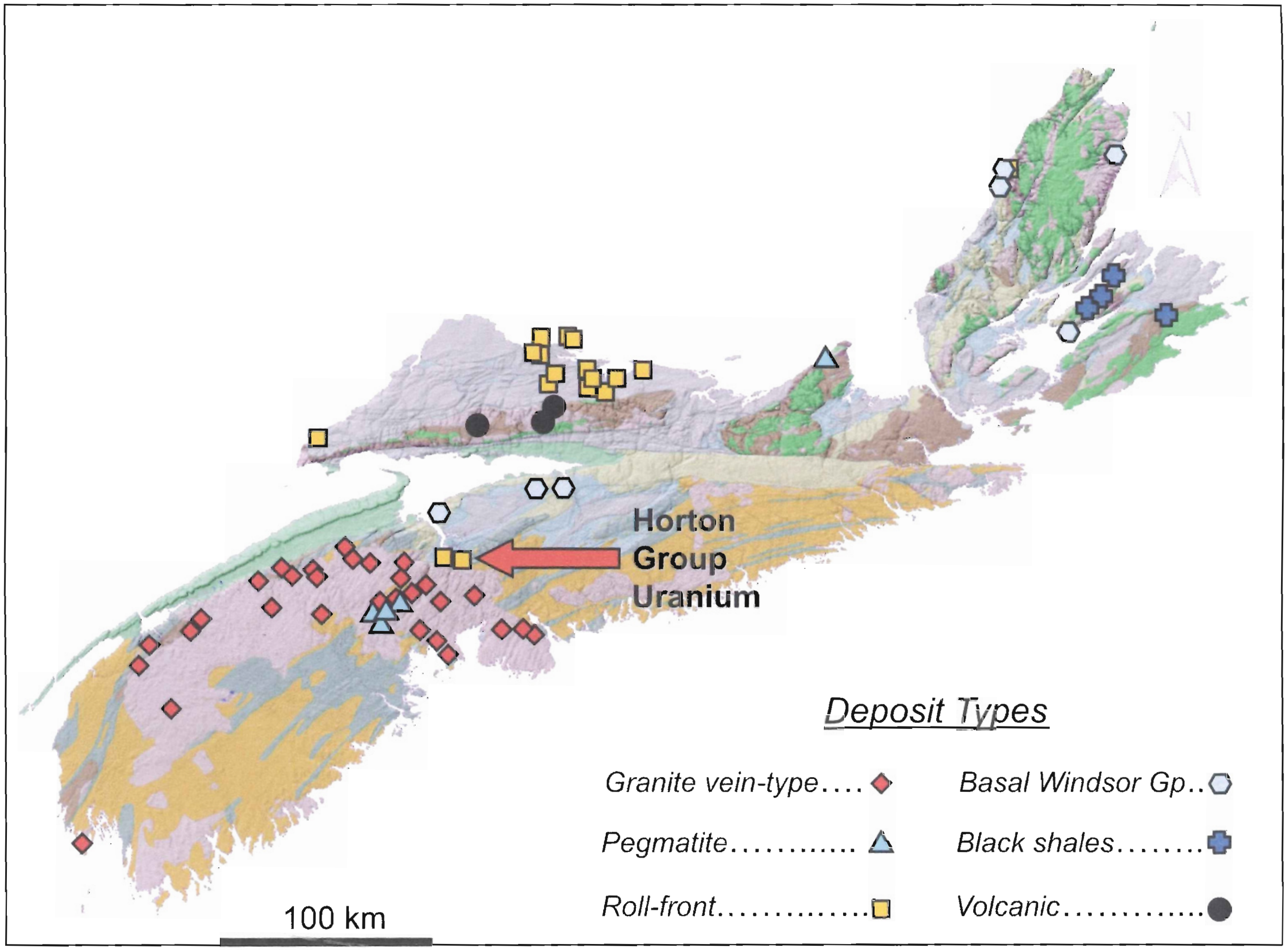


Figure 2.5 – Uranium occurrences in Nova Scotia (Modified from O'Reilly, 2005)

2.4.3 Summary

In order to evaluate the nature of the uranium occurrence at Green Street, petrographic study, microprobe analysis of selected minerals, whole rock geochemistry, and analysis of the waters produced under weathering experiments was undertaken. Details of the methods used are outlined in the following chapter.

3.0 Methods: Field Sampling and Experiments

3.1 Introduction

This chapter provides a summary of the methods that were used to investigate the potential for mobility of uranium in the Horton Group Sandstones. Field sampling was used in conjunction with laboratory work to gather evidence for uranium migration from various sources. Samples collected in the field were used for several analytical procedures, including petrographic and microprobe analysis, whole rock geochemical analysis, and artificial weathering and water sample analysis.

3.2 Field Work

Rock samples were collected from the Horton Group near Windsor, Nova Scotia (Figure 1.1) on September 28th, 2006. Descriptions of the samples are listed in Table 3-1. All observations made while at the site are recorded in chapter 4. Rocks sampled for analysis were selected based on elevated uranium counts from a McPhar model TV-A1 handheld spectrometer.

Samples were collected from sections where a higher response was obtained from the handheld spectrometer, indicating higher uranium values. Values read from the spectrometer for rocks with elevated uranium ranged from 70,000->100,000 counts per minute (cpm), typical background readings in the area range from 4,000 to 6,000 cpm. As the rocks of the outcrop were damp, the samples were later dried and any noticeable organic debris was removed by hand.

Table 3.1 – Sample Descriptions, see Figure 1.1 for sampling locations.

Sample #	Lithology	Description
AP06-01	Highly weathered sandstone, collected from Location 1	Outwash from cliff face, contains some organic matter. Grains are rounded to sub-rounded with a maximum grain size of 4mm. Dry sample is light reddish brown in colour, with some slightly consolidated black material and minor organics
AP06-02	Siltstone, collected from Location 1	Siltstone layer is located within the sandstone horizons. The layer is highly fractured and contains roots and moss. The siltstone is light to medium brown in colour with some orange staining on the outer surfaces.
AP06-03	Weathered sandstone, collected from Location 1	Sandstone makes up the majority of the cliff face in Location 1. The material is fairly homogeneous throughout, with the exception of the silt layers and some hematization. Grains are sub-rounded with a maximum size of 4mm. Grains range from brown to purple. The sandstone fractures very easily.
AP06-04	Siltstone, collected from Location 2	This siltstone band is located in a bank of consolidated sandstone horizons. The layer is slightly less fractured than the siltstone in Location 1 and contains less organic material. The layer is light brown to dark brown with a small amount of orange staining along fracture surfaces.
AP06-05	Weathered sandstone, collected from Location 2	Sandstone banks are located along the sides of the road from Location 1 to Location 2. Again, the sandstone from Location 2 is homogeneous throughout with the exception of the silt layers. Sandstones at Location 2 are less hematized. The grains are sub-rounded with a maximum grain size of 4mm. The sandstones are light reddish brown in colour and fracture very easily.
AP06-06	Unconsolidated sediment, collected from location 2	Outwash from banks, contains debris and organic matter. Grains are rounded to sub-rounded with a maximum grain size of 4mm. Sample is light to dark brown in colour. This sample is a mixture of sandstone and siltstone.
AP06-07	Fresh sandstone, collected from Location 2	This sample was collected from a sandstone bank by chipping the weathered surface to expose a fresh surface. This sample is lighter in colour than the other sandstone samples and does not contain any staining or hematization. This sample is also more consolidated than the other sandstones.

3.3 Petrographic and Microprobe Analysis

Based on observations of thin sections using standard petrographic techniques, a number of grains were selected for microprobe analysis. This analysis was done using a JEOL JXA-8200 WD/ED Combined Microanalyzer. Microprobe analysis focused on biotites and opaque minerals, based on previous work involving fission track analysis. This fission track work by O'Beirne-Ryan (2006) indicated that uranium within the Horton Group is largely concentrated within biotite grains (Figure 3.1), and in associated opaque minerals.

3.4 Geochemical Analysis

Whole rock geochemical analysis of major, trace, and rare earth elements were undertaken at ACME Laboratories (Vancouver) using XRF and ICP-MS techniques. Samples analyzed included two siltstones, two sandstones, one more highly weathered sandstone, and a mixed sample collected from loose debris at the base of the outcrop.

3.5 Water Sample Analysis

In order to determine whether any elements were leachable out of the Horton Group sedimentary rocks, an artificial weathering experiment was carried out, using the same samples as the whole rock geochemical analysis. 200g of gently-crushed rock (average diameter ~1cm) was placed in a conical flask with 400ml of natural "rainwater" (pH 5.06). The "rainwater" was collected following a snowfall on January 6th, 2007 ("rainwater" refers to the melted snow, and for simplicity will hereafter be referred to simply as rainwater). The water was stored in sealed jars and refrigerated until use. The samples were prepared in two batches: the first batch was agitated for a period of 48 hours, the second batch for 144 hours, both at a constant temperature of 25°C. The sediments were allowed to settle for approximately one hour before the water

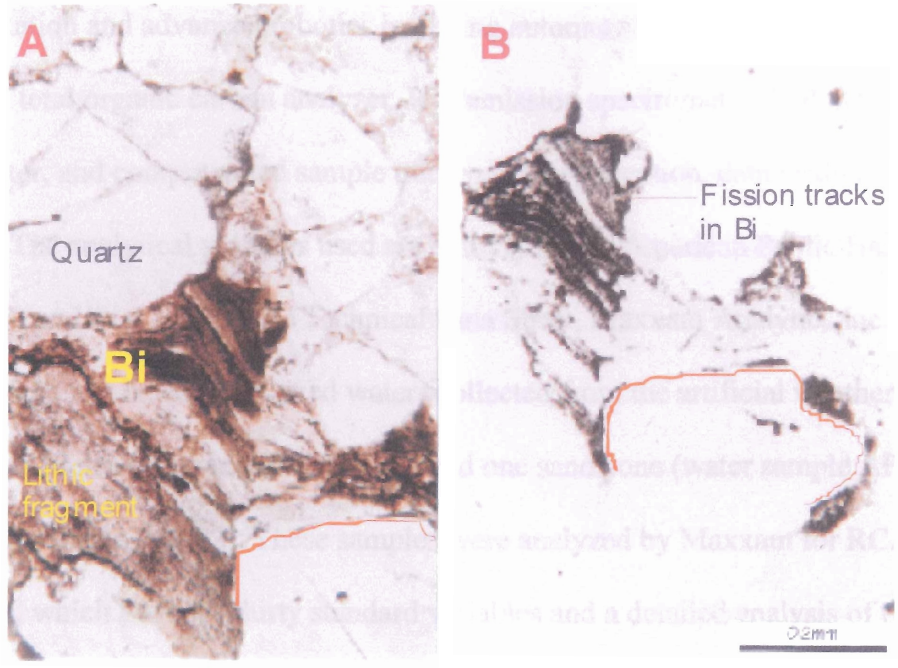


Figure 3.1 – Fission track analysis of a biotite grain from the Horton Group (from O'Beirne-Ryan, 2006). A shows a biotite grain in thin section. B shows uranium fission tracks are concentrated within the biotite grain, and along grain boundaries. These fission tracks are revealed as black areas within the slide.

was drained and immediately brought to Maxxam Analytics (Bedford) for analysis. Measures of pH, total dissolved solids, conductivity, and temperature were taken over the course of the agitation.

Analysis at Maxxam Analytics was performed using highly automated instrumentation and advanced robotics including automated colorimetric analyzers, automated total organic carbon analyzer, ICP/emission spectrometer, ICP/mass spectrometer, and computerized sample tracking, data collection, data evaluation and reporting. The analytical methods used are defined by the American Public Health Association and/or the US EPA (Technical Data Sheet, Maxxam Analytics Inc., 2005)

Samples in batch 1 analyzed waters collected from the artificial weathering of the two samples highest in uranium. This included one sandstone (water sample AP06-05) and one siltstone (AP06-02). These samples were analyzed by Maxxam for RCAp-MS parameters, which includes thirty standard variables and a detailed analysis of trace metal concentrations. Metals for this sample batch were analyzed as dissolved metals.

Samples in batch 2 analyzed waters from three remaining rock samples, one sandstone (water sample AP06-03), one siltstone (AP06-04), and the more highly weathered sample (AP06-01). These samples were analyzed only for total metals, as it was not feasible to do a full analysis on all samples.

Analysis was carried out on a sample of the natural rainwater used in the artificial weathering experiments (water sample AP06-RW). Like the samples from batch 1, this sample was analyzed for RCAp-MS parameters, but the metals were analyzed as total metals, rather than just the dissolved species. The results of the all water analyses are presented in Chapter 4.

4.0 Results

Data for this thesis were collected from a variety of sources, including observations from the field, microprobe analysis, whole rock geochemistry, and an analysis of water samples. The results for each are discussed in the following sections.

4.1 Field Observations

The outcrops studied in this thesis are located in the vicinity of Green Street, near Windsor, NS. The outcrops are accessible as road cuts along the sides of the road (Figure 1.1). The topography is predominantly gently rolling hills, resulting in differences in elevation over the study area, with Location 1 located down slope from Location 2 (Figure 1.1). The stream near Location 1 (Section 1.2) is located down slope from the outcrop.

There were several important observations made in and around the study area, the most important feature being the visible weathering of the outcrops. The cliff at Location 1 is weathering and eroding rapidly, as could be seen by the overhang of the overlying soil layers at the top of the cliff (Figure 2.2). There was a large amount of loose, weathered material at the base of the cliff and in surrounding ditches. Similar weathering could be seen in the smaller outcrops along the side of the road toward the small quarry (Figure 1.1). Weathering of the siltstone layers could also be established by the presence of extensive fracturing, oxidation on exposed surfaces, and the pile of debris at the base of the cliff.

A second important observation in the area is the proximity of the weathering outcrops to surface water. The closest source of surface water in the area is a stream that lies approximately 5m down slope from the outcrop at Location 1 (Figure 4.1). Water levels in this stream were low at the time of sampling (September, 2006) following an extensive period of little rainfall.

Figure 4.1 – Proximity of uranium-bearing outcrop to surface water. Red arrow indicates the stream, blue arrow indicates outcrop at Location 1. Distance between arrows is approximately 4m (photo by A. Parsons, September, 2006).

4.2 Mineralogical Data

4.2.1 Introduction and Sample Descriptions

Microprobe data focused on determining the compositions of biotite grains and opaque minerals. Several grains in thin sections from both sandstones and both siltstones were examined to establish a consistency in the data. Table 4.1 lists the compositional data from the examined points.

4.2.2 Sandstones

The sandstones are K-feldspar rich, with quartz, muscovite, and variably weathered biotite. The samples also contain 5-10% small opaque grains and accessory zircon and apatite. The amount of quartz in the samples ranges from 40-45%, quantities of K-feldspar range from 35-40%. There are minor amounts of muscovite/ illite (1-2%), and biotite (5-10%). The less weathered sample (AP06-07) contains some small lithic fragments and has a fine-grained matrix. The more weathered samples (AP06-03, AP06-05) have a higher proportion of matrix material than the less weathered sample, and also have a higher proportion of ragged opaque minerals. As might be expected, the feldspars in the more weathered samples are more weathered than those in the fresher sample.

Table 4.1 – Mineralogical data from microprobe analysis (mass percent)

Sample	Lithology	K_2O	TiO_2	Na_2O	Al_2O_3	FeO	CaO	Cr_2O_3	MgO	SiO_2	MnO	Total	Composition
AP06-07	Sandstone	1.52	0.55	0.05	35.78	2.76	0.00	0.00	0.84	46.68	0.01	88.20	kaolinite
AP06-07	Sandstone	0.09	0.00	0.00	38.38	0.28	0.00	0.00	0.00	47.33	0.00	86.08	kaolinite
AP06-05	Sandstone	2.29	0.69	0.05	35.55	4.62	0.01	0.00	1.14	47.10	0.07	91.53	kaolinite
AP06-05	Sandstone	0.59	0.05	0.03	37.09	0.81	0.03	0.00	0.37	46.00	0.00	84.97	kaolinite
AP06-03	Sandstone	0.99	0.00	0.04	37.28	0.24	0.01	0.00	0.06	44.68	0.00	83.30	kaolinite
AP06-03	Sandstone	0.51	0.00	0.04	39.67	0.24	0.01	0.00	0.03	47.28	0.00	87.79	kaolinite
AP06-03	Sandstone	0.58	0.13	0.01	38.26	1.02	0.00	0.00	0.24	47.69	0.03	87.98	kaolinite
AP06-07	Sandstone	6.25	2.88	0.07	25.22	14.49	0.05	0.01	3.99	40.91	0.20	94.08	biotite
AP06-07	Sandstone	8.10	3.07	0.09	21.01	17.17	0.04	0.04	4.00	38.72	0.23	92.47	biotite
AP06-05	Sandstone	8.37	2.92	0.12	22.49	16.60	0.04	0.05	5.00	38.38	0.46	94.44	biotite
AP06-05	Sandstone	7.80	3.65	0.11	20.34	15.37	0.07	0.03	5.08	40.53	0.21	93.19	biotite
AP06-05	Sandstone	8.49	3.12	0.09	20.74	16.36	0.07	0.04	5.35	39.54	0.30	94.10	biotite
AP06-04	Siltstone	8.71	3.44	0.03	20.35	16.53	0.03	0.09	4.25	37.20	0.34	90.97	biotite
AP06-04	Siltstone	7.23	2.66	0.08	19.88	19.24	0.06	0.11	3.07	36.07	0.25	88.65	biotite
AP06-05	Sandstone	1.87	0.10	0.04	15.01	3.11	0.05	0.00	0.43	62.34	0.00	82.95	vermiculite
AP06-05	Sandstone	4.64	1.84	0.05	29.36	9.54	0.02	0.00	3.68	42.09	0.15	91.36	vermiculite
AP06-03	Sandstone	6.31	0.00	0.13	32.12	4.42	0.01	0.00	2.06	47.28	0.09	92.43	vermiculite
AP06-03	Sandstone	6.87	3.29	0.09	22.77	13.49	0.06	0.10	2.85	40.19	0.23	89.94	vermiculite
AP06-03	Sandstone	5.21	2.33	0.06	27.00	10.00	0.04	0.06	2.05	41.89	0.15	88.78	vermiculite
AP06-03	Sandstone	5.85	1.73	0.03	27.52	9.04	0.04	0.08	1.78	44.95	0.14	91.17	vermiculite
AP06-04	Siltstone	9.49	0.03	0.30	34.58	2.10	0.00	0.02	0.59	46.28	0.03	93.41	muscovite
AP06-04	Siltstone	10.11	0.15	0.31	35.36	1.73	0.01	0.03	0.61	47.37	0.04	95.72	muscovite
AP06-04	Siltstone	9.10	0.05	0.39	35.06	2.34	0.00	0.01	0.39	46.48	0.05	93.87	muscovite
AP06-04	Siltstone	9.57	0.75	0.31	33.49	1.69	0.00	0.02	1.05	47.44	0.09	94.41	muscovite
AP06-02	Siltstone	7.45	0.70	0.26	38.00	0.90	0.00	0.01	0.27	48.29	0.00	95.89	muscovite
AP06-02	Siltstone	8.89	0.01	0.38	37.51	1.25	0.00	0.02	0.42	46.59	0.03	95.09	muscovite
AP06-02	Siltstone	6.65	0.34	0.27	37.70	1.01	0.00	0.00	0.19	46.66	0.01	92.84	muscovite
AP06-03	Sandstone	13.61	0.00	1.17	19.04	0.05	0.05	0.00	0.00	65.09	0.00	99.02	K feldspar
AP06-03	Sandstone	14.98	0.00	1.13	19.42	0.05	0.01	0.00	0.00	63.32	0.00	98.90	K feldspar
AP06-03	Sandstone	15.61	0.00	0.76	19.07	0.08	0.03	0.05	0.00	64.12	0.02	99.73	K feldspar
AP06-03	Sandstone	15.64	0.01	0.69	19.29	0.06	0.02	0.06	0.00	62.90	0.02	98.68	K feldspar
AP06-03	Sandstone	14.81	0.01	1.17	18.77	0.13	0.02	0.04	0.00	63.67	0.02	98.64	K feldspar

AP06-03	Sandstone	14.77	0.02	0.37	18.57	0.26	0.01	0.01	0.00	63.16	0.03	97.19	K feldspar
AP06-04	Siltstone	16.14	0.01	0.21	18.52	0.30	0.01	0.06	0.00	63.97	0.02	99.23	K feldspar
AP06-04	Siltstone	14.38	0.00	1.43	18.74	0.16	0.03	0.03	0.00	63.55	0.02	98.34	K feldspar
AP06-05	Sandstone	0.20	0.32	0.06	0.51	76.18	0.12	0.17	0.19	1.69	0.12	79.56	Fe oxide
AP06-05	Sandstone	0.08	0.21	0.04	0.26	77.82	0.10	0.18	0.02	1.78	0.12	80.62	Fe oxide
AP06-03	Sandstone	0.22	0.32	0.06	3.41	73.94	0.15	0.19	0.04	8.16	0.11	86.60	Fe oxide
AP06-03	Sandstone	0.11	0.87	0.08	2.16	78.67	0.11	0.21	0.02	6.60	0.13	88.97	Fe oxide
AP06-03	Sandstone	0.34	0.51	0.05	2.41	79.18	0.10	0.18	0.03	7.52	0.12	90.45	Fe oxide
AP06-03	Sandstone	0.26	0.41	0.01	3.37	76.28	0.09	0.26	0.00	6.49	0.13	87.30	Fe oxide
AP06-04	Siltstone	0.05	0.12	0.00	0.00	76.15	0.06	0.20	0.00	0.46	0.13	77.18	Fe oxide
AP06-04	Siltstone	0.05	0.15	0.00	0.00	75.71	0.06	0.20	0.00	0.94	0.13	77.25	Fe oxide
AP06-04	Siltstone	0.06	0.20	0.00	0.00	77.42	0.05	0.25	0.00	0.37	0.14	78.49	Fe oxide
AP06-04	Siltstone	0.07	0.16	0.00	0.00	74.46	0.06	0.22	0.00	1.10	0.42	76.49	Fe oxide
AP06-02	Siltstone	0.12	98.46	0.01	0.00	0.63	0.05	0.19	0.00	0.13	0.09	99.68	Ti oxide
AP06-02	Siltstone	0.24	98.26	0.00	0.00	0.72	0.03	0.17	0.00	0.11	0.08	99.61	Ti oxide

4.2.3 Siltstones

The siltstones are fine grained with K-feldspar, quartz, muscovite, variably weathered biotite, and opaque mineral grains in a clay-rich matrix. The main difference between the two siltstone samples is a lamination as defined by the biotites in AP06-04. This, along with the presence of chlorite and a higher proportion of clay, suggests that the two siltstone samples are from different silty layers and are not two samples from the same horizon.

4.2.4 Biotite and Opaque Minerals

The most important information that was gathered from the microprobe analysis was the observed pattern of biotite weathering. Compositions obtained from many grains indicated the presence of biotite, kaolinite, and an intermediate composition that will be referred to as vermiculite. A typical grain with traces of all three compositions can bee seen in Figure 4.2. The composition of the parent biotite is within the range of biotite compositions for fresh biotite in the monzogranite of the South Mountain Batholith (O'Beirne-Ryan, 2006).

The opaque grains show a variety of compositions. Iron oxides were common in all samples (Figure 4.3), and small amounts of titanium oxide were also found in a siltstone layer from Location 1 (AP06-02) (Figure 4.4).

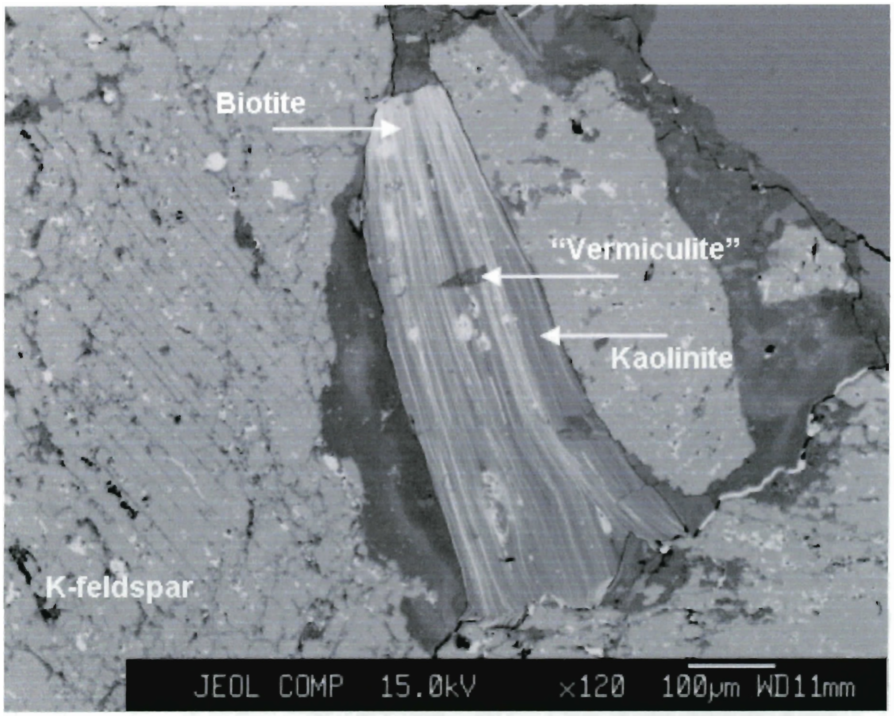


Figure 4.2 - Sandstone from Location 2 showing relationship of biotite to its weathering products

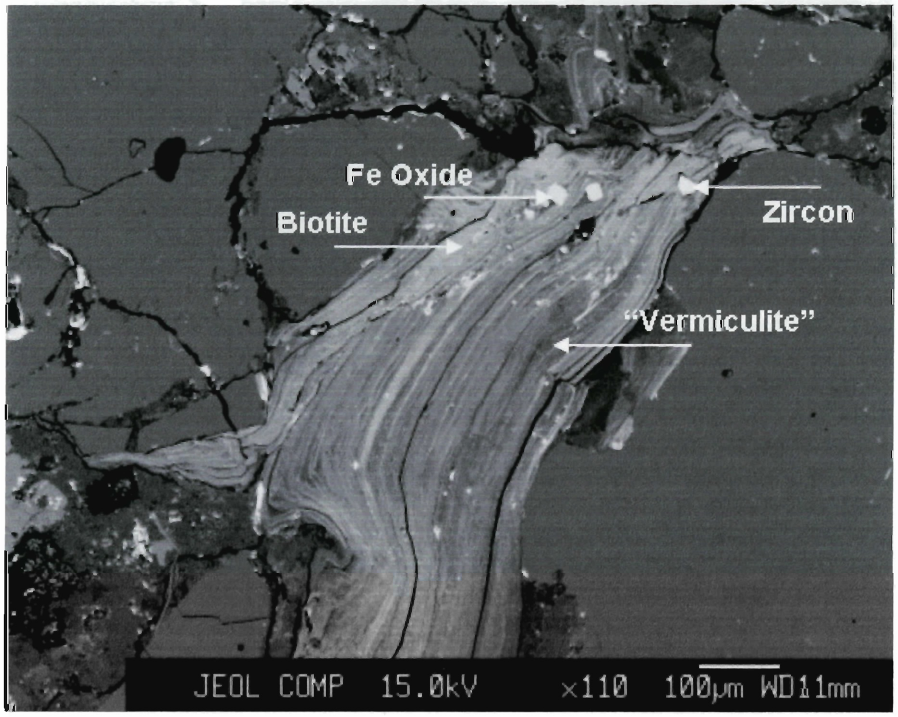


Figure 4.3 – Sandstone from location 2 showing relationship of accessory minerals to biotite grains

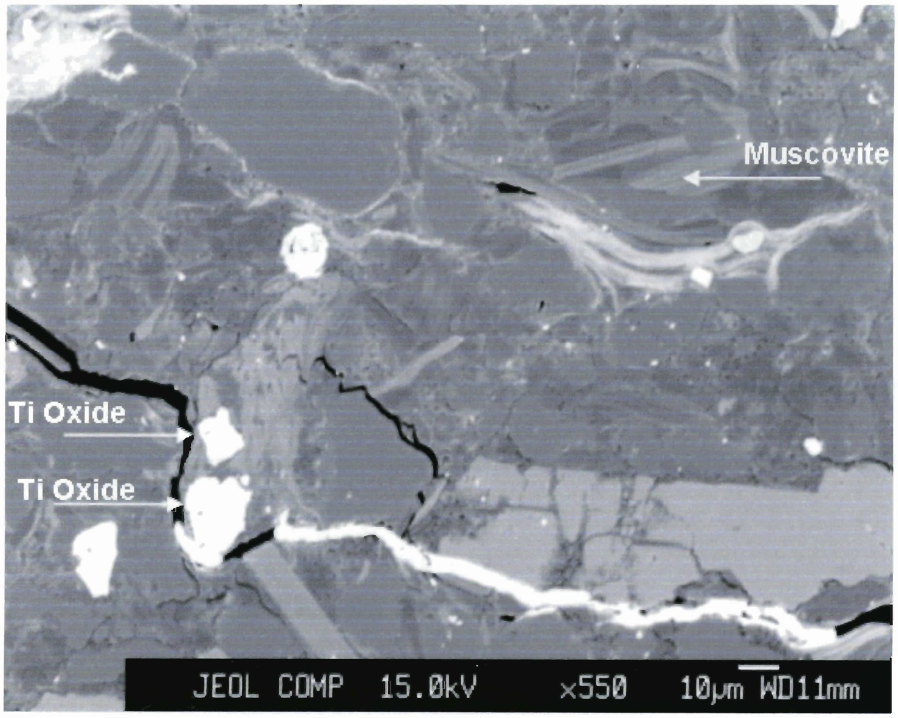


Figure 4.4 – Siltstone from location 1, contains titanium oxides

4.3 Whole Rock Geochemistry

Geochemical analysis was carried out on samples of two sandstones (one from each location), two siltstones (one from each location), the loose, weathered debris from the base of the sandstone at location 1 (Figure 2.2), and a sample of mixed, loose material from location 2. A full suite of data was obtained, including major elements (Table 4.2), trace elements (Table 4.3), and rare earth elements (Table 4.4). Results for major element analysis and trace element analysis are discussed in the following sections. Results for rare earth elements will not be discussed further in this thesis.

Rock classification of samples was done based on whole rock geochemical results, by plotting log(Fe_2O_3/K_2O) and log(SiO_2/Al_2O_3) (Figure 4.5).

Using Minpet software, several graphs were generated from selected geochemical data. The graphs were used to find correlations between major elements as well as between uranium and other trace elements. Correlations were calculated using R^2 values.

4.3.1 Major Element Results

In the plots for major elements (Figure 4.6), strong correlations between Al_2O_3 and SiO_2 (R^2=0.9802), TiO_2 and SiO_2 (R^2=0.9560), and MgO and SiO_2 (R^2=0.9707) are evident. Not all major elements showed a strong correlation with silica (Fe_2O_3 - R^2=0.2167). A ternary plot (Figure 4.7) was produced using mol proportions of Al_2O_3 vs. K_2O vs. CaO+Na_2O_3 (after Nesbitt and Young, 1989). The plot shows that the siltstones were more aluminous than the sandstones. The sandstones plot to the right of the Nesbitt and Young weathering trend, showing slight relative K_2O enrichment.

Table 4.2 – Whole rock geochemical results – Major elements

Element		SiO_2	TiO_2	Al_2O_3	Fe_2O_3	MnO	MgO	CaO	Na_2O	K_2O	P_2O_5	LOI
Method		XRF	XRF	XRF	XRF	XRF	XRF	XRF	XRF	XRF	XRF	XRF
Sample	Lithology	%	%	%	tot. %	%	%	%	%	%	%	%
AP06-01	Sand	83.49	0.08	8.90	0.98	0.05	0.11	0.03	0.16	3.70	0.07	2.30
AP06-02	Siltstone	59.93	1.00	22.63	3.16	0.02	0.71	0.04	0.18	3.74	0.09	8.40
AP06-03	Sandstone	77.36	0.08	10.58	4.40	0.02	0.14	0.04	0.16	3.84	0.09	3.30
AP06-04	Siltstone	60.74	0.90	22.33	3.02	0.03	0.68	0.02	0.16	3.77	0.10	8.00
AP06-05	Sandstone	78.75	0.17	12.06	1.13	0.04	0.16	0.02	0.17	3.58	0.07	3.70
AP06-06	Sediment	67.21	0.59	17.56	2.52	0.03	0.46	0.04	0.16	3.57	0.11	7.70

Table 4.3 – Whole rock geochemical results – Trace elements

Element		Ba	Rb	Sr	Y	Zr	Nb	Th	Pb	Ga	Zn	Cu
Method		XRF	ICP-MS	XRF	ICP-MS	ICP-MS	ICP-MS	ICP-MS	ICP-MS	ICP-MS	ICP-MS	ICP-MS
Sample	Lithology	ppm	ppm	ppm	ppm	ppm	ppm	ppm	ppm	ppm	ppm	ppm
AP06-01	Sand	228.0	83.0	39.0	4.4	23.3	2.4	2.2	17.4	9.1	6.8	13.9
AP06-02	Siltstone	286.0	150.5	88.0	33.5	181.3	22.1	13.0	88.1	45.7	41.0	95.5
AP06-03	Sandstone	245.0	84.7	74.0	74.5	24.5	2.6	2.5	27.1	11.2	6.3	30.5
AP06-04	Siltstone	223.0	135.7	198.0	11.4	177.8	19.6	16.2	38.7	34.9	56.3	51.5
AP06-05	Sandstone	214.0	98.0	62.0	7.3	44.8	4.4	5.3	43.1	13.4	14.6	31.2
AP06-06	Sediment	219.0	142.1	123.0	10.8	123.1	14.0	13.4	33.7	27.5	30.4	70.3

Element		Ni	V	Cr	Hf	Cs	Sc	Ta	Co	Be	U
Method		XRF	ICP-MS	ICP-MS	ICP-MS	ICP-MS	XRF	ICP-MS	ICP-MS	ICP-MS	ICP-MS
Sample	Lithology	ppm	ppm	ppm	ppm	ppm	ppm	ppm	ppm	ppm	ppm
AP06-01	Sand	7.0	26.0	7.0	0.9	5.5	2.0	0.5	2.7	1.0	6.3
AP06-02	Siltstone	5.0	159.0	43.0	6.2	45.9	10.0	3.0	8.3	29.0	212.1
AP06-03	Sandstone	<5	34.0	7.0	0.9	6.2	2.0	0.5	21.1	3.0	39.7
AP06-04	Siltstone	12.0	106.0	27.0	6.4	41.5	10.0	2.9	12.1	7.0	56.2
AP06-05	Sandstone	<5	53.0	6.0	1.6	9.6	4.0	0.9	31.9	4.0	109.2
AP06-06	Sediment	11.0	88.0	22.0	4.6	30.9	8.0	2.2	18.8	5.0	52.3

Table 4.4 – Whole rock geochemical results – Rare earth elements

Element		La	Ce	Pr	Nd	Sm	Eu	Gd	Tb	Dy
Method		ICP-MS	ICP-MS	ICP-MS	ICP-MS	ICP-MS	ICP-MS	ICP-MS	ICP-MS	ICP-MS
Sample	Lithology	ppm	ppm	ppm	ppm	ppm	ppm	ppm	ppm	ppm
AP06-01	Sand	5.2	11.0	1.2	5.4	1.2	0.3	1.2	0.2	1.0
AP06-02	Siltstone	12.0	25.0	2.7	11.3	3.5	1.1	5.1	1.2	7.8
AP06-03	Sandstone	8.2	16.3	1.8	7.3	2.4	1.0	7.5	1.7	12.3
AP06-04	Siltstone	31.0	67.4	7.1	29.6	5.5	0.4	3.5	0.5	2.4
AP06-05	Sandstone	11.7	26.6	2.4	9.2	1.7	0.3	1.5	0.2	1.4
AP06-06	Sediment	27.7	59.3	6.2	23.0	4.8	0.5	3.1	0.4	2.1

Element		Ho	Er	Tm	Yb	Lu
Method		ICP-MS	ICP-MS	ICP-MS	ICP-MS	ICP-MS
Sample	Lithology	ppm	ppm	ppm	ppm	ppm
AP06-01	Sand	0.1	0.4	0.1	0.4	0.1
AP06-02	Siltstone	1.4	3.7	0.5	3.4	0.5
AP06-03	Sandstone	2.5	6.4	0.9	5.0	0.7
AP06-04	Siltstone	0.4	1.0	0.2	1.3	0.2
AP06-05	Sandstone	0.3	0.7	0.1	0.7	0.1
AP06-06	Sediment	0.4	1.0	0.2	1.1	0.2

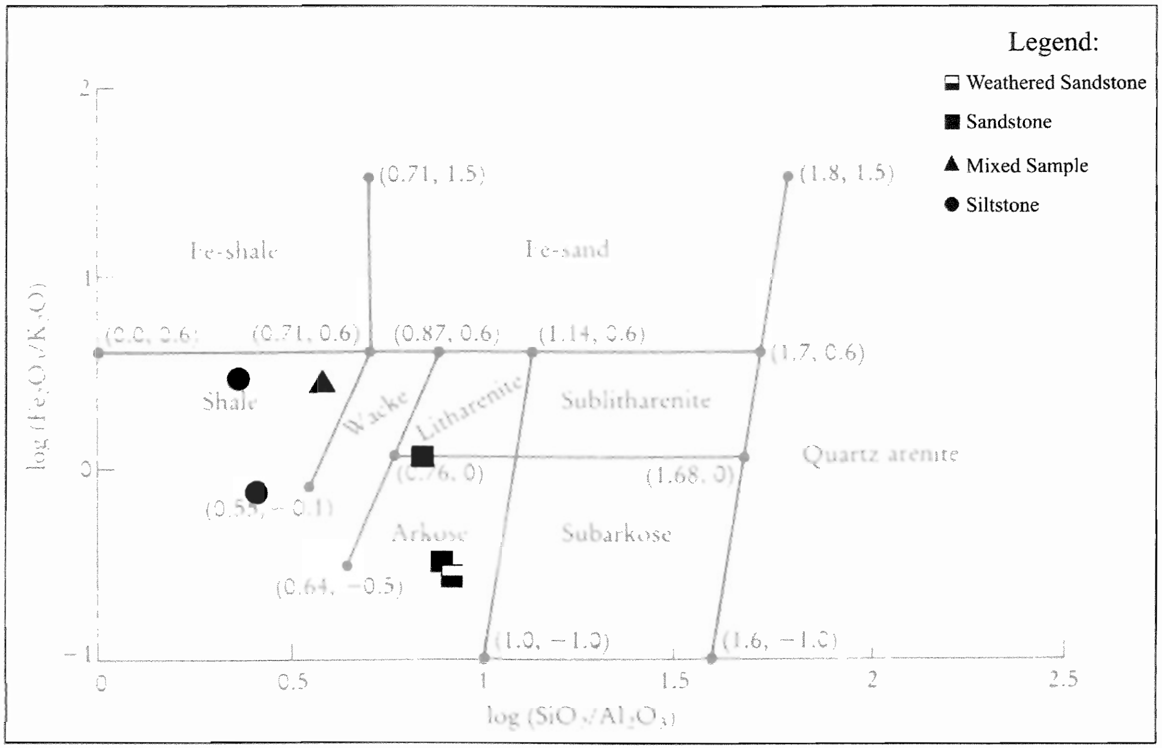
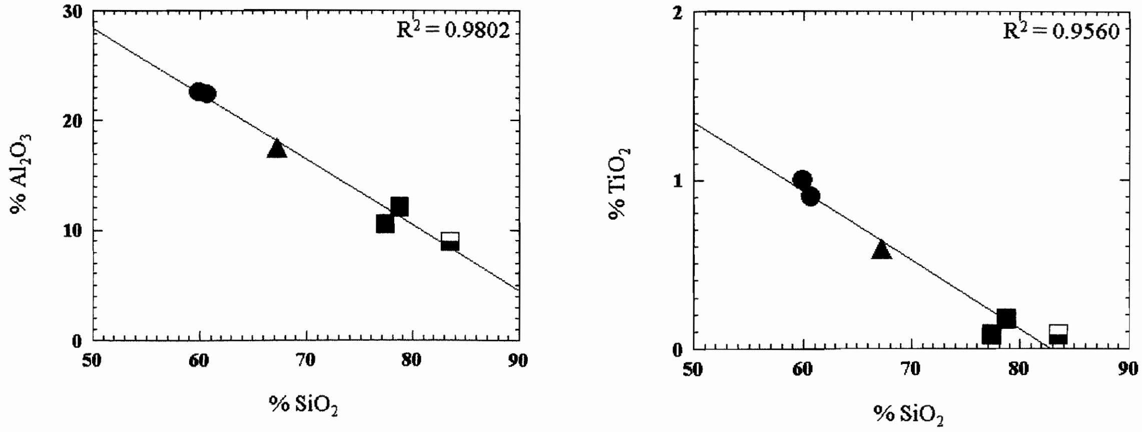
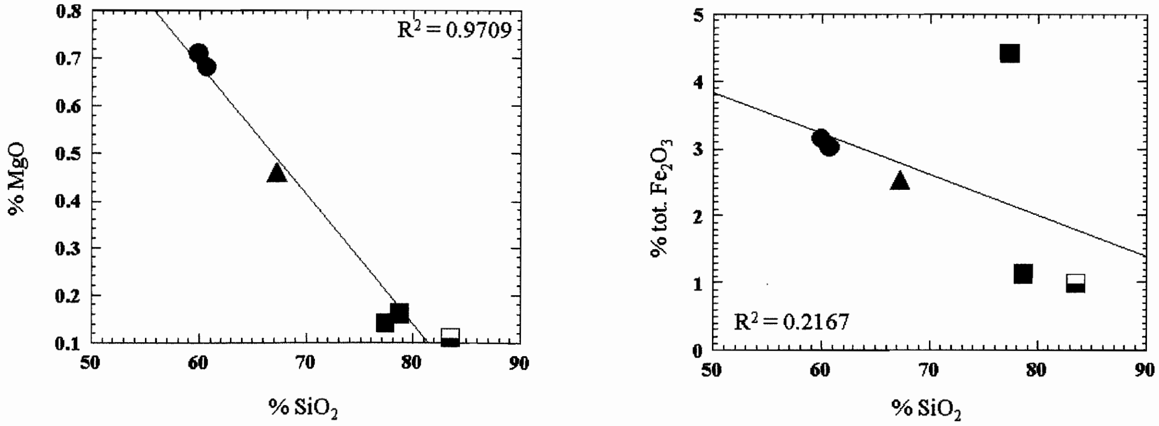


Figure 4.5 – Classification of terrigenous sandstones and shales using log(Fe_2O_3/K_2O) and log(SiO_2/Al_2O_3) (after Herron, 1988). The numbers shown in parenthesis are the plotting coordinates for the field boundaries.



4.5a. %SiO_2 vs. %Al_2O_3

4.5b. %SiO_2 vs. TiO_2



4.5c. %SiO_2 vs. %MgO

4.5d. %SiO_2 vs. %Fe_2O_3 (tot.)

Legend:

- ⬓ Weathered Sandstone
- ■ Sandstone
- ▲ Mixed Sample
- ● Siltstone

Figure 4.6 – Selected Major Element Plots

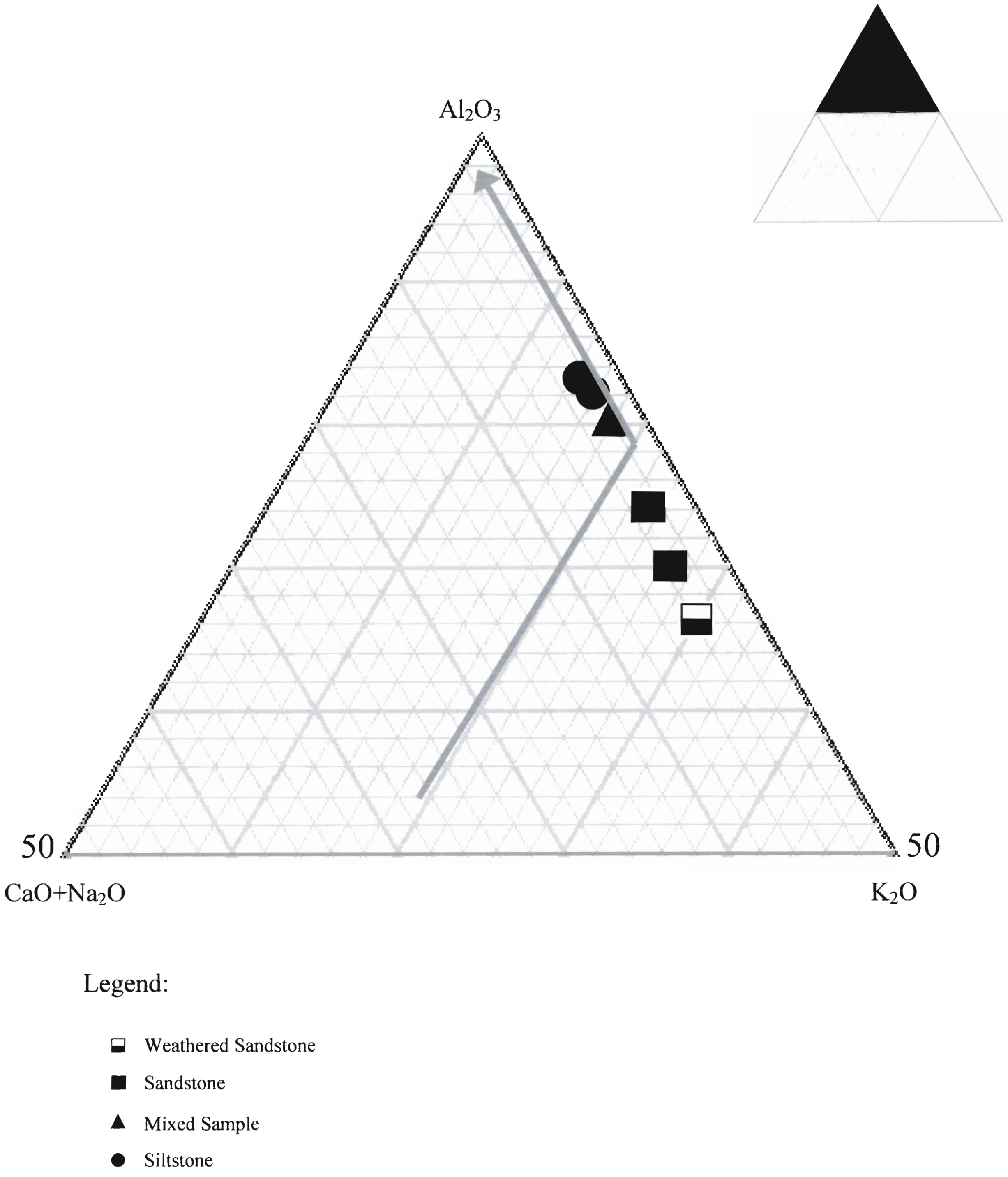


Figure 4.7 – Ternary Plot (values in molar proportions). Grey arrow indicates granite weathering trend (Nesbitt and Young, 1989).

4.3.2 Trace Element Results

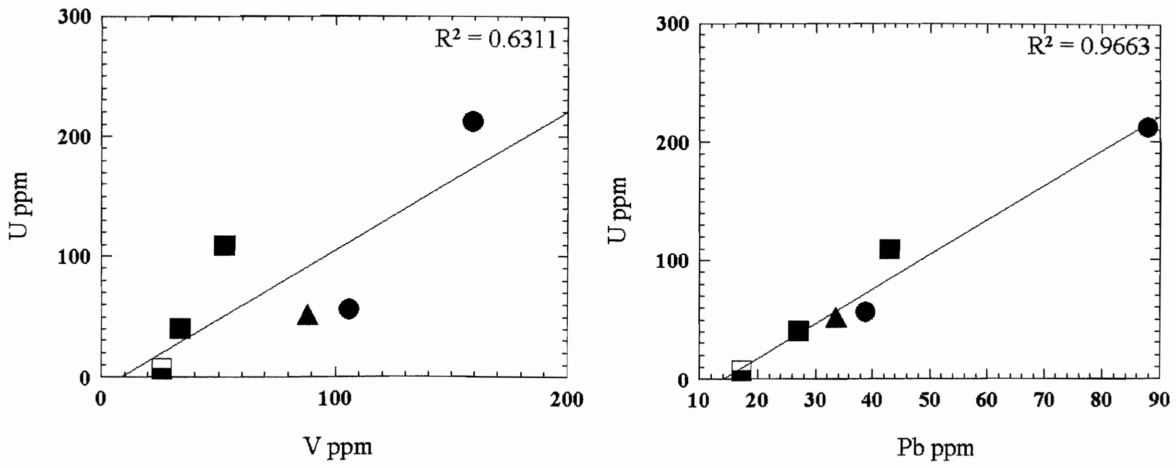
Plots for trace elements were generated showing concentrations of various elements against uranium (Figure 4.8). The strongest correlation (R^2=0.9663) was seen between uranium and lead. Uranium also had relatively strong correlations with vanadium (R^2=0.6311), and copper (R^2=0.5723). Uranium had little or no correlation with thorium or zirconium, or with iron or titanium.

4.3.3 Uranium Data

The geochemical data indicates that the highest uranium values are found in one siltstone (AP06-02) and one sandstone (AP06-05), with values of 212.1ppm and 109.2ppm respectively (Table 4.3). Uranium values for other samples range from 39.7ppm to 52.3ppm, with the exception of the more highly weathered sandstone sample (AP06-01) having only 6.3ppm (Table 4.3). All of these values exceed the background level for uranium in arkose sandstones in the area, which is typically 4-6ppm (R.J. Ryan, personal communication).

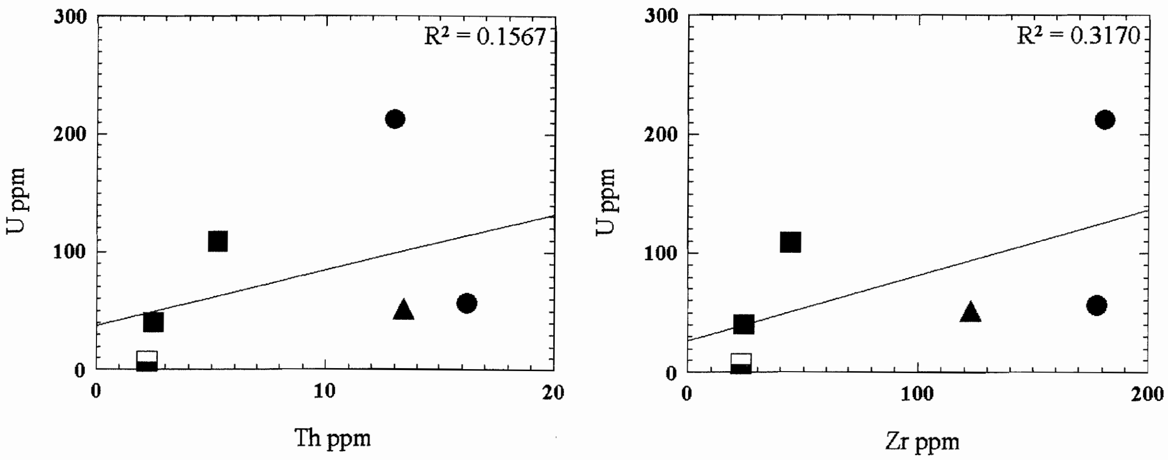
4.4 Water Geochemistry

Rock samples collected from the study area were artificially weathered (as described in section 3.5), after which time the water was collected and submitted for analysis at Maxxam Analytics Inc. The purpose of this weathering was to determine whether uranium would leach from the rocks during a typical rainstorm event, and if so, to determine the associated cations, anions, and other metals.



4.6a. U (ppm) vs. V (ppm)

4.6b. U (ppm) vs. Pb (ppm)



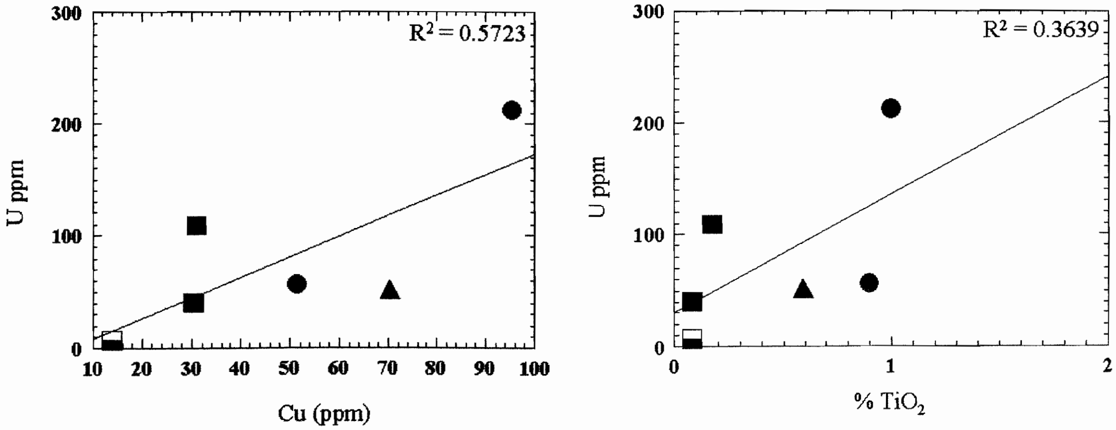
4.6c. U (ppm) vs. Th (ppm)

4.6d. U (ppm) vs. Zr (ppm)

Legend:

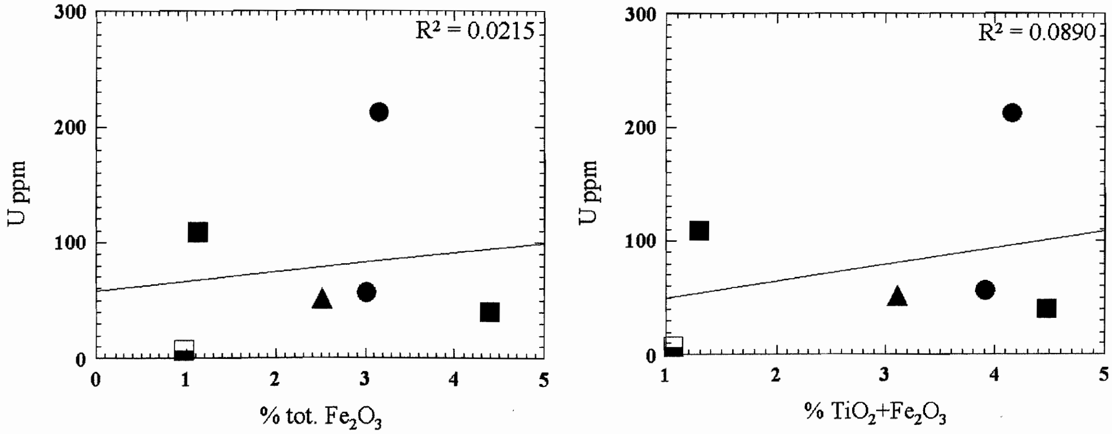
- ⬓ Weathered Sandstone
- ■ Sandstone
- ▲ Mixed Sample
- ● Siltstone

Figure 4.8 – Selected Trace Element Plots



4.6e. U (ppm) vs. Cu (ppm)

4.6f. U (ppm) vs. %TiO_2



4.6g. U (ppm) vs. %Fe_2O_3 (tot.)

4.6h. U (ppm) vs. % TiO_2+Fe_2O_3

Legend:

- ⬓ Weathered Sandstone
- ■ Sandstone
- ▲ Mixed Sample
- ● Siltstone

Figure 4.8 – Selected Trace Element Plots

While the samples were being artificially weathered, pH, temperature, total dissolved solids, and conductivity data were collected (Table 4.5). Changes in pH did not occur at a constant rate, but there was a gradual increase in values over time (Figure 4.9). Similarly, changes in conductivity and total dissolved solids did not occur at constant rates over time, or in relation to pH (Figures 4.10-4.12).

The two samples containing the highest uranium values (one sandstone and one siltstone) were analyzed for inorganics and dissolved metals (Table 4.6a). Three other samples, one sandstone, one siltstone, and one highly weathered sandstone were analyzed for total metals (Table 4.6b). Total rather than dissolved metals were analyzed for the second batch of samples to obtain approximate minimum (dissolved) and maximum (total) values respectively for metals in the water. A full suite analysis of the rainwater used during the artificial weathering experiment was also analyzed (Table 4.6c).

4.4.1 Rainwater

The natural rainwater used in the artificial weathering experiments was slightly acidic, having a initial pH of 5.08. Values for most inorganics and metals were below detection limits, with few exceptions: total organic carbon was present, with a concentration of 1mg/L, dissolved chloride also had a concentration of 1mg/L; aluminum was slightly elevated, with a concentration of 24µg/L; nickel, vanadium, and zinc were present, with values of 5µg/L, 9µg/L, and 10µg/L respectively.

Table 4.5a – data for AP06-05

Time (min)	pH	TDS (ppm)	Conductivity (uS)	Temperature (°C)
0	5.06	2.0	4.0	17.5
60	5.33	5.0	11.0	21.4
160	5.16	6.0	12.0	22.7
225	5.17	6.0	12.0	23.6
290	5.21	6.0	13.0	24.7
345	5.17	7.0	13.0	24.6
1010	5.31	7.0	14.0	24.5
1125	5.19	6.0	13.0	24.6
1530	5.45	5.0	10.0	24.7
2700	5.50	5.0	9.0	24.9

Table 4.5b – data for AP06-02

Time (min)	pH	TDS	Conductivity (uS)	Temperature (°C)
0	5.06	2.0	4.0	17.5
60	5.51	6.0	14.0	20.7
160	5.57	6.0	14.0	21.0
225	5.33	7.0	14.0	22.9
290	5.34	10.0	21.0	24.9
345	5.16	9.0	17.0	24.9
1010	5.62	8.0	16.0	23.3
1125	5.36	9.0	18.0	23.7
1530	5.49	11.0	19.0	24.4
2700	5.57	15.0	26.0	24.9

Table 4.5c – data for AP06-01

Time (min)	pH	TDS	Conductivity (uS)	Temperature (°C)
0	5.94	2.0	3.0	19.8
60	6.12	5.0	11.0	21.3
140	5.98	6.0	12.0	22.4
270	5.84	6.0	11.0	23.3
480	5.89	7.0	14.0	24.6
1440	5.65	7.0	16.0	24.7
2520	5.52	9.0	17.0	24.4
4020	5.49	12.0	17.0	24.6
5780	5.37	11.0	19.0	24.6
6800	5.42	11.0	18.0	24.8
8610	5.48	9.0	21.0	24.5

Table 4.5d – data for AP06-03

Time (min)	pH	TDS	Conductivity (uS)	Temperature (°C)
0	5.94	2.0	3.0	19.8
60	6.08	5.0	9.0	21.6
140	6.02	5.0	12.0	22.1
270	5.87	7.0	12.0	23.8
480	5.74	8.0	14.0	24.6
1440	5.46	7.0	26.0	24.4
2520	5.67	7.0	24.0	23.9
4020	5.49	7.0	24.0	23.7
5780	5.44	8.0	20.0	24.2
6800	5.47	6.0	17.0	24.6
8610	5.51	6.0	16.0	24.7

Table 4.5e – data for AP06-04

Time (min)	pH	TDS	Conductivity (uS)	Temperature (°C)
0	5.94	2.0	3.0	19.8
60	6.23	6.0	15.0	21.3
140	6.34	6.0	15.0	21.5
270	6.19	8.0	16.0	22.4
480	5.84	11.0	18.0	23.1
1440	5.59	9.0	19.0	24.6
2520	5.12	8.0	18.0	24.7
4020	5.26	10.0	21.0	24.5
5780	5.71	10.0	23.0	24.4
6800	5.73	12.0	25.0	24.7
8610	5.64	14.0	23.0	24.8

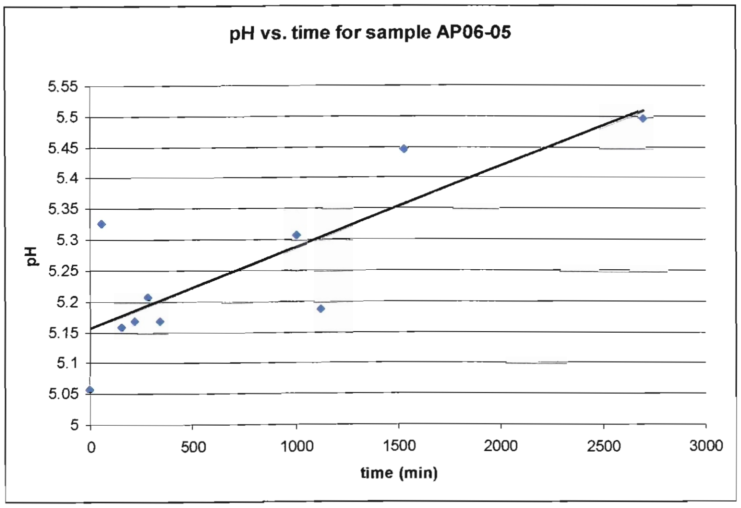


Figure 4.9 – pH vs. time for water sample AP06-05. Trend line indicates there was a gradual, nonlinear increase in pH over time.

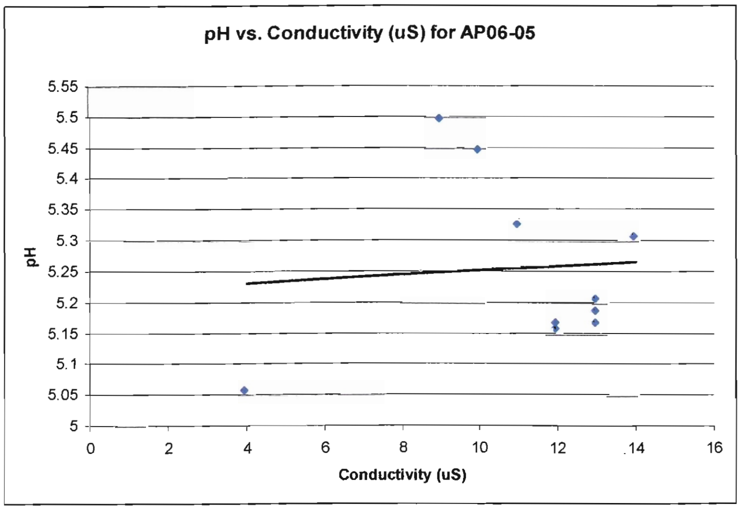


Figure 4.10 – pH vs. conductivity for water sample AP06-05. R^2=0.0056, indicating that there is no correlation between pH and conductivity.

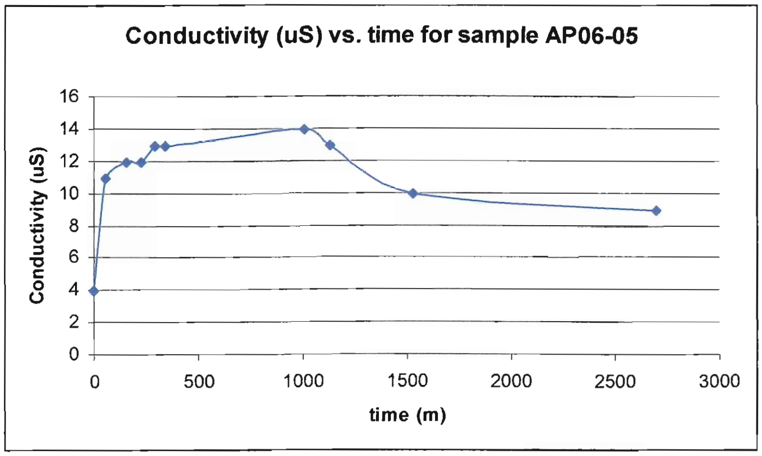


Figure 4.11 – Conductivity vs. time for water sample AP06-05. Conductivity did not change at a constant rate over time.

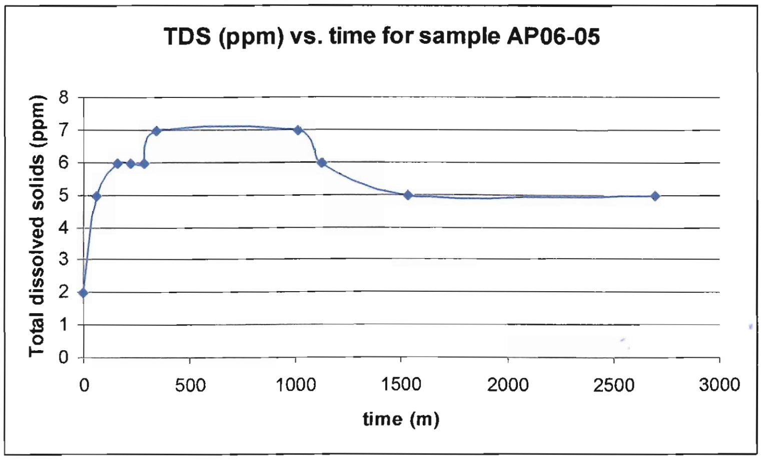


Figure 4.12 – Total Dissolved Soils vs. time for water sample AP06-05. TDS did not change at a constant rate over time.

Table 4.6a – Atlantic RCAp-MS dissolved in water - Analysis of a sandstone (AP06-05) and a siltstone (AP06-02).

	Units	AP06-05	AP06-02	RDL
INORGANICS				
Total Alkalinity (Total as CaCO3)	mg/L	ND	ND	5
Dissolved Chloride (Cl)	mg/L	1	4	1
Colour	TCU	ND	ND	5
Hardness (CaCO3)	mg/L	ND	1	1
Nitrate + Nitrite	mg/L	ND	ND	0.05
Nitrite (N)	mg/L	ND	ND	0.01
Nitrogen (Ammonia Nitrogen)	mg/L	ND	ND	0.05
Total Organic Carbon (C)	mg/L	8	ND	5
Orthophosphate (P)	mg/L	0.01	ND	0.01
pH	pH	5.5	5.24	N/A
Reactive Silica (SiO2)	mg/L	3.6	6.9	0.5
Dissolved Sulphate (SO4)	mg/L	3	3	2
Turbidity	NTU	350	430	3
Conductivity	uS/cm	17	26	1
RCAP CALCULATIONS				
Anion Sum	me/L	0.1	0.17	N/A
Bicarb. Alkalinity (calc. as CaCO3)	mg/L	ND	ND	1
Calculated TDS	mg/L	11	19	1
Carb. Alkalinity (calc. as CaCO3)	mg/L	ND	ND	1
Cation Sum	me/L	0.13	0.21	N/A
Ion Balance (% Difference)	%	13	10.5	N/A
Langelier Index (@ 20C)	N/A	NC	NC	N/A
Langelier Index (@ 4C)	N/A	NC	NC	N/A
Saturation pH (@ 20C)	N/A	NC	NC	N/A
Saturation pH (@ 4C)	N/A	NC	NC	N/A

Elements (ICP-MS)	**Units**	**AP06-05**	**AP06-02**	**RDL**
Dissolved Aluminum (Al)	ug/L	78	29	10
Dissolved Antimony (Sb)	ug/L	ND	ND	2
Dissolved Arsenic (As)	ug/L	ND	ND	2
Dissolved Barium (Ba)	ug/L	ND	ND	5
Dissolved Beryllium (Be)	ug/L	ND	ND	2
Dissolved Bismuth (Bi)	ug/L	ND	ND	2
Dissolved Boron (B)	ug/L	12	20	5
Dissolved Cadmium (Cd)	ug/L	ND	ND	0.3
Dissolved Chromium (Cr)	ug/L	ND	ND	2
Dissolved Cobalt (Co)	ug/L	2	ND	1
Dissolved Copper (Cu)	ug/L	12	18	2
Dissolved Iron (Fe)	ug/L	ND	ND	50
Dissolved Lead (Pb)	ug/L	ND	ND	0.5
Dissolved Manganese (Mn)	ug/L	21	5	2
Dissolved Molybdenum (Mo)	ug/L	ND	ND	2
Dissolved Nickel (Ni)	ug/L	ND	ND	2
Dissolved Selenium (Se)	ug/L	ND	ND	2
Dissolved Silver (Ag)	ug/L	ND	ND	0.5
Dissolved Strontium (Sr)	ug/L	ND	ND	5
Dissolved Thallium (Tl)	ug/L	ND	ND	0.1
Dissolved Tin (Sn)	ug/L	ND	ND	2
Dissolved Titanium (Ti)	ug/L	2	ND	2
Dissolved Uranium (U)	ug/L	11	3.3	0.1
Dissolved Vanadium (V)	ug/L	ND	ND	2
Dissolved Zinc (Zn)	ug/L	ND	6	5
Elements (ICP-OES)				
Dissolved Calcium (Ca)	mg/L	0.1	0.2	0.1
Dissolved Magnesium (Mg)	mg/L	0.1	0.1	0.1
Dissolved Phosphorus (P)	mg/L	ND	ND	0.1
Dissolved Potassium (K)	mg/L	1.4	1.7	0.1
Dissolved Sodium (Na)	mg/L	1.8	3.1	0.1
RCAP CALCULATIONS				
Nitrate (N)	mg/L	ND	ND	0.05

Table 4.6b – Total metals - Analysis of two sandstones (AP06-01 and AP06-03) and a siltstone (AP06-04).

	Units	AP06-01	AP06-03	AP06-04	RDL
Elements (ICP-MS)					
Total Aluminum (Al)	ug/L	2500	1500	4500	10
Total Antimony (Sb)	ug/L	ND	ND	ND	2
Total Arsenic (As)	ug/L	12	14	3	2
Total Barium (Ba)	ug/L	39	27	38	5
Total Beryllium (Be)	ug/L	ND	ND	ND	2
Total Bismuth (Bi)	ug/L	ND	ND	ND	2
Total Boron (B)	ug/L	39	27	11	5
Total Cadmium (Cd)	ug/L	ND	ND	ND	0.3
Total Chromium (Cr)	ug/L	ND	ND	2	2
Total Cobalt (Co)	ug/L	28	5	6	1
Total Copper (Cu)	ug/L	19	15	18	2
Total Iron (Fe)	ug/L	4800	1800	5500	50
Total Lead (Pb)	ug/L	33	14	25	0.5
Total Manganese (Mn)	ug/L	750	110	2100	2
Total Molybdenum (Mo)	ug/L	ND	ND	ND	2
Total Nickel (Ni)	ug/L	5	ND	4	2
Total Selenium (Se)	ug/L	ND	ND	ND	2
Total Silver (Ag)	ug/L	ND	ND	ND	0.5
Total Strontium (Sr)	ug/L	ND	ND	ND	5
Total Thallium (Tl)	ug/L	0.5	0.2	0.3	0.1
Total Tin (Sn)	ug/L	ND	ND	ND	2
Total Titanium (Ti)	ug/L	57	3	27	2
Total Uranium (U)	ug/L	11	24	16	0.1
Total Vanadium (V)	ug/L	4	4	11	2
Total Zinc (Zn)	ug/L	14	22	20	5

Table 4.6c – Atlantic RCAp-MS dissolved in water - Analysis of the natural rainwater sample (N/A = not applicable; ND = value below detection limit; NC = constituent was not analyzed).

	Units	AP-06-RW	RDL
INORGANICS			
Total Alkalinity (Total as CaCO3)	mg/L	ND	5
Dissolved Chloride (Cl)	mg/L	1	1
Colour	TCU	ND	5
Hardness (CaCO3)	mg/L	ND	1
Nitrate + Nitrite	mg/L	0.06	0.05
Nitrite (N)	mg/L	ND	0.01
Nitrogen (Ammonia Nitrogen)	mg/L	ND	0.05
Total Organic Carbon (C)	mg/L	1	0.5
Orthophosphate (P)	mg/L	ND	0.01
pH	pH	5.08	N/A
Reactive Silica (SiO2)	mg/L	ND	0.5
Dissolved Sulphate (SO4)	mg/L	ND	2
Turbidity	NTU	0.3	0.1
Conductivity	uS/cm	10	1
RCAP CALCULATIONS			
Anion Sum	me/L	0.04	N/A
Bicarb. Alkalinity (calc. as CaCO3)	mg/L	ND	1
Calculated TDS	mg/L	3	1
Carb. Alkalinity (calc. as CaCO3)	mg/L	ND	1
Cation Sum	me/L	0.06	N/A
Ion Balance (% Difference)	%	20	N/A
Langelier Index (@ 20C)	N/A	NC	N/A
Langelier Index (@ 4C)	N/A	NC	N/A
Saturation pH (@ 20C)	N/A	NC	N/A
Saturation pH (@ 4C)	N/A	NC	N/A

Elements (ICP-MS)	**Units**	**AP06-RW**	**RDL**
Total Aluminum (Al)	ug/L	24	10
Total Antimony (Sb)	ug/L	ND	2
Total Arsenic (As)	ug/L	ND	2
Total Barium (Ba)	ug/L	ND	5
Total Beryllium (Be)	ug/L	ND	2
Total Bismuth (Bi)	ug/L	ND	2
Total Boron (B)	ug/L	ND	5
Total Cadmium (Cd)	ug/L	ND	0.3
Total Chromium (Cr)	ug/L	ND	2
Total Cobalt (Co)	ug/L	ND	1
Total Copper (Cu)	ug/L	2	2
Total Iron (Fe)	ug/L	ND	50
Total Lead (Pb)	ug/L	0.7	0.5
Total Manganese (Mn)	ug/L	ND	2
Total Molybdenum (Mo)	ug/L	ND	2
Total Nickel (Ni)	ug/L	5	2
Total Selenium (Se)	ug/L	ND	2
Total Silver (Ag)	ug/L	ND	0.5
Total Strontium (Sr)	ug/L	ND	5
Total Thallium (Tl)	ug/L	ND	0.1
Total Tin (Sn)	ug/L	ND	2
Total Titanium (Ti)	ug/L	ND	2
Total Uranium (U)	ug/L	ND	0.1
Total Vanadium (V)	ug/L	9	2
Total Zinc (Zn)	ug/L	10	5
Elements (ICP-OES)			
Total Calcium (Ca)	mg/L	0.1	0.1
Total Magnesium (Mg)	mg/L	ND	0.1
Total Phosphorus (P)	mg/L	ND	0.1
Total Potassium (K)	mg/L	0.7	0.1
Total Sodium (Na)	mg/L	0.5	0.1
RCAP CALCULATIONS			
Nitrate (N)	mg/L	0.06	0.05

4.4.2 Sandstones

In the water collected from weathering sandstones in the first batch of samples, total organic carbon increased during the 48 hour weathering process from 1µg/L to 8µg/L. There was also a slight increase in pH from 5.08 to 5.50. There was no change in the amount of dissolved chloride, a value of 1mg/L was obtained for both the rainwater and the sandstone leached sample. Dissolved sulphate increase from below the detection limit to 3mg/L in the sandstone leached sample. Metal analysis was done as dissolved metals in the first batch, and total metals in the second batch. While the values obtained were not identical, most of the metals that were leached from the sandstones were found in both the dissolved and total metal analysis. Uranium was found in waters from both batches, with a concentration of 11-16ppb. Aluminum, manganese, and titanium were elevated in both the dissolved metals and total metals analyses. Iron was found in concentrations up to 5500ppb in the total metals, but was not found in the dissolved metal analysis.

4.4.3 Siltstones

Waters collected from the artificial weathering of siltstones showed similar values similar to those obtained from the artificial weathering of sandstones (Table 4.5). There was a slight pH increase in the waters from the siltstones, from 5.09 to 5.24. There was more chloride in the waters from the siltstones, 4mg/L compared to 1mg/L in the rainwater. Dissolved sulphate was found in the same concentration as in the water from the sandstones, 3mg/L. Most of the metals that were elevated in waters from the sandstones were also found in waters from the siltstones, but in lower amounts. Aluminum and iron, which had concentrations of 2500-4500µg/L and 4800-5500µg/L in the waters from the sandstones, had concentrations of 1500µg/L and 1800µg/L in the waters from the siltstones. The exception was uranium, which in the total metal analysis had a value of 73µg/L in the water from the siltstone, but values of 27-57µg/L in the waters from the sandstones.

4.5 Summary

All three data sets are consistent in showing the more highly weathered whole rock sandstone sample (AP06-01) to be depleted in most elements, but enriched in silica relative to the other samples. Elevated uranium values were found in all whole rock samples from the study area, and are one to two orders of magnitude above background values. Uranium values were highest in samples of different lithologies from different locations. Water analysis suggests that a number of elements are indeed leached from these rocks during weathering.

5.0 Discussion

The previous chapter presented data collected from various observations and experiments. The following sections focus on the interpretation of these data, and implications of naturally elevated levels of uranium in Horton Group sandstones.

5.1 Field Observations

There were two important factors in the study area that have contributed to the potential for uranium in surface waters in the area: (1) proximity of the outcrops to surface water, and (2) weathering. The buildup of loose sediments and debris at the base of the cliff indicated weathering was causing the uranium enriched sediment to become physically mobile. Further analysis was needed to determine whether the sediments could also be chemically weathering, as outlined below.

5.2 Mineralogical Data

Previous work on fission track analysis (O'Beirne-Ryan, 2006) indicated that uranium was concentrated within the biotite grains of the Horton Group sedimentary strata. For this reason, many of the grains examined during microprobe analysis were biotites. The microprobe analyses confirm the sequential weathering pattern occurring in the biotite grains. The observed weathering pattern was as follows (Figure 4.2):

Biotite → "Vermiculite" → Kaolinite

The sequence of mineralogical weathering, also described by Velde (1995), infers that if there is uranium locked up in the parent biotite, as the grains weather, there is potential for uranium to become mobile. This weathering pattern is similar to that observed in the weathering of the biotites of the SMB monzogranite (O'Beirne-Ryan, 2006).

The K-feldspars in the sandstones appear unweathered in thin section. A granite weathering trend by Nesbitt and Young (1989) suggests that mol% K_2O approaches zero as granite weathers (Figure 4.7). This suggests that the Horton Group sediments were derived from relatively fresh granite of the South Mountain Batholith, as opposed to a more weathered granite, which would have more highly weathered grains of K-feldspar.

5.3 Whole Rock Geochemistry

5.3.1 Major Elements

A number of the major elements (Al_2O_3, TiO_2, MgO) have a strong positive correlation with silica (Figure 4.6). Siltstones are more enriched in these elements than the sandstones, however the highly weathered sandstone sample is depleted in most major elements, with the exception of silica, in which it is relatively enriched. Elements, such as CaO, NaO, and K_2O, were found in the same relative proportions in all samples (Table 4.2).

5.3.2 Trace Elements

Trace element analyses show that siltstones are enriched in a number of trace elements relative to the sandstones (Table 4.3); siltstones have relatively higher values of Zr, Th, Zn, Cu, and V than the sandstones. Cobalt is elevated in sandstones relative to siltstones. The more highly weathered sandstone sample is depleted in trace elements, except for Ba and Rb.

5.3.3 Uranium Data

O'Reilly (1982) stated that in sedimentary rocks, uranium tends to be concentrated in shale or clay rich layers. Data from this study are only partially consistent with this statement, as the two samples with highest uranium were a siltstone from location 1 (212.1ppm) and a sandstone from location 2 (109.2ppm). This observation suggests that the uranium is not tied to a specific lithology within the Cheverie Formation. Strong positive correlations between uranium and other trace elements (such as V and Pb) indicate that the uranium may be mobilized with these elements. Such association is typical of roll front deposits, as can be seen in Figure 5.1 (Skinner, 1999). There is a poor correlation with TiO_2 and Fe_2O_3, indicating that uranium is not linked only to these elements. The more highly weathered sandstone sample is depleted in uranium, at least compared to the fresher sandstone, confirming that the uranium is mobilized upon weathering (Table 4.3).

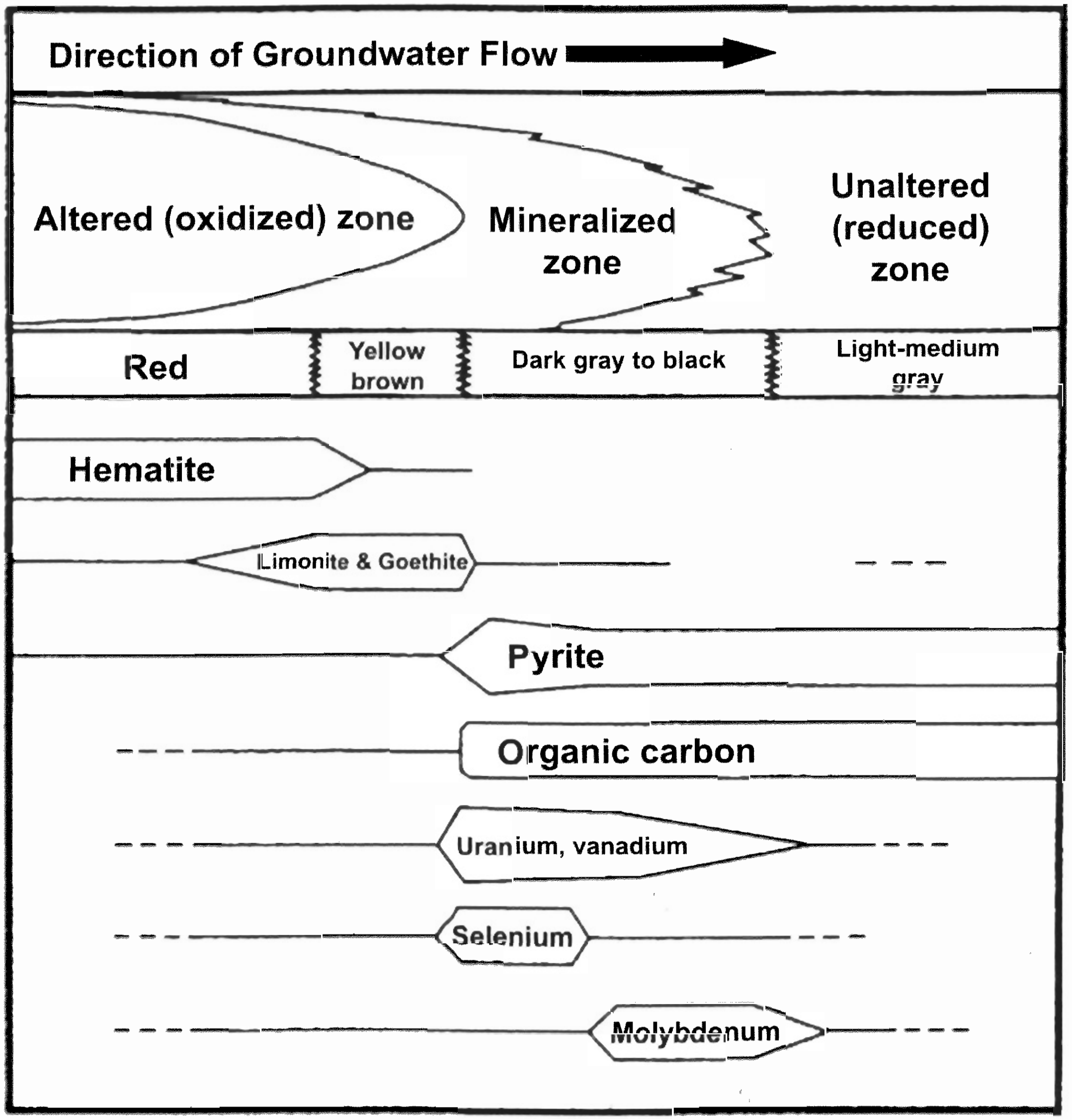


Figure 5.1 – Roll front cross section illustrating the mineralogical, elemental, and colour changes as a result of groundwater flow (Modified from Skinner, 1999).

5.4 Water Geochemistry

Based on the artificial weathering experiments, it is possible to determine which elements are leaching out of the rocks by comparing the chemistry of water drained from the rock samples to the chemistry of the natural rainwater sample. Analysis for total metals involved analysis of an unfiltered water sample, which may have included fine particulate matter. Analysis for dissolved species represents data collected from filtered samples, and therefore is a measure of the metal species in solution only. In both the dissolved and total metal data there were elements leached in varying proportions from both lithologies. Uranium, copper, aluminum, iron, and manganese were found in elevated concentrations in waters from sandstones and siltstones (Table 5.1a). Selected values of Canadian Drinking Water Quality Guidelines (Health Canada, 2006) are given in Table 5.1b. Because concentrations of aluminum, iron, lead, and manganese are above the guideline levels and are likely present as suspended particulate matter or adsorbed onto suspended particulates, it would be recommended that drinking water in the area be filtered prior to consumption.

As was found with the whole rock geochemistry results, lithology was not the only determining factor for the level of uranium leached from a sample. The higher uranium values in the water samples were from the rocks with the lower total uranium concentrations (Table 5.1a). This suggests uranium may be mobilized more readily from rocks that are already partially weathered. For example, whole rock geochemical analysis of the more highly weathered sandstone yielded a value of 6.3ppm, the value for a less weathered sandstone was 39.7ppm. Uranium values in the leached waters were 16ppb and 11ppb, respectively, confirming that more uranium was mobilized more intensly from a sample with a lower total uranium value in whole rock geochemistry.

Table 5.1a – Elements leached from sandstones and siltstones during artificial weathering, (d) indicates analysis was for dissolved metals, (tot) indicates analysis was for total metals. Numbers marked in yellow indicate a concentration above the value given in the Guidelines for Canadian Drinking Water Quality (Health Canada, 2006).

Element	**Unit**	**Sandstones**		**Siltstones**	**Rainwater**
	Sample>	**AP06-05**		**AP06-02**	
Al (d)	**µg/L**	78		29	24
B (d)	**µg/L**	12		20	N/D
Co (d)	**µg/L**	2		N/D	N/D
Cu (d)	**µg/L**	12		18	2
Mn (d)	**µg/L**	21		5	N/D
Ti (d)	**µg/L**	2		N/D	N/D
U (d)	**µg/L**	11		3.3	N/D
K (d)	**mg/L**	1.4		1.7	0.7
Na (d)	**mg/L**	1.8		3.1	0.5
	Sample>	**AP06-03**	**AP06-01**	**AP06-04**	
Al (tot)	**µg/L**	2500	4500	1500	24
As (tot)	**µg/L**	12	3	14	N/D
Ba (tot)	**µg/L**	39	38	27	N/D
B (tot)	**µg/L**	39	11	27	N/D
Co (tot)	**µg/L**	28	6	5	N/D
Cu (tot)	**µg/L**	19	18	15	2
Fe (tot)	**µg/L**	4800	5500	1800	N/D
Pb (tot)	**µg/L**	33	25	14	0.7
Mn (tot)	**µg/L**	750	2100	110	N/D
Tl (tot)	**µg/L**	0.5	0.3	0.2	N/D
Ti (tot)	**µg/L**	57	27	73	N/D
U (tot)	**µg/L**	11	16	24	N/D
Zn (tot)	**µg/L**	14	20	22	10

Table 5.1b - Selected element values for upper limits in drinking water, from the Guidelines for Canadian Drinking Water Quality (Health Canada, 2006). All values are in µg/L.

Parameter	**Health-based or Aesthetic Guideline**
Aluminum (Al)	100
Iron (Fe)	300
Lead (Pb)	10
Manganese (Mn)	50
Uranium (U)	20

As stated in Section 2.4.1, uranium can form soluble complexes with carbonate, phosphate, sulphate, fluoride, and silicate ions (Langmuir, 1978). Data collected from the artificial weathering experiments was used to determine the association of uranium in the experimentally leached waters, by assessing the amounts of each ion mobilized during the artificial weathering.

Low to undetectable levels of carbonate, phosphate, and sulphate anionic species in the water analyzed indicate that the elevated levels of uranium found in the water are not likely the result of uranium complexation with these species only (Figure 2.3). However, because the levels of uranium being mobilized at the current time are low, sulphate may be contributing to the mobilization. Langmuir (1978) also suggests that uranium can complex with fluorine (Figure 2.3) at pH values similar to water analyzed in this study.

Although the water chemistry analysis does not include data for fluorine, MacDonald (2001), reports typical values for fluorine within plutons of the South Mountain Batholith as ~200-1000ppm. Because the SMB is the most likely source rock for the Horton Group sandstones, it is likely that similar concentrations would be found in the sandstones of this study, or that waters flowing through the granite terrane into the sandstones and shales could be elevated in fluorine, thus supplying the fluorine in the water to form a complex with uranium.

5.5 Implications for Groundwater and Surface Waters in Nova Scotia

Whole rock geochemical analysis of samples collected from the study area confirmed that Horton Group sandstones and shales have elevated levels of uranium. The concern with the elevated uranium in the study area is that when mobilized, it may potentially contaminate water sources. The whole rock geochemical analysis of all the sandstone and shale samples from the area confirmed that uranium levels in these rocks are 10-50 times greater than the background levels in the area. This elevation in uranium levels is of particular concern because of the close proximity to water and farmland, and to well waters in the region.

Chemical mobility of uranium in the Horton Group sandstones and shales was confirmed by the artificial weathering experiments. Table 5.1 lists the elements that were found to leach from the rocks during weathering. Uranium was mobilized from all samples tested, the highest value (24µg/L), from a siltstone (AP06-04), was above the Canadian Drinking Water Quality guideline of 20µg/L. Because the artificial weathering was carried out over a period of only several days, it is likely that amounts leached from the outcrops over a period of years would be higher.

As discussed in Section 5.4, uranium may be mobilized more readily from rocks that are already partially weathered. The outcrop in the study area is weathering readily in sections (Figure 2.2), and as it continues to weather, it is likely that greater amounts of uranium will become mobilized, causing a greater risk to waters in the area.

Elemental associations with uranium can be seen from whole rock geochemical analysis (Figures 4.6-4.8), and results of water sample analysis (Tables 4.6, 5.1). While the whole rock geochemical analysis was useful to determine what elements are associated with the uranium in situ, the water analysis allowed for the determination of elements that are mobilized with uranium. Uranium in sandstones and siltstones in the study area may be mobilized by forming a soluble complex with fluoride. Because the granites of the South Mountain Batholith contain fluoride, it is likely that there is sufficient fluoride available in the area to complex with the uranium. Uranium can also become mobilized by attaching to OH or O_2 (Figure 2.3).

6.0 Conclusions

Based on results from microprobe analysis, whole rock geochemical analysis, and artificial weathering experiments, I conclude the following about the uranium in my study area:

(1) The weathering sequence observed in the biotite grains confirms that there is potential for any uranium in the biotite to become mobile as the biotite weathers to vermiculite and kaolinite.

(2) Weathered uranium-enriched sandstones have lower levels of uranium and other elements than the less weathered samples. This confirms that elements are liberated from the rocks as they weather.

(3) Leached uranium values are similar in both the total metal analysis and the dissolved metal analysis. This suggests that a high proportion of the uranium is dissolved in the water and is not just suspended particulate matter.

(4) Of the possible soluble complexes that mobilize uranium, fluoride may be causing the mobilization of uranium from the sandstones of the Horton Group. Other ions such as chlorine and sulphate may also be contributing to the uranium movement although they are present in very low quantities in the leached waters.

(5) Uranium is mobilized more readily from rocks that are partially weathered. This observation is found in both sandstones and siltstones, regardless of the total amount of uranium in the rock.

(6) From the water analysis for total metals and dissolved metals it appeared that much of the uranium leached from the rocks in is the dissolved phase. While simply filtering drinking water in the area will not reduce the amount of uranium dissolved, it will lower levels of aluminum, iron, lead, and manganese, as these are found in the water as suspended particulate matter.

6.1 Recommendations for Future Work

This thesis provides preliminary observations of the Horton Group sandstones, however there are several recommendations that can be made for future work in this area.

(1) A wider group of samples would be useful to verify the whole rock geochemical results, and also to determine uranium distribution in the different siltstone layers within the sandstone outcrops. Because this study focused primarily on a section of outcrop ~200m long, it would be beneficial to sample over a wider area, to determine if the geochemical results are unique to this particular section, or are representative of a larger area.

(2) Determination of the presence of the fluoride ion, its complexation with uranium, and possible sources of fluoride would be an interesting complementary work to this study. Rock samples should be evaluated by whole rock geochemical analysis and

water analysis, specifically for the fluoride ion, and should include both waters in the study area, and waters collected from artificial weathering.

(3) This study established that there is risk of contamination to waters in the area, both from uranium and from other metals. Testing of well waters in the area would establish if there are any health risks associated with local drinking water supplies.

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