POTENTIAL FOR THE PRESENCE OF RADON IN WATER FROM WEATHERED OUTCROPS OF URANIUM-BEARING STRATA OF SOUTHERN NOVA SCOTIA

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Submitted in partial fulfillment of the requirements
for the Degree of Bachelor of Science, Honours
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March 2008

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DATE: March, 2008.

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TITLE: Potential For The Presence of Radon in Water From Weathered Outcrops of Uranium-Bearing Strata of Southern Nova Scotia

Degree: B. Sc. Convocation: Spring Year: 2007

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Acknowledgements

I would like to thank Patrick Ryall, honours supervisor, for his support and guidance which made this thesis possible.

To Anne Marie O'Beirne-Ryan, my supervisor: I thank you for your guidance, patience and support. Also, your knowledge and advice was a great resource throughout the progression of this thesis.

I would also like to thank Bob Ryan and George O'Reilly from the NSDNR who had endless knowledge of the study area and were continuous resources of information during the extent of this thesis. Thank you to the NSDNR for financial support and for the access to the RAD7 radon detector which was essential to this study.

Abstract

Radon is a radioactive daughter product of uranium that is naturally occurring in sedimentary sequences and granitic intrusions throughout Nova Scotia. Occurrences can be found in Southern Nova Scotia in such outcrops as the South Mountain Batholith and the Horton Group. Health Canada has recently lowered the national regulated radon guideline for indoor air to 200 Bq/m^3 due to evidence that radon is the second leading cause of lung cancer.

This study was undertaken in order to establish whether rocks with variable uranium content from weathered outcrops of known uranium concentration, contributed significant amounts of radon to waters and soils associated with these rocks. Through a series of experiments it was determined that radon is readily leached from weathered rocks both of high uranium concentrations of 212 ppm and low uranium concentrations of 4 ppm. The uranium concentration of the source rock is not the sole indicator in identifying a radon problem in water and soil gas. Source rocks with low or high uranium concentrations yielded elevated radon levels in water of >1,000,000 Bq/m^3. This research has found that radon is not only being leached from the uranium in the rock but also from radium which is the direct parent rock of radon. Further analysis of the leached water identified that uranium is being leached into the water with concentrations as high as 22μg/L dissolved in the water, which exceeds the recommended guideline for Canadian drinking water. The main concern for the leaching of the radon from the source rock is the potential for the radon and radium to be redistributed through surface and groundwater. This study also confirmed that radon outgases more rapidly from warm water than from cold water, and although there is no current Canadian guideline for radon in water, radon in hot water outgases readily and so can contribute to the level of radon in indoor air, particularly in bathrooms of homes.

1.0 Introduction

1.1 Statement of Purpose

Radon is one of the radioactive daughter products of the uranium series, which also includes radium, the immediate parent of radon, and is naturally occurring in water, soil, and rocks throughout many areas in Nova Scotia. Among the rocks in Nova Scotia which have elevated levels of uranium are the sedimentary units of the Carboniferous Horton Group and granitic intrusions, such as the South Mountain Batholith of Devonian age. Radon associated with the Horton sedimentary rocks and granites of the South Mountain Batholith is the focus of this study.

Radon is currently a topic of interest in Canada as in 2006, the regulated Health Canada guideline for indoor air was lowered from 800 Becquerel per meters cubed (Bq/m^3) to 200 Bq/m^3. The topic is of particular interest to residents in Nova Scotia because of the presence of elevated levels in uranium and radon within the local bedrock, till and soil. The radioactivity and toxicity of these elements are a health concern to residents, as evidence indicates that radon is the second leading cause of lung cancer. (Health Canada, 2008)

The main focus of my thesis is to determine whether there is a correlation between the uranium and/or radium content of rocks and the concentration of radon in waters running over and through these rocks. The uranium concentration in a rock is not the only indicator of the possible presence of radon gas; radium is the immediate decay parent of radon and in some cases the uranium can be mobilized and transported elsewhere leaving behind a high radium concentration which then decays into radon gas. For this reason in the absence of uranium, radium would be the source of elevated radon. In order to assess the nature of the relationship between uranium and radium content in weathered rocks and their interaction with waters, I worked with fresh and weathered rock samples of known uranium content, ranging from a high concentration of 212 ppm to a lower concentration of 4ppm (Parsons, 2007; O'Beirne-Ryan, pers.comm. 2008). Saprolites, the in situ weathered profiles found at a number of locations on the South Mountain Batholith have higher permeability and therefore have a greater affect on groundwater flow. Saprolites also undergo chemical weathering changes which release radioactive elements such as uranium, radium, and radon, and due to the higher permeability these can be easily be mobilized into the hydrologic system under the right Eh and pH conditions (O'Beirne-Ryan, 2006). Parsons (2007) determined the uranium content in the parent material, and measured the uranium content of waters associated with sandstones and siltstones of the Lower Carboniferous Horton Group. However, a study of the distribution and nature of behavior of radon was not a part of the study by Parsons (2007). Prior to this study, the relationship between the levels of uranium in the rocks and radon in the waters draining these rocks was not known~~,~~ nor was it known what the impact of different parent rock and varying concentrations of uranium on the nature of this relationship might be. In addition, the possibility that temperature of the waters plays a role will be addressed, as waters of differing temperatures will be used to artificially leach radon from the natural samples, over differing timeframes. The technique for measuring radon in water is relatively straight forward, well established and economic, and working with rocks of known uranium content can provide data related to uranium in water and radon in water, which may in turn, serve as a proxy for radon in indoor air. In addition, one of the locations chosen (the South Mountain Batholith site) was selected because despite low concentrations of uranium in the saprolite, soil gas levels at the outcrop were elevated, so the question arises as to whether radon is being released from the saprolite. Understanding the relationship between uranium, radium and radon in soil, rock and water is key to providing important information to homeowners in terms of testing for radon and/or uranium and in mitigating effects of elevated radon and/or uranium levels.

1.2 Study Area

For this thesis the samples collected from outcrops with known uranium concentrations are from two different geological units. The first area of interest (Sample Site 1, Fig.1.1) is in the vicinity of the Green Street near Windsor, NS where the sedimentary samples were collected from the Lower Carboniferous Cheverie Formation of the Horton Group (Ryan et al., 2007). In September 2007, two sets of samples of siltstone and sandstone were obtained from the vicinity of Green Street located in Windsor, Nova Scotia which is also the sample location used in Parsons, (2007). The sequence of sandstone and interlayered siltstone beds within the outcrop has layers of elevated uranium content and layers in which the uranium content is low. The outcrop is adjacent to Sam's Brook and surface water from the creek was used in the experiments. This location is easily accessible by dirt road and the outcrop covered a length of 50 – 60 meters (Ryan, 1998).

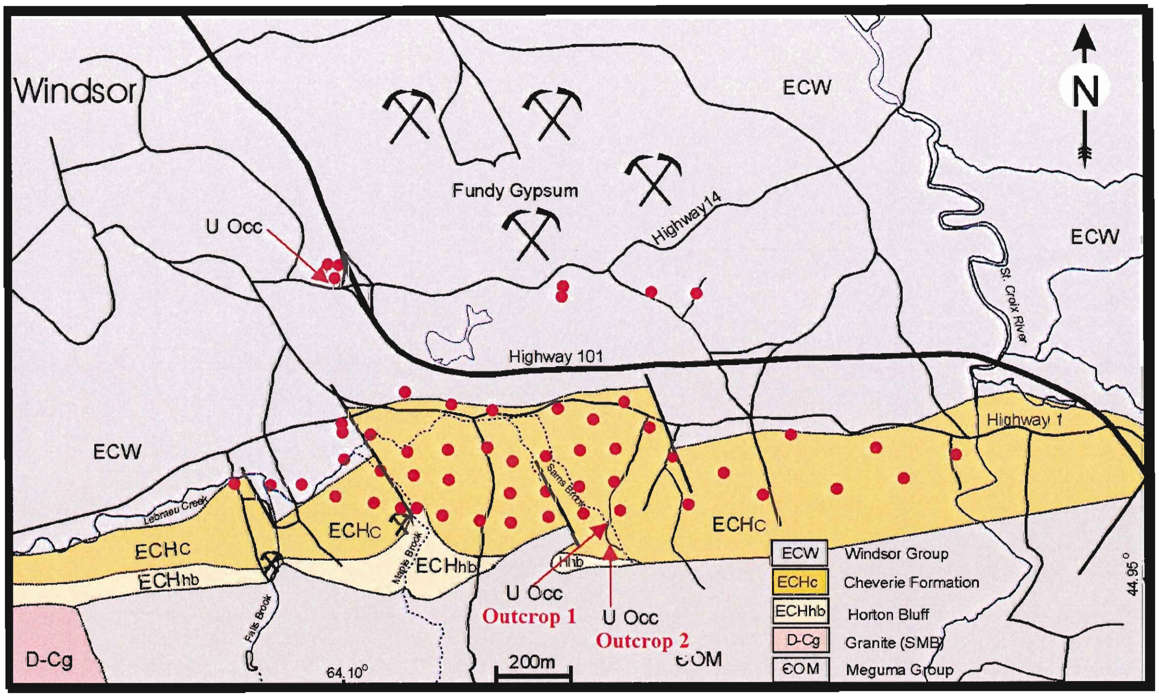

Figure 1.1 – Location of the Green Street Uranium Occurrence, near Windsor, Nova Scotia. Outcrop 1 is the location of uranium-rich samples; outcrop 2 is location of low-uranium samples. (After Ryan and O'Beirne-Ryan, 2007)

The second sampling locality is a weathered granite occurrence and is located on Morton Reeves Road near New Ross, Nova Scotia (Figure 1.2) within the South Mountain Batholith of Devonian age (MacDonald, 1992). This site is the same site from which samples were selected for geochemical analysis by O'Beirne-Ryan (pers com, 2008). Geochemical analysis of these weathered samples does not indicate elevated uranium content; however scintillometer readings of the outcrop indicate elevated radioactivity, and elevated levels of radon, as tested by a RAD 7 radon detector. The outcrop is accessible via an old mining road and covers a small area of approximately 10 – 15 m^2.

Figure 1.2 – Location of Reeve's Road Occurrence, near New Ross, Nova Scotia. (Modified from O'Beirne-Ryan and Zentilli, 1999)

Table 1.0 Sample Identification, locations as indicated on Figure 1.1 and Figure 1.2

Sample location	**Sample ID**	**Sample Type**	**Uranium Concentration**	**Gamma Anomaly**
Green Street, Windsor, NS	DF-07-01	Uranium Enriched Siltstone	212ppm	70,000- >100,000
Green Street, Windsor, NS	DF-07-02	Uranium Enriched Sandstone	109ppm	70,000- >100,000
Green Street, Windsor, NS	DF-07-03	Low Uranium Siltstone	56ppm	10,000-20,000
Green Street, Windsor, NS	DF-07-04	Low Uranium Sandstone	39ppm	10,000-20,000
Reeve's Road, New Ross, NS	DF-07-05	Granite	5ppm	20,000-40,000

1.3 Project Objectives

The objective of this project is to determine if there is a direct and predictable correlation between the content of uranium in sandstone, siltstone and granites and the radon measured in the water that is exposed to the rocks over various timeframes, and under variable pH and temperature conditions. In addition, because radium is radioactive and the immediate parent of radon, in the absence of elevated uranium in rocks, can elevated radon occur, and if so, does this serve as a potential indicator for elevated radium?

Page (1999), in a study which integrated data on uranium, radium, and radon in well waters from uranium exploration activity in the late 1970s, suggested that assessing the interrelationship between uranium, radium, and radon in soils, rocks, and waters was a promising approach to establishing potentially problematic areas. The study by Page (1999) however, did not undertake an intercomparison between uranium concentrations in a specific rock outcrop, radon in the overlying soil, and radon in waters draining this sample.

1.4 Format of Thesis

Chapter 1 introduces the topic, and examines the rationale of the study; Chapter 2 focuses on the background and geology of the study areas and the rationale for this study. Chapter 3 discusses the methods used while completing the field work as well as the protocol set in place to insure that each set of experiments were completed strictly and consistently to provide an accurate set of data. Chapter 4 reviews the results that were found once the experiments were completed and Chapter 5 is a discussion on the results. Lastly, Chapter 6 is a summary and conclusion of the findings of the thesis and recommendation for further study.

2.0 Geology

2.1 Introduction

The generalized geology of the study area within southwestern Nova Scotia consists of three primary units including the Meguma Supergroup, South Mountain Batholith, and the Horton Group. The South Mountain Batholith is a peraluminous granitoid body which intruded the Meguma Supergroup circa 370-380 million years ago, (MacDonald, 2001) The South Mountain Batholith is thought to have intruded in two main phases: the first stage is dominated by biotite grandiorite to leucomonzogranites, while the second stage is comprised of monzogranite to coarse and fine grained leucogranite (MacDonald, 2001). When compared to other granitic bodies in Nova Scotia the South Mountain Batholith is high in uranium (MacDonald, 2001).

The Horton Group of Late Devonian-Early Carboniferous age consists of red and grey polymictic conglomerates, arkose sandstone, mudstones, siltstones, and minor non-marine evaporites (Martel 1990; Van de Poll et al., 1995; Martel and Gibling, 1996). The Horton Group is lies as an angular unconformity on the Meguma Supergroup and also is found as a nonconformity to the South Mountain Batholith granitoids. (Martel 1990; Van de Poll et al., 1995; Martel and Gibling, 1996)

Nova Scotia has localized areas of elevated uranium, particularly in rocks of Devono-Carboniferous age (Figure 2.6). For the purpose of this thesis the geological units of interest vary from low level uranium concentrations found in weathered granitoid (saprolite) of the South Mountain Batholith (sample locality 2, Figure 1.2) to elevated levels of uranium found in sandstones and siltstones of the Horton Group (sample locality 1, Figure 1.1). The following discussion focuses on the detailed geology of sample location one in Horton Group sandstones and siltstones of Carboniferous age, and sample location two, in a leucogranite phase of the South Mountain Batholith.

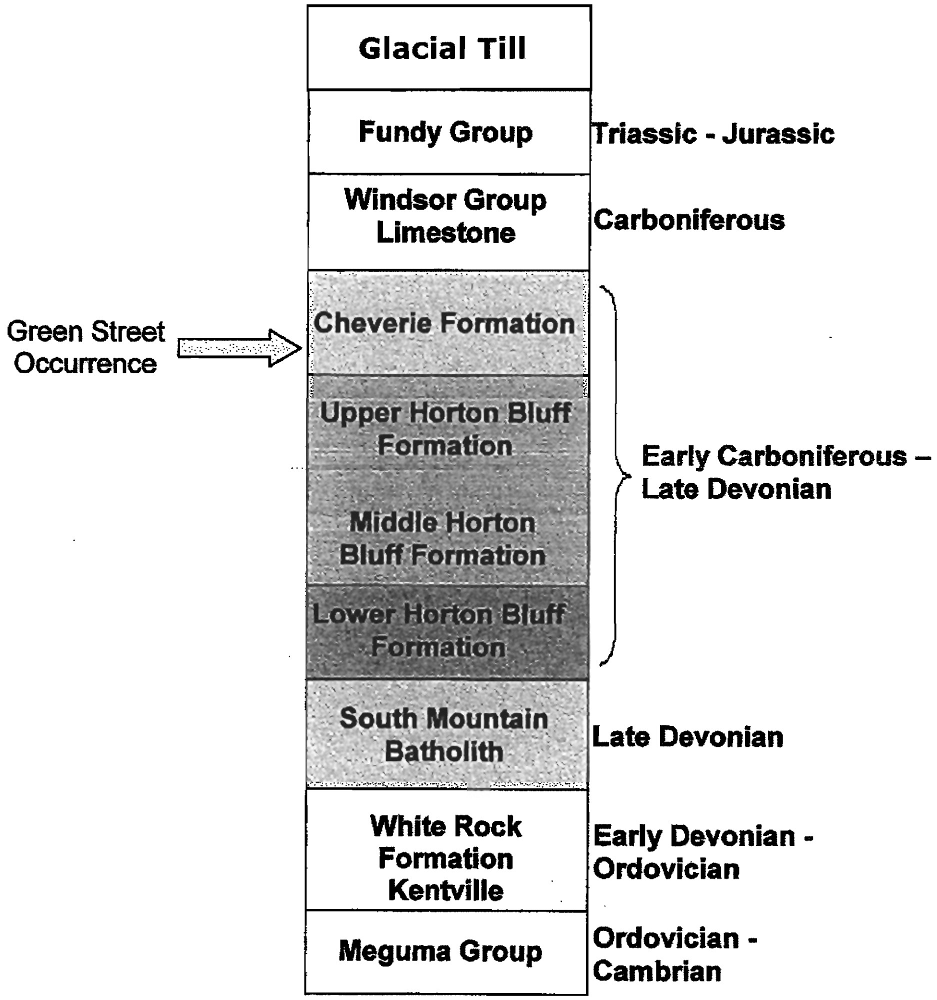

Figure 2.1 – Geological time sequence: Simplified stratigraphic column of both study areas. Reeve's Road granitoid occurs within the South Mountain Batholith. (Modified from Ryan, 1998; Parsons, 2007)

2.2 The Horton Group

Sample location one is within the Horton Group, a geological unit consisting of sedimentary rocks of Lower Carboniferous age (Bell, 1929) (Fig. 1.1 and 2.1). The Horton Group is comprised of red to grey beds of sandstone, conglomerate, mudstone, and siltstone. The Horton Group is overlain by limestone of the Windsor Group, also of Carboniferous age, and unconformably overlies granitoids of the South Mountain Batholith of late Devonian age, and the Cambrian – Ordovician metamorphic rocks of the Meguma Group in the study area (Bell, 1929).

The Horton Group includes the Horton Bluff Formation and the Cheverie Formation (Figure 1.2, Figure 2.1). For this project the arkosic sandstone and fine grained siltstone of the Cheverie Formation were sampled. The Cheverie Formation consists of coarse grained braided channel deposits along with fine grained floodplain sediments (Bell, 1929). The rocks of this formation contain naturally elevated uranium within some of the sandstone and siltstone layers. The sandstone is weathered and is weakly cemented, therefore easily eroded. Within the unit are small (10 – 15 centimetres) siltstone lenses which are also weathered (Ryan & O'Beirne-Ryan, 2006).

Underlying much of the Horton Group is the South Mountain Batholith, which consists of a series of granitic plutons ranging from granodiorite to leucogranite in composition (MacDonald, 2001). Weathering of the South Mountain Batholith was the source of many of the sediments deposited into basins throughout the eastern part of Canada during the late Devonian and early

Carboniferous, which resulted in the accumulation of the sediments of the Horton Group (Bell, 1929). The depositional environment of the Horton Bluff Formation was that of fluvial-lacustrine system. During the formation of the middle and upper portion of the Horton Bluff Formation regional flooding changed the depositional environment to a lacustrine environment (Martel and Gibling, 1996).

The Cheverie Formation unconformably overlies the Horton Bluff Formation. The unconformity represents a period of erosion of the Horton Bluff however, data from spores indicate that the period in question represents very short time (Martel and Gibling, 1996). The depositional environment of the Cheverie Formation near the basin margins was that of an alluvial system, with sediment sourced predominantly from the South Mountain Batholith (Martel and Gibling, 1996).

The Cheverie Formation at sample locality 1 (Figure 1.1) is exposed in outcrops of sandstone with siltstone lenses at the Green Street occurrence near Windsor, Nova Scotia (Figure 2.2). These outcrops have been oxidized and weathered, and erosion is ongoing with the eroded sediments deposited in nearby ditches and the local creek (Sam's Brook). Ryan and O'Beirne-Ryan (2007) have described the uranium occurrence at the Green Street location as being a preserved tail of a uranium roll front deposit. Uranium roll front deposits reflect the reconcentration of uranium at redox boundaries within sandstone bodies down hydrologic gradient (Figure 2.3). (Ryan and O'Beirne-Ryan 2007)

Figure 2.2 – Outcrop 2 at Green Street occurrence, near Windsor, Nova Scotia.

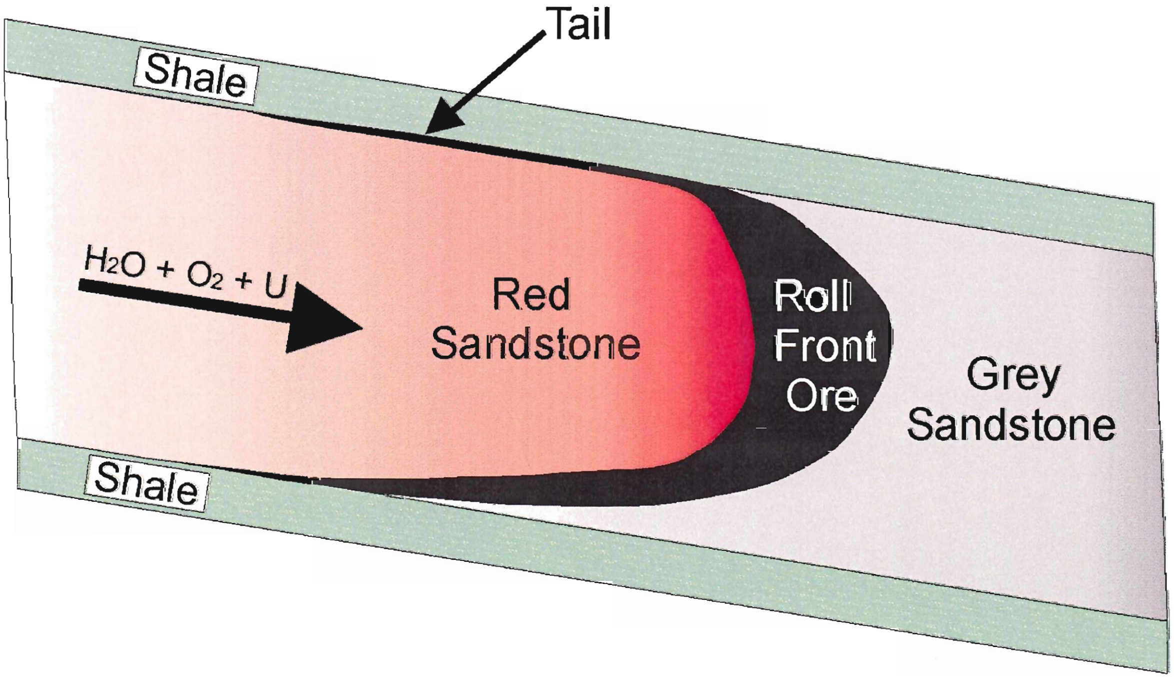

Figure 2.3 – Uranium roll front deposits. (Ryan and O'Beirne-Ryan 2007)

2.3 South Mountain Batholith

There are numerous uranium and polymetallic deposits and occurrences found within the South Mountain Batholith. Previous interpretations of these occurrences have suggested that they formed from fluid migration in late-stage granitoid emplacement (MacDonald, 2001). These occurrences can be found within shear and fracture zones or in late stage plutons (Chatterjee et al., 1982; MacDonald, 2001). MacDonald (2001) also suggests theses occurrences may be peribatholithic and found in the Meguma country rocks. Within the South Mountain Batholith of Nova Scotia there are five locations that have tin occurrences one of which is that of the Reeves Tin Mine (Smith and Turek, 1976), which is approximately 150 metres from sample location 2. (Figure 1.2) East Kemptville tin deposit also has elevated uranium concentration. (Chatterjee et al, 1982).

The Reeves Road granitoid of the South Mountain Batholith is categorized as a leucomonzogranite (McDonald, 2001). This coarse grained equigranular leucomonzogranite is predominately buff-colored and at sample location two, the focus of this study, due to the outcrop's intensely weathered state, it is categorized as a saprolite in which the rock can be hand-picked from the surface of the outcrop. This weathered saprolite crumbles very easily in your hand and is coarse grained with 70% whitish-pinkish potassium feldspar, 10% biotite, and 15% quartz. Leucomonzogranites accounts for approximately 22% of the South Mountain Batholith with the remaining consisting of granodiorite and monzogranite (MacDonald, 2001). Because the granite at sample location 2 has weathered to a saprolite, leaching of the more soluble elements has occurred (O'Beirne-Ryan, 2006).

The outcrop is approximately six meters in length and two meters in height. The outcrop is currently over grown with natural revegetation almost hiding the outcrop from roadside view since previously visited, in 2003 (Figure 2.4).


Figure 2.4a – Reeve's Road occurrence, near New Ross, Nova Scotia


Figure 2.4b – Sample taken at Reeve's Road occurrence, near New Ross, Nova Scotia.

2.4 Properties of Uranium and Radon

Uranium is a naturally occurring radioactive element which has an atomic number of 92 and has three different isotopes ^{234}U, ^{235}U, and ^{238}U all of which are radioactive. For the purpose of this project the uranium isotope of interest is ^{238}U because ^{238}U is approximately 140 times more abundant than the other isotopes. This radioactive isotope is unstable and decays into a series of daughter products, all except the last of which are themselves radioactive (Figure 2.5). The resulting stable isotope of ^{238}U is ^{206}Pb. One of the radioactive decay daughters of ^{238}U is ^{226}Ra which is the immediate parent of ^{222}Rn, the only isotope in the series that is a gas. When radon decays, it releases an alpha particle and transforms to solid daughter products ^{218}Po and ^{214}Po (Figure 2.5). As radon is a gas, radon in air is a concern in soil and homes. Radon in water is also a concern due to the ability of the gas to be transported through water and therefore enter groundwater sources or out-gas away from the source of the radon. There is currently a guideline for indoor air quality in Canada (200 Bq/m^3), but the US EPA is currently considering a radon in water guideline, of 4,000 pCi/L (1pCi/L = 37 Bq/m^3). The relationship between radon and uranium in waters and soils associated with the Cheverie sandstones and siltstones and with the Reeves leucomonzonite is the focus of this project.

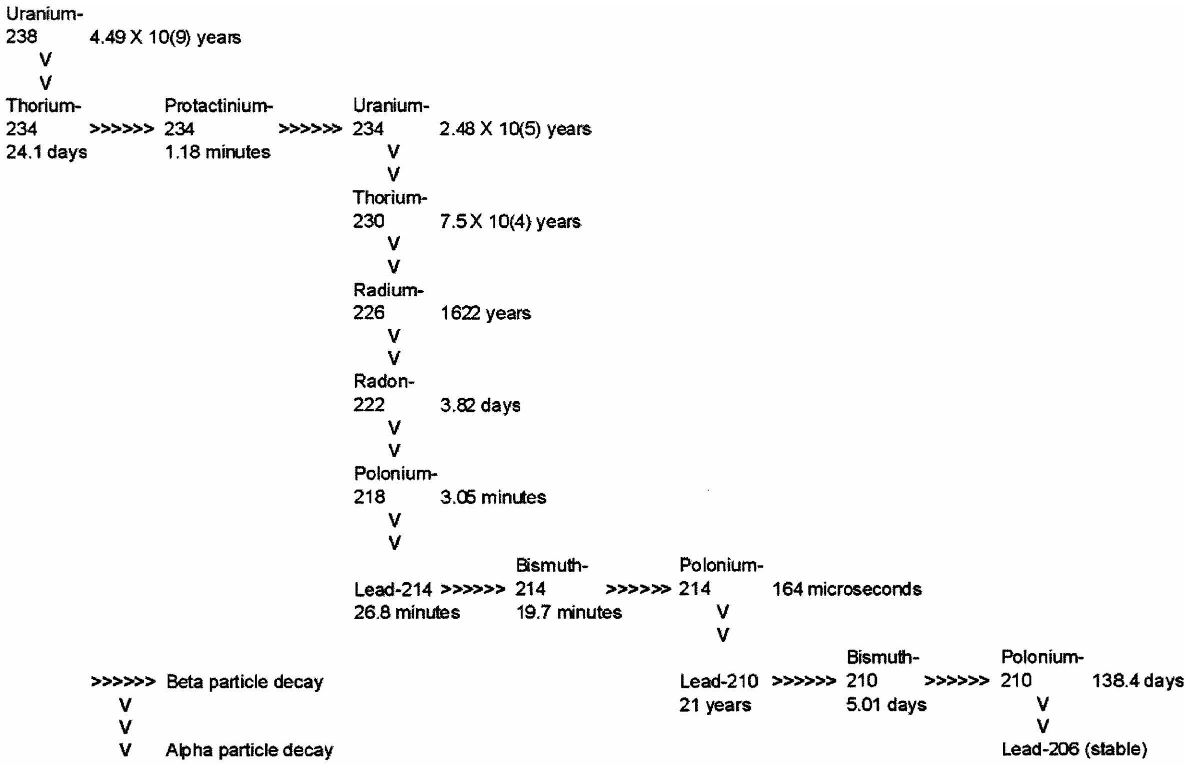

Figure 2.5 – Uranium decay series (USGS, 1999)

Uranium has two main valence states; +4 and +6. When found in the +6 state, uranium is chemically mobile whereas when uranium is present in the +4 state it is not mobile. In the weathering environment, and indeed any time uranium is oxidized from U^{4+} to U^{6+}, the uranium may form soluble complexes in water (Ivanovich and Harmon, 1992).

^{222}Rn is a naturally occurring radioactive decay product from the ^{238}U decay series. Radon is an inert gas that is colorless and odourless and has no static electrical charge. ^{222}Rn-has an atomic number of 86 and has a half-life of 3.8 days. Because it is a noble gas, it does not react with other elements or compounds and therefore will not be bound up within the soils, therefore becoming free to move through the soil with the rate determined by the permeability of the soil. When ^{222}Rn decays, an alpha particle is emitted into the atmosphere and the atomic number changes by two, and atomic mass changes by four, to form Po-218 (Figure 2.5). Because the remainder of the daughter products are solids they may either be trapped within the rock or soil as they form or be mobilized into waters, if pH and/or Eh conditions are suitable. If an atom of radon forms at a grain boundary, then the radon may escape along the boundary, moving upwards into the soil and ultimately into the air, particularly if the rock is highly fractured, or already weathered and therefore quite permeable. (O'Beirne-Ryan, 2006)

^{222}Rn and its immediate decay products ^{218}Po (3.05 minutes) and ^{214}Po (164 microseconds), which also emit alpha particles, are among the major potential health issues in relation to the uranium decay series, as alpha particles can permanently damage DNA (US EPA, 2008). The most likely way in which these isotopes can enter the body is through inhalation of radon gas when we breathe. The health effects are attributed to the radon decay products ^{218}Po and ^{214}Po which are metals, chemically reactive and both have a half-life of less than 30 minutes (US EPA, 2008). It is breathing in radon, and the subsequent release of alpha particles, radon has become the second leading cause of lung cancer in North America, with smoking being the leading cause. Canadian Cancer Statistics 2007 indicated 19, 000 lung cancer deaths were expected for 2007 and 1, 900 of those lung cancer deaths are attributed to radon. In Canada, the current indoor air guideline is 200 Bq/m^3 for indoor air and levels of radon should be established by considering a series of readings over time, which are then averaged; it is this average value that should not exceed 200 Bq/m^3 which is the set level of risk determined by Health Canada, any radon level below this guideline is seen as acceptable (Health Canada, 2008). If the limit is reached or surpassed in a dwelling, the health risk significantly increases with the increased concentration and with increased time spent in an enclosed room with elevated radon (and its daughters) (US EPA, 2008).

2.5 Uranium and Radon Occurrences in Nova Scotia

Uranium occurs at a number of localities within Nova Scotia where the uranium levels are naturally elevated. Various companies and provincial organizations have carried out many surveys and exploration activity to determine the overall background levels of uranium in Nova Scotia. (O'Reilly, 1982). During the late 1970s and early 1980s there was an intense period of uranium exploration in the Province, following the identification of a number of localities with elevated uranium, as identified by a regional airborne radiometric survey (Ford, 1997). In the early 1980s, a moratorium on any further exploration or exploitation of uranium was emplaced, and this remains in place today. However, details of a number of regions with elevated levels of uranium and radon are included in assessment reports from the time.

A "uranium potential in groundwater" map was produced by the then Nova Scotia Department of Environment (1998), (Figure 2.7) and the Nova Scotia Department of Health also published a map outlining potential areas with elevated uranium occurrences in groundwater and potential occurrence of radon gas in

Nova Scotia (Figure 2.8). Both maps are based solely on the distribution of granitic and sedimentary rocks and do not actually provide uranium level data of specific areas, making the map generalized and not an accurate source for alerting the public to areas of concern. (Figure 2.6, Figure 2.7, Figure 2.8)

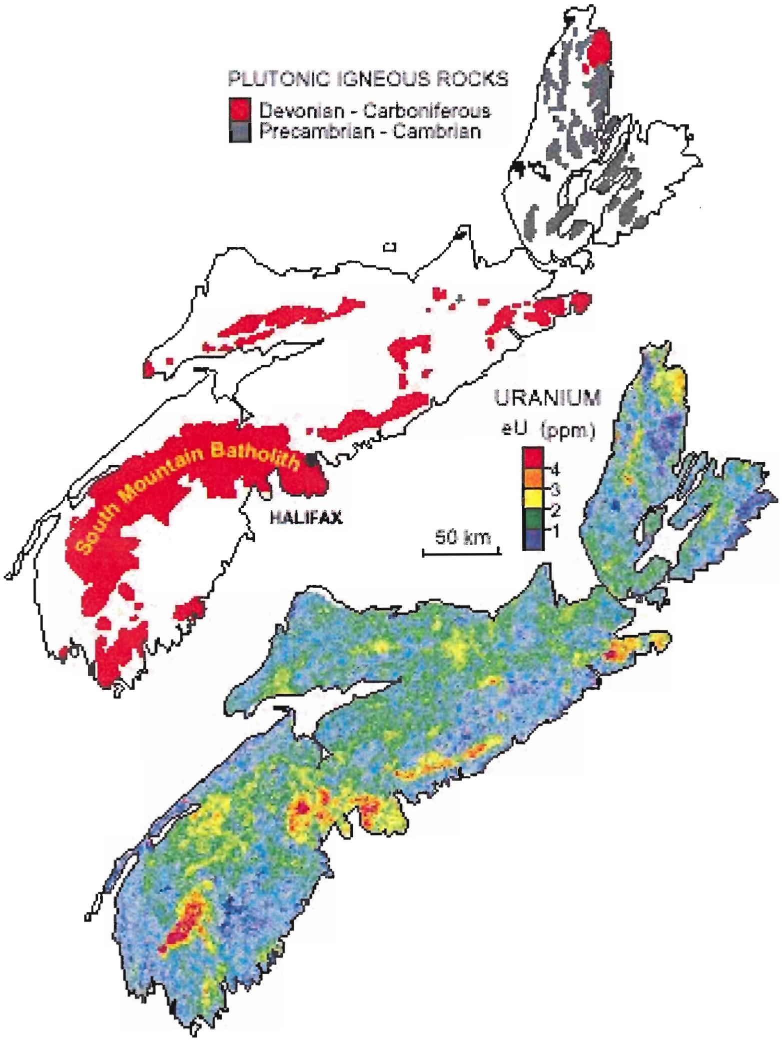

Figure 2.6 – Geological Survey of Canada Uranium map of Nova Scotia. (Geological Survey of Canada, 2007)

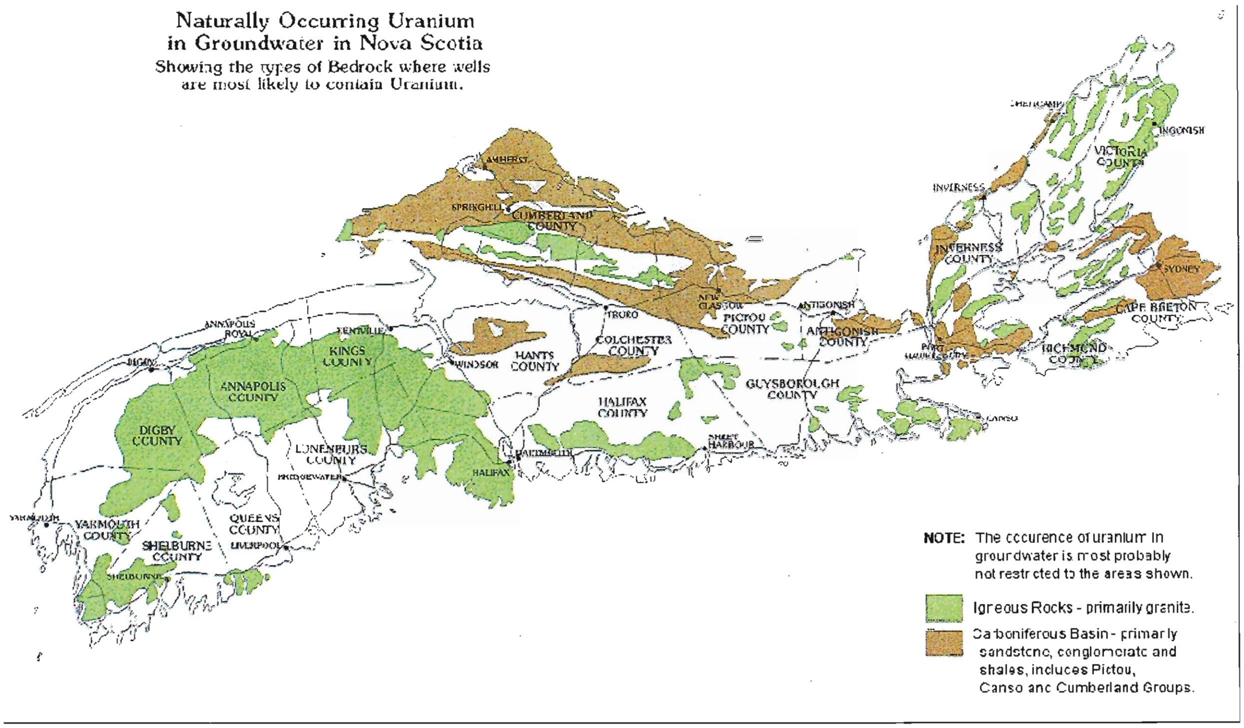

Figure 2.7 – Naturally Occurring Uranium in Groundwater in Nova Scotia. (Department of Environment, 1998)

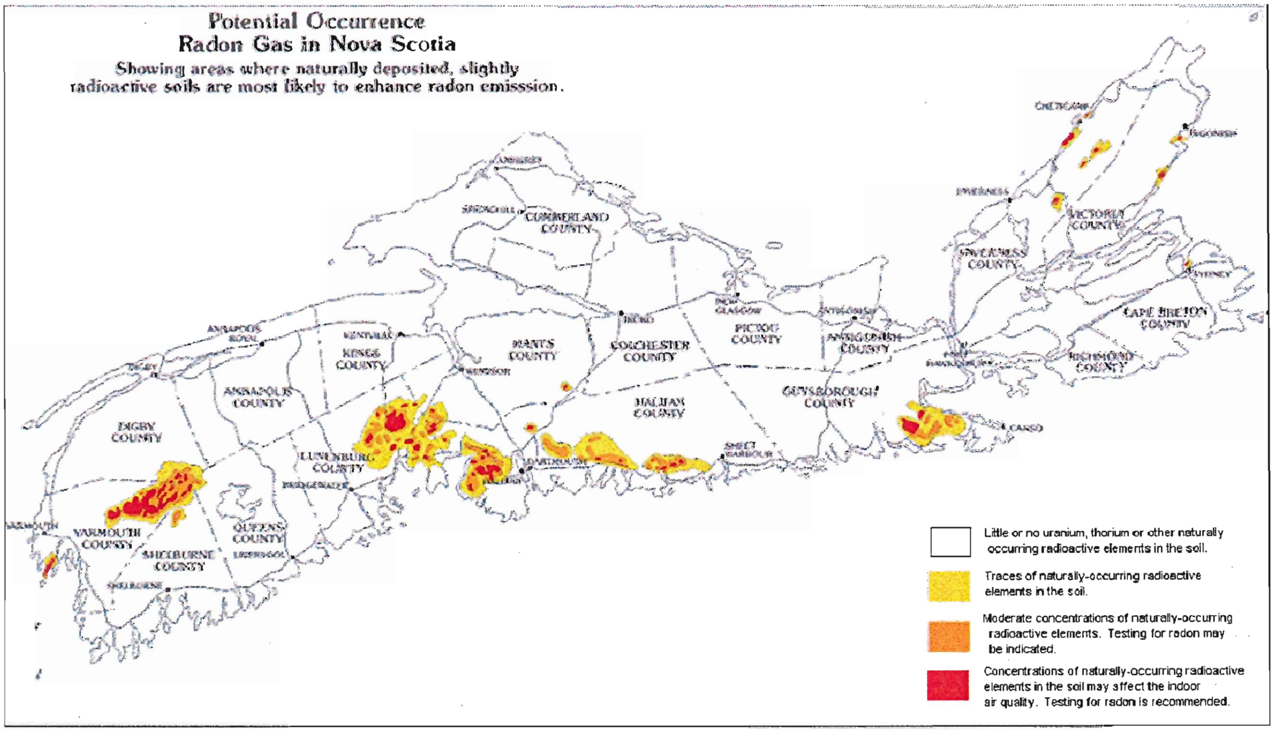

Figure 2.8 – Potential Occurrence Radon Gas in Nova Scotia. (Department of Environment, 1998)

2.6 Summary

Parsons (2007) conducted leaching experiments of the Horton Group which was from the Cheverie Formation from sample locality 1 of this study. Parsons (2007) study indicated a number of elements, including uranium migrated into the water, attributed to the weathering of the rock. Parsons (2007) also found slightly elevated levels of uranium in the waters from the leaching experiment, but radon was never measured, either in the soil or the water. Given that uranium was found to be present in the waters of the leaching experiments, it follows that radon should also be found in such waters, as long as there is equilibrium between the radon and the uranium. This project determines whether radon is indeed found in the waters resulting from weathering of rocks from the two sample locations, with the idea that as detection of radon in water is a simple and cost-efficient process, detecting its presence may be used as a proxy for uranium and/or radium in waters, which is more costly to determine. Radon in water is also a concern due to the transport and subsequent liberation of radon in soils creating potential radon indoor air concerns along with contamination of groundwater supplies.

3.0 Methods: Field Sampling and Experiments

3.1 Introduction

Throughout the duration of this investigation various methods were used to collect data, and the following section outlines the procedures involved. The main methods are field work and a series of laboratory experiments to gather ample evidence of the decay of uranium and/or radium to the daughter product radon gas in the weathered material selected. Rock samples were collected in the field and then used in various experiments to "artificially weather" the samples and subsequently analyze the resultant waters for radon and in select cases for general water chemistry and dissolved metals, including uranium.

3.2 Field Work

Rock samples were collected from two distinct locations near Windsor and New Ross, Nova Scotia on September 22, 2007. The locations were chosen based on elevated radiation counts measured by a handheld McPhar TVIA gamma spectrometer, and of outcrops of known geochemistry, both with and without elevated uranium concentration in the samples. (Table 1.0)

In total, roughly 20 kg of sample was collected (Table 1. 0). The organic portion of the samples were removed and the samples were allowed to air dry, then placed in bags, labelled and stored in a cool area while awaiting analysis. At each sample location (Figure 1.1, Figure 1.2) radon in soil gas was tested using a RAD7 detector which measures the radon in the soil overlying the outcrops, the results are present in Chapter 4.

3.3 DURRIDGE RAD7 Radon Detector

The RAD7 is a sniffer and a continuous monitor of both radon and thoron, which is radon-220. The RAD7 uses a solid state alpha detector which is a semiconductor material (usually silicon). The solid state detector converts the alpha radiation into an electrical signal. One of the important advantages of the solid state detector is the ability to determine the energy of each alpha particle therefore distinguishing different isotopes. (Durridge, 2006) When radon-222 decays the nucleus emits radiation in the form of alpha particles along with 5.49 MeV of energy. When radon-220 (thoron) decays it emits a 6.29MeV alpha particle and then decays into different decay daughters then that of radon-222. The RAD7 displays and prints out the data in radon-222 counts and radon-220 counts along with audible radon and thoron counts every time an alpha particle or energy is released (Durridge, 2006). The range of the RAD7 is 4 to >1 000 000 Bq/m^3 and with special attachments it is possible to test indoor air, soil gas, and radon in water. The upper detection limit is 1 000 000 Bq/m^3, so all values above this register as >1 000 000 Bq/m^3.

During the testing process the RAD7 tests for a total of 30 minutes. For the first 5 minutes the count rate increases then the counts begin to approach a steady level and after 10 minutes radon-222 has reached equilibrium with polonium-218, which is the daughter product of radon-222. When this equilibrium is reached the radon-222 level is stabilized, and a true radon-222 reading is reached. The RAD7 takes readings in 5 minutes increments over the 30 minute period and the Mean, Standard Deviation, and High/Low levels are recorded (Durridge, 2006).

In order to test the soil gas radon level there is a soil gas probe which is 30 centimetres in length. The probe has to be inserted in the soil and a reasonable seal between the probe and the surrounding soil must be created so no ambient air dilutes the soil gas sample. The probe must be connected to the RAD7 prior to starting a test, but before the test can be started the machine must be purged bringing the moisture level within the machine down to a level between 10-14% for the best results. In order to purge the machine, the input valve is connected to the filter as well as the output valve, so the air being used to purge is recycled. Once the machine is purged, the probe is reconnected to the RAD7 and the testing can begin (Durridge, 2006). (Figure 3.1)

The RAD-H20 is an accessory to the RAD7 that measures the radon in water levels with high accuracy and the detectable levels range from 370 Bq/m3 to >1 000 000 Bq/m^3 with measurements taken over 30 minutes with immediate results. With a maxim level of 1,000,000 Bq/m^3 levels were still categorized as being a problematic level of radon in water. (US EPA, 2008). Before starting the test the RAD7 should be purged for 10 minutes to remove any radon from the machine and to reach the appropriate humidity levels (10-14%).

Figure 3.1 – RAD7 with soil gas probe (Durridge, 2006).

Once ready to start the test, the water is measured into a 40mL vial filled to the top allowing no air space. The stainless steel aeration device provided is inserted into the water. The aeration system is then fed by tubing to a filter to collect any moisture from the water sample to ensure no water enters the RAD7. The tubing then goes from the filter to the RAD7 where the air is tested; the filter must be placed higher than the 40mL for the duration of the test to limit the amount of moisture entering the tubing. The radon levels are measured every 5 minutes but the counts are taken by the last four cycles after equilibrium has been reached and a summary reading is printed off after the 30 minute test is completed. Once the test is complete, the aeration system is unscrewed and the 40mL vial. If testing another sample allow the RAD7 to purge for 2 minutes in between the testing to remove any radon that might be left in the machine (Durridge, 2006) (Figure 3.2).

Figure 3.2 – RAD-H20 setup to RAD7 detector. (Durridge, 2006)

3.4 Radon in Water

To determine if there is a correlation between the uranium concentrations of the rock samples and the subsequent water used to weather the rock sample, through possible leaching, multiple suites of experiments were conducted using:

(a) distilled water at room temperature (20 °C)
(b) distilled water with a average temperature of 5°C
(c) distilled water at room temperature (20 °C) agitated every 24 hours
(d) river water at room temperature (20 °C)
(e) river water with an average temperature of 5°C

The resultant water was measured for radon using the RAD7 over increasing lengths of time. The longest period of time was chosen based on the length of ten half lives of radon-222 which is 3.8 days. Therefore the radon was tested over duration of up to 40 days.

The rock samples were broken to maximum grain size of 1 cm to increase surface area and were divided into 30g per 250mL sample jar. The samples were then filled to the top of the sample jars with no air space using 200mL of distilled water for one suite then a second suite was run using 200mL of river water which was collected on February 8th, 2008 in Windsor, Nova Scotia at the location of the Horton Group sample site 1 (Figure 1.1). The samples were contained in tightly sealed jars and prepared in multiple suites with 10 – 20 duplicates per suite.

The river water was tested for radon using the RAD7 prior to being utilized for the leaching experiments. The river water was also decanted from one suite with peak radon results and removed from the source for 3 half lives and the radon levels were measured after each half-life.

3.5 Water Analysis

There were a total of six water samples that were analyzed at Maxxam Analytics for general chemistry and available dissolved metals. The water tested included:

- River water prior to experiments
- Cold river water after 30 days of leaching from uranium enriched sandstone of the Horton Group (outcrop 1, Figure 1.1)
- Cold river water after 30 days of leaching from uranium enriched siltstone of the Horton Group (outcrop 1, Figure 1.1)
- Cold distilled water after 35 days of leaching from granite for Reeve's Road of South Mountain Batholith (Figure 1.2)
- Cold distilled water after 35 days of leaching from uranium enriched sandstone of the Horton Group (outcrop 1, Figure 1.1)
- Cold distilled water after 30 days of leaching from uranium enriched siltstone of the Horton Group (outcrop 1, Figure 1.1)

The water samples were those of the refrigerated suites and were taken at the peak of radon levels after 30-35 days. The water was immediately decanted from the samples and divided into two sample jars; a 200mL jar for general chemistry and a 50mL jar for dissolved metals. The sample jars were kept refrigerated until taken to Maxxam Analytics in Bedford, Nova Scotia, where they were filtered prior to analysis, so only dissolved metals and anions were measured. The methods used at Maxxam Analytics are those defined by the American Public Association and the U.S. EPA (Maxxam, 2005). The results of all the water analysis are presented in Chapter 4.

3.6 Quality Control

Quality control was a very important aspect of this study. In order to monitor this aspect of the data recorded steps were taken to measure duplicate samples to check for precision.

When conducting the radon in water tests, the same protocol was used every time. The machine was purged until the ideal humidity level was achieved and in that way, any radon remaining in the machine was removed. The aeration device was rinsed with distilled water to ensure there was no cross-contamination between samples. There were also systematic duplicates which were used at least three times during the testing process for each rock sample in each suite to ensure and provide confirmation of the precision of the RAD7 and gave affirmation of the precision used to preparing the samples prior to testing. When preparing the samples, the rock was measured using an electronic scale and the water was measured in a graduated cylinder to ensure each sample had the same amount of rock and water used in each sample.

When the samples were taken to Maxxam Analytics, they were subjected to the Maxxam Quality Assurance which consists of blind and double-blind checks sample programs and Corporate Standard QC sample frequency meaning the processing of Quality Control samples within each batch (1 per 20). Maxxam's facility undergoes an assessment by an external accrediting body to ensure that their policies and procedures are accredited. Maxxam is the Industry Leader for implementing the recognized Quality practices such as ISO 9002 standard.

4.0 Results

4.1 Field Observations

The sample location 1 (Figure 1.1) was in the area of Green Street located in Windsor, Nova Scotia. The outcrops at this location were accessible by a farming road with the outcrop visible in road side cuts (Figure 2.2). The overall topography of the area is gentle hills resulting in changing elevations between outcrops along the roadside over a section approximately 200 meters in length. The main outcrop where mineralized sandstone and mineralized siltstone was collected is adjacent to Sam's Brook where the river water used for testing was collected on February 8, 2008 (Figure 1.1). Sam's Brook runs South/North along the road. Significant observations at both sites in the Green Street area include the intense oxidation and weathering that has occurred within some horizons. Both outcrops have experienced erosion that is evident by the overhanging nature of the cliff face and weathered material that was found loose at the base of the cliff and along the roadside. Siltstone occurs as lenses centimetres thick within the sandstone units. A contributing factor to the potential redistribution of uranium and its daughters is the stream that runs along the west side of the outcrop. The stream bed cuts approximately five meters into the outcrop (Figure 4.1). The water level of this stream was low at the time of sampling the rock; upon return in February 2008 the water level had increased significantly from 10-20 centimetres to a depth of 30-40 centimetres and doubled in width, following a period of rain and snow. The soil gas level of radon was measured on September 22, 2007 and the results are represented in Table 4.1. The sample outcrops were measured using a handheld spectrometer that yielded a higher response over regions with elevated uranium levels. Given the presence of radon gas in the overlying soil, the higher levels of uranium are consistent with higher levels of radon gas that is being produced. Typical background levels of gamma radiation in the area yield scintillometer readings between 4,000 and 6,000 counts per minute. Readings of the uranium enriched horizons from the Horton Group ranged from 70,000 to >100,000 counts per minute.

Figure 4.1 – Sam's Brook located 5 meters along the west side of outcrop 1, sample location 1 (Green Street, near Windsor, Nova Scotia)

The second outcrop location was in the surrounding area of Morton Reeves Road located in New Ross, Nova Scotia (Figure 1.2). This single outcrop location was accessible by an old wood road that was heavily over grown with vegetation. The outcrop was five meters from the road side but due to vegetation growth, the outcrop is difficult to see from the road. This granitic outcrop was small in area (10-15m^2) and was the only outcrop visible along the old wood road, less than a kilometer from the Forties Road. The overall topography of the area is relatively flat, with an average elevation of 200 meters. An important observation was the intense amount of weathering that was evident at this outcrop. This granite outcrop was so severely weathered that the rock crumbles when placed in your hand, and is readily classed as a saprolite (Figure 2.4b). There was no water source in the area, although the ditch between the outcrop and the road was boggy which suggests poor drainage (Figure 2.4a). The granite outcrop has a relatively low uranium concentration of 5ppm consistent with non-mineralized areas in the South Mountain Batholith, but when soil gas radon levels were tested the area showed significant levels of radon, averaging at 4800 Bq/m^3 which was comparable to that of the results found of the Horton Group sample location. The handheld spectrometer gave readings on the low uranium samples from the Horton Group ranged from 10, 000 – 20, 000 counts per minute, five times above the background levels. The South Mountain Batholith yielded spectrometer readings were in between the low uranium outcrop of the Horton Group and the enriched outcrop of the Horton Group (Table 4.1).

Table 4.1 – The radon levels measured at the Green Street outcrop in Windsor, NS are measured in Bq/m^3 at a temperature of 24 ^{0}C with relative humidity at 9%. The radon levels measured at the Reeves Road outcrop in New Ross, NS are measured in Bq/m3 at a temperature of 23 ^{0}C with relative humidity at 11.5%. Measurements were taken in five minute cycles.

RADON IN SOIL		
Time (mins)	**Green Street, Location 1**	**Reeves Road, Location 2**
	Radon	**Radon**
5	5700 +/- 957	2580 +/-676
10	6220 +/- 999	4800 +/-887
15	6630 +/- 1030	4230 +/- 834
20	6290 +/- 1010	5180+/-923
25	6180 +/- 1000	6180 +/-1000
30	5000 +/- 915	5840+/-973
35	6010 +/- 989	
40	5830 +/- 981	
Average	**5982**	**4802**

4.2 Whole Rock Geochemistry

Geochemical analysis was carried out on the Horton Group sandstone and siltstone previously by Parsons, 2007 and on the Reeve's granite by O'Beirne-Ryan pers-com., 2008. Parsons completed a geochemical analysis on samples taken from the Green Street area outcrops consisting of two sandstone samples and two siltstone samples (Table 4.2a). These samples were taken from the loose, weathered debris from the base of location one adjacent to the stream. O'Beirne-Ryan completed a geochemical analysis on the weathered granitic sample from the Reeve's Road outcrop (Table 4.2b). A full suite of data was obtained and the results are presented in the following sections which include major element analysis and trace element analysis (Parsons, 2007).

Table 4.2a Geochemical Analysis of Horton Group Sandstone and Siltstone (after Parsons, 2007)

SAMPLE	SiO2	TiO2	Al2O3	Fe2O3t	MnO	MgO	CaO	Na2O	K2O	P2O5	LOI
AP06-02 Siltstone	59.93	1	22.63	3.16	0.02	0.71	0.04	0.18	3.74	0.09	8.4
AP06-03 Sandstone	77.36	0.08	10.58	4.4	0.02	0.14	0.04	0.16	3.84	0.09	3.3
AP06-04 Siltstone	60.74	0.9	22.33	3.02	0.03	0.68	0.02	0.16	3.77	0.1	8
AP06-05 Sandstone	78.75	0.17	12.06	1.13	0.04	0.16	0.02	0.17	3.58	0.07	3.7

SAMPLE	Ba	Rb	Sr	Y	Zr	Nb	Th	Pb	Ga	Zn	Cu
AP06-02 Siltstone	286	150.5	88	33.5	181.3	22.1	13	88.1	45.7	41	95.5
AP06-03 Sandstone	245	84.7	74	74.5	24.5	2.6	2.5	27.1	11.2	6.3	30.5
AP06-04 Siltstone	223	135.7	198	11.4	177.8	19.6	16.2	38.7	34.9	56.3	51.5
AP06-05 Sandstone	214	98	62	7.3	44.8	4.4	5.3	43.1	13.4	14.6	31.2

SAMPLE	Ni	V	Cr	Hf	Sc	Ta	Co	Be	U
AP06-02 Siltstone	5	159	43	6.2	10	3	8.3	29	212
AP06-03 Sandstone	<5	34	7	0.9	2	0.5	21.1	3	39.7
AP06-04 Siltstone	12	106	27	6.4	10	2.9	12.1	7	56.2
AP06-05 Sandstone	<5	53	6	1.6	4	0.9	31.9	4	109

Table 4.2b Geochemical Analysis of Reeves Road Granite (O'Beirne-O'Ryan, pers comm., 2008).

SAMPLE	SiO2	TiO2	Al2O3	Fe2O3t	FeO	MnO	MgO	CaO	Na2O	K2O	P2O5	LOI	H2Op	Total
128 mild weathering	72.1	0.282	14.55	2.33	1.17	0.024	0.37	0.72	2.87	5.56	0.213	0.99	0.74	100.01
129 moderate weathering	72.95	0.24	13.76	2.26	1.16	0.026	0.33	0.47	2.48	5.57	0.166	1.03	0.67	99.28

SAMPLE	V	Cr	Co	Zr	Ba	La	Nd	Ni	Cu	Zn	Ga	Rb	Sr	Y	Nb	Pb	Th	U
128 mild weathering	38	12	1	110	688	20	21	3	6	73	19	232	73	22	10	17	5	5
129 moderate weathering	34	8	1	107	446	17	22	1	5	74	19	248	43	29	10	18	3	4

4.3 Radon in Water

During the leaching process samples were divided into two major sets (Table 4.3); one set using distilled water and the other using river water collected from Sam's Brook, Green Street, Windsor, Nova Scotia. These various suites were further split by temperature to mimic seasonal changes; one suite was maintained at room temperature of 20^0C while the other suite was kept at a cooler temperature of 5^0C. A third suite was maintained at room temperature but was continuously agitated (20 repetitions) on a daily basis to stimulate a heavy rainstorm to increase the rate of erosion.

4.3.1 Radon in River Water

The river water used in the leaching process had an initial pH reading of 5.6, and an initial radon reading of >1 000 000Bq/m^3. The river water was used in two suites of leaching experiments, each using three rock samples: uranium-enriched sandstone, and uranium-enriched siltstone of location one, and the granite of location 2. One suite was maintained at room temperature (20°C) (Table 4.3a) whereas the second suite was maintained at a cooler temperature (5°C) (Table 4.3b). The timeframe was chosen based on eight complete half lives of radon to ensure an ample amount of time to monitor the nature of change in radon over a number of half lives. With a half-life of 3.8 days the timeframe used was between 30-40 days. River water was also decanted from each type of source rock once the radon levels peaked. The decanted water was placed in 3 40mL containers filled to the top, and tested after 3 full half lives to determine whether radon levels dropped through time. This suite was completed using the three rock types found in Table 4.3b, the results for the decanted water can be found in Table 4.3c.

Table 4.3a

RADON IN RIVER WATER UNSHAKEN ROOM TEMPERATURE (20^{0}C)			
	Sample Type		
	DF-07-02	DF-07-01	DF-07-05
Time	Uranium enriched Sandstone	Uranium enriched Siltstone	Reeves Granite
2 days	>1 000 000*	>1 000 000*	>1 000 000*
4 days	>1 000 000	>1 000 000	>1 000 000
6 days	516 000	610 000	587 000
8 days	521 000	717 000	691 000
10 days	677 000	912 000	913 000
13 days	663 000	701 000	681 000
16 days	787 000	937 000	941 000
20 days	>1 000 000*	>1 000 000*	>1 000 000*
23 days	>1 000 000	>1 000 000	>1 000 000
27 days	>1 000 000	>1 000 000	>1 000 000
30 days	125 000*	312 000*	547 000*
34 days	250 000	347 000	490 000

*Duplicate samples taken, mean of results was used for data.

Table 4.3b

RADON IN RIVER WATER UNSHAKEN REFRIDGERATED (5^{0}C)			
	Sample Type		
	DF-07-02	DF-07-01	DF-07-05
Time	Uranium enriched Sandstone	Uranium enriched Siltstone	Reeves Granite
2 days	>1 000 000 *	>1 000 000*	>1 000 000*
4 days	>1 000 000	>1 000 000	>1 000 000
6 days	423 000	566 000	533 000
8 days	694 000	807 000	885 000
10 days	915 000	910 000	927 000
13 days	535 000	673 000	695 000
16 days	901 000	>1 000 000	925 000
20 days	>1 000 000*	>1 000 000*	>1 000 000*
23 days	>1 000 000	>1 000 000	>1 000 000
27 days	>1 000 000	>1 000 000	>1 000 000
30 days	509 000*	480 000*	822 000*
34 days	497 000	495 000	788 000

* Duplicate samples taken, mean of results was used for data.

Table 4.3c

RADON IN RIVER WATER UNSHAKEN ROOM TEMPERATURE (2^0C) Decanted from Source Rock			
	Sample Type		
	DF-07-02	DF-07-01	DF-07-05
Time	Uranium enriched Sandstone	Uranium enriched Siltstone	Reeves Granite
0 days	677 000	912 000	913 000
4 days	966 000	>1 000 000	993 000
8 days	>1 000 000	>1 000 000	>1 000 000
16 days	940 000	944 000	950 000

4.3.2 Radon in Distilled Water

The distilled water was initially assumed to be pure water and therefore no testing was conducted prior to the leaching experiments. The distilled water was used for multiple suites including all rock samples, cold and warm temperatures and agitation. The timeframe varied between 30 – 40 days for each suite based on the results of the previous suite. The radon levels were elevated after one half-life which was the starting measurement of the first suite conducted, the following suites began the measurements prior to one half-life, after 12 hours – 2 days for the remaining suites. The results follow in Table 4.3d – Table 4.3g.

Table 4.3d

RADON IN DISTILLED WATER UNSHAKEN ROOM TEMPERATURE (20^{0}C)			
	Sample Type		
	DF-07-02	DF-07-01	DF-07-05
Time	Uranium enriched Sandstone	Uranium enriched Siltstone	Reeves Granite
8 days	83 000	422 000	3200
10 days	754 000*	558 000	605 000
11 days	656 000*	>1 000 000*	>1 000 000*
12 days	>1 000 000*	>1 000 000	>1 000 000
14 days	846 000*	> 1 000 000	859 000
16 days	820 000*	464 000*	155 000*
21 days	389 000*	985 000	266 000
29 days	318 000*	950 000*	>1 000 000*
33 days	847 000	>1 000 000	>1 000 000

* Duplicate samples taken, mean of results was used for data

Table 4.3e

RADON IN DISTILLED WATER UNSHAKEN REFRIDGERATED (5^{0}C)			
	Sample Type		
	DF-07-02	DF-07-01	DF-07-05
Time	Uranium enriched Sandstone	Uranium enriched Siltstone	Reeves Granite
12 hours	239 000	326 000	306 000
24 hours	238 000	312 000	287 000
2 days	545 000	325 000	345 000
3 days	579 000*	456 000*	384 000*
5 days	511 000	412 000	402 000
8 days	745 000	756 000	792 000
10 days	> 1 000 000*		
13 days	598 000		
16 days	>1 000 000	741 000	610 000
20 days	808 000*	>1 000 000*	>1 000 000*
23 days	833 000	>1 000 000	>1 000 000
27 days	741 000	>1 000 000	>1 000 000
30 days	>1 000 000*	>1 000 000	>1 000 000
34 days	>1 000 000	>1 000 000 *	>1 000 000*
38 days	949 000	873 000	793 000

* Duplicate samples taken, mean of results was used for data

Table 4.3f

RADON IN DISTILLED WATER UNSHAKEN REFRIDGERATED (5^{0}C)		
	Sample Type	
	DF-07-04	DF-07-03
Time	Low Uranium Sandstone	Low Uranium Siltstone
4 days	334 000	314 000
7 days	>1 000 000*	>1 000 000*
10 days	>1 000 000	921 000
12 days	508 000	543 000
14 days	664 000	687 000
16 days	967 000	>1 000 000
19 days	>1 000 000*	>1 000 000*
23 days	986 000	962 000
27 days	973 000	907 000
30 days	875 000*	776 000*
34 days	791 000	739 000

* Duplicate samples taken, mean of results was used for data

Table 4.3g

RADON IN DISTILLED WATER UNSHAKEN ROOM TEMPERATURE (20^{0}C)		
	Sample Type	
	DF-07-04	DF-07-03
Time	Low Uranium Sandstone	Low Uranium Siltstone
4 days	366 000	350 000
7 days	>1 000 000	>1 000 000
10 days	789 000	>1 000 000
12 days	575 000	531 000
14 days	881 000	746 000
16 days	891 000	742 000
19 days	>1 000 000*	>1 000 000*
23 days	947 000	951 000
27 days	>1 000 000	>1 000 000
30 days	607 000*	612 000*
34 days	702 000	707 000

* Duplicate samples taken, mean of results was used for data

Table 4.3h

RADON IN DISTILLED WATER SHAKEN ROOM TEMPERATURE (20^{o}C)			
	Sample Type		
	DF-07-02	DF-07-01	DF-07-05
Time	Uranium enriched Sandstone	Uranium enriched Siltstone	Reeves Granite
1 day	609 000	622 000	600 000
4 days	266 000	512 000	508 000
6 days	561 000	>1 000 000	910 000
8 days	>1 000 000*	>1 000 000*	>1 000 000*
10 days	868 000	917 000	627 000
13 days	499 000*	>1 000 000*	>1 000 000*
16 days	>1 000 000	>1 000 000	980 000
20 days	736 000	>1 000 000	>1 000 000
23 days	367 000	340 000	604 000
27 days	804 000	978 000	>1 000 000
30 days	839 000*	>1 000 000*	>1 000 000*
34 days	>1 000 000*	>1 000 000*	>1 000 000
36 days	924 000	>1 000 000	>1 000 000
38 days	525 000	794 000	734 000

* Duplicate samples taken, mean of results was used for data

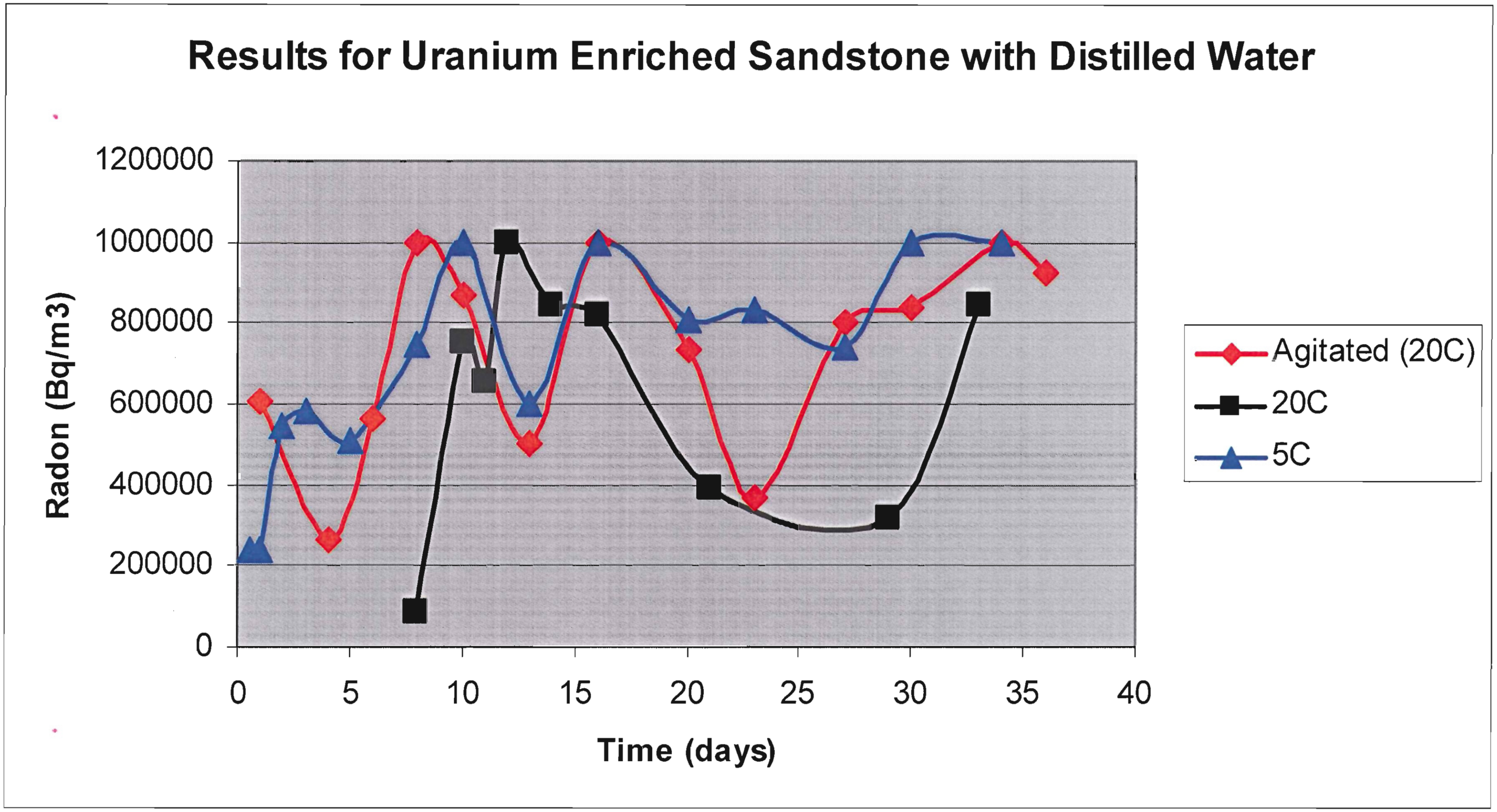

Figure 4.3(a) Uranium Enriched Sandstone – Distilled water suites

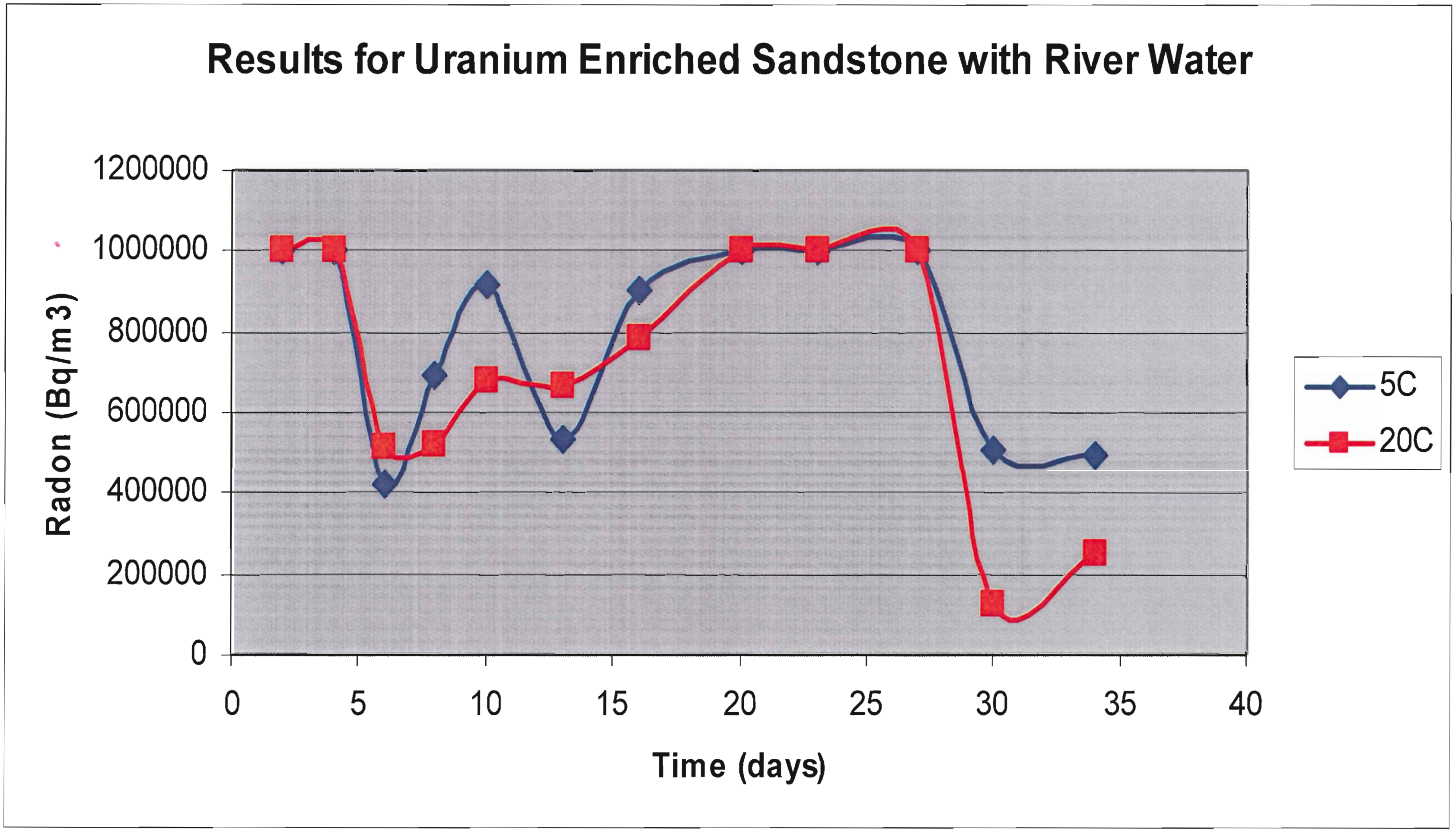

Figure 4.3 (b) Uranium Enriched Sandstone – River water suites

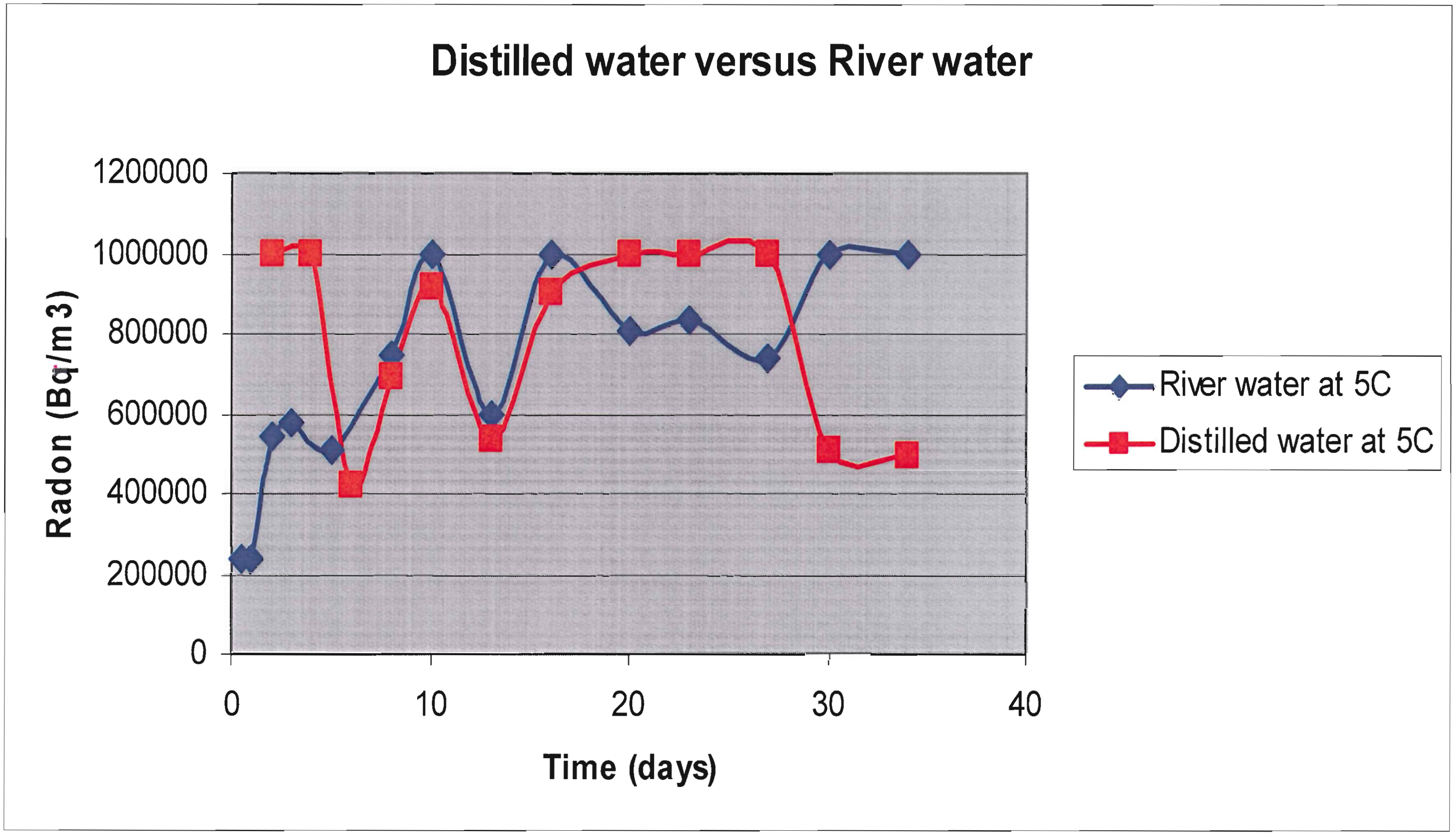

Figure 4.3(c) Uranium Enriched Sandstone – Cold distilled water versus cold river water

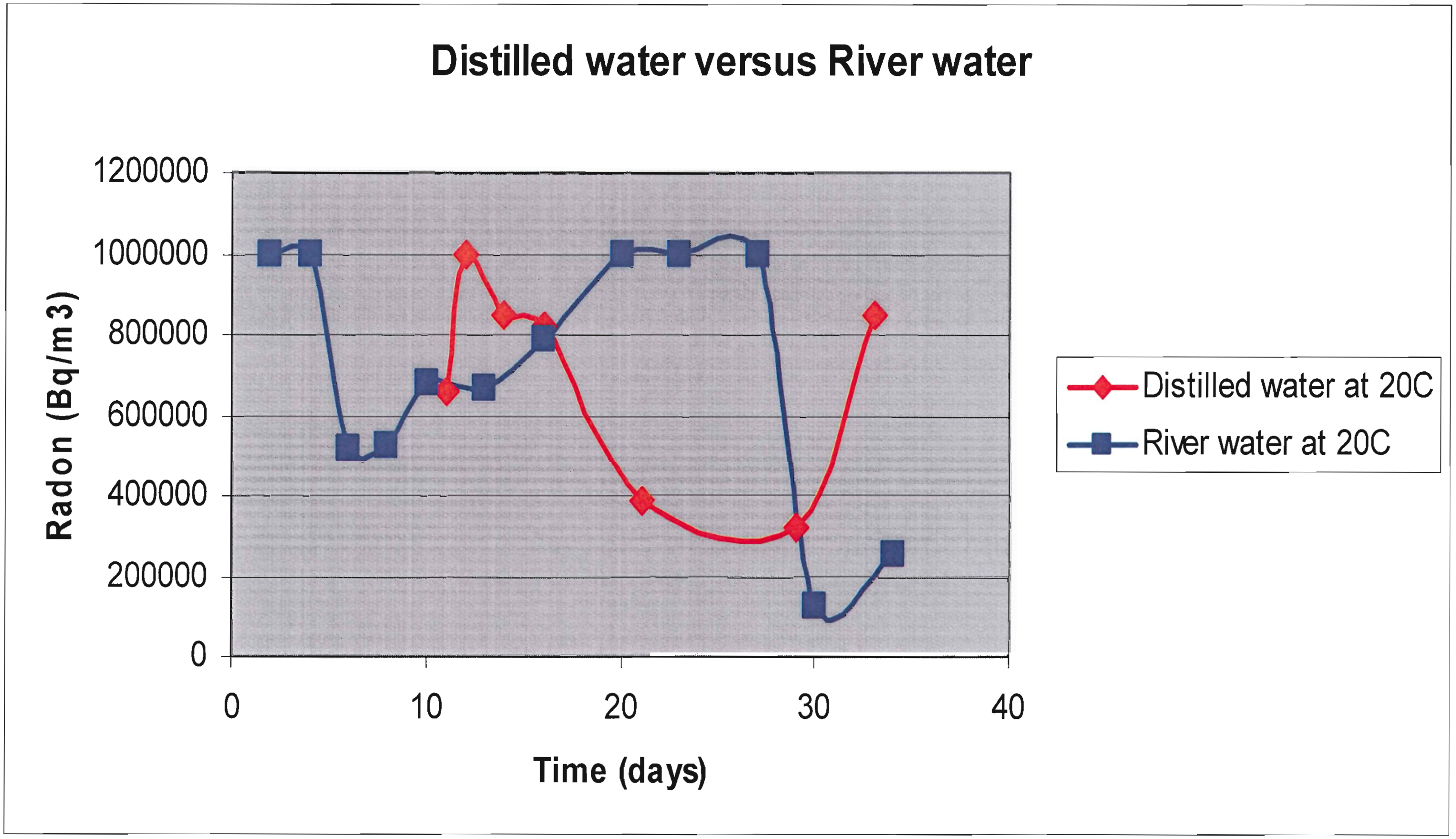

Figure 4.3(d) Uranium Enriched Sandstone – Distilled water versus river water at room temperature

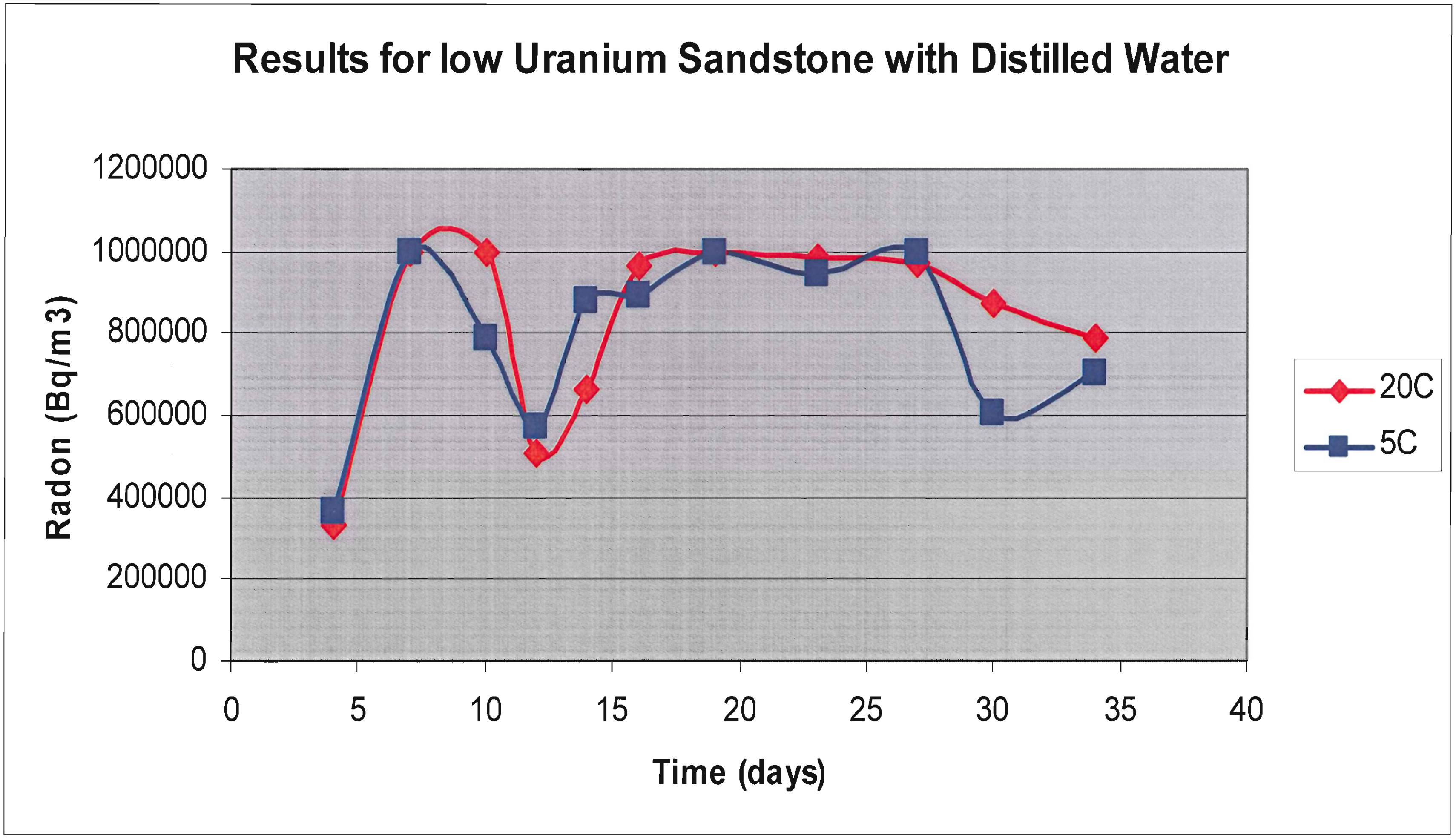

Figure 4.3(e) Low Uranium Concentration Sandstone – Distilled water suites

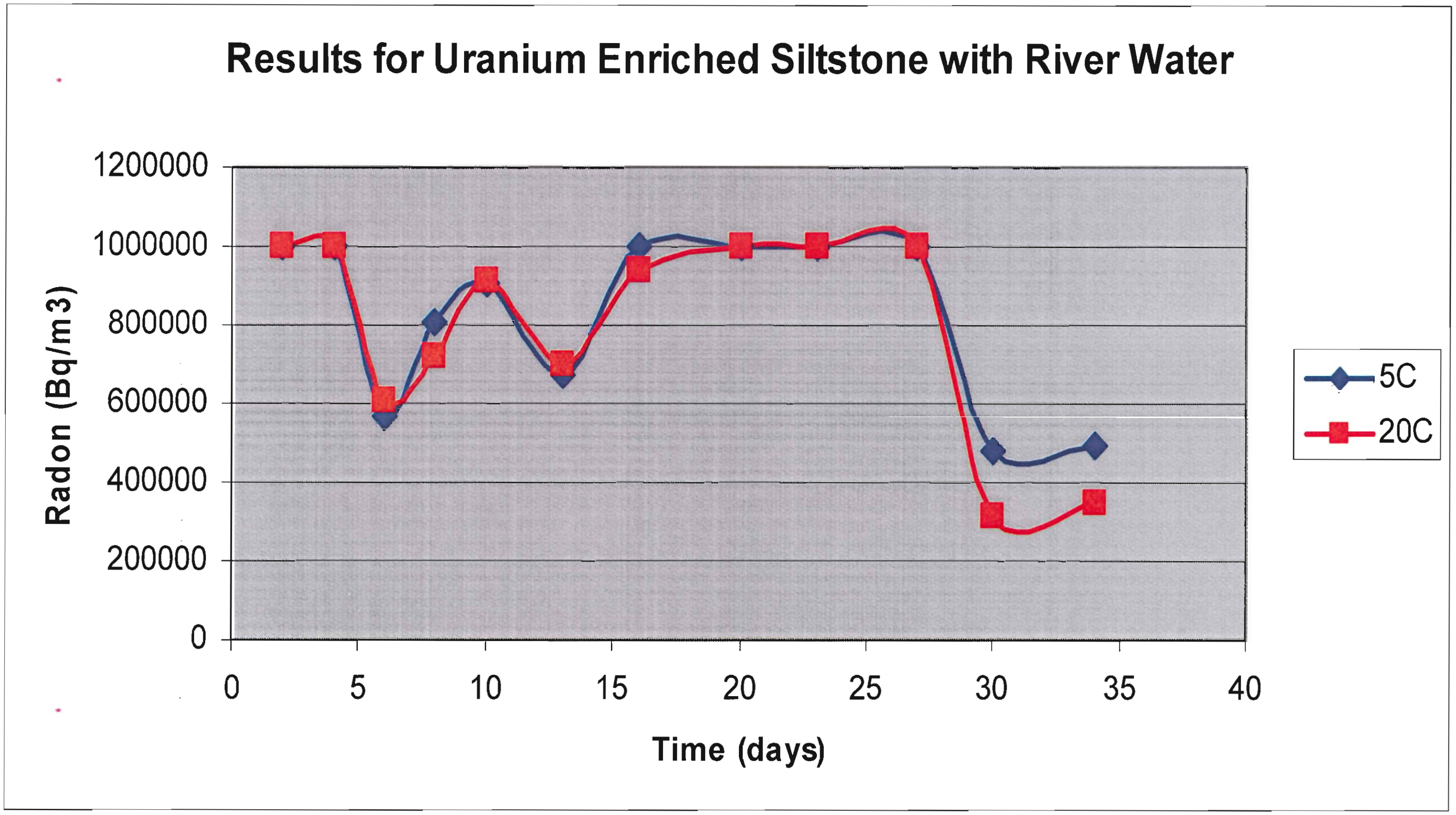

Figure 4.3(f) Uranium Enriched Siltstone – River water suites

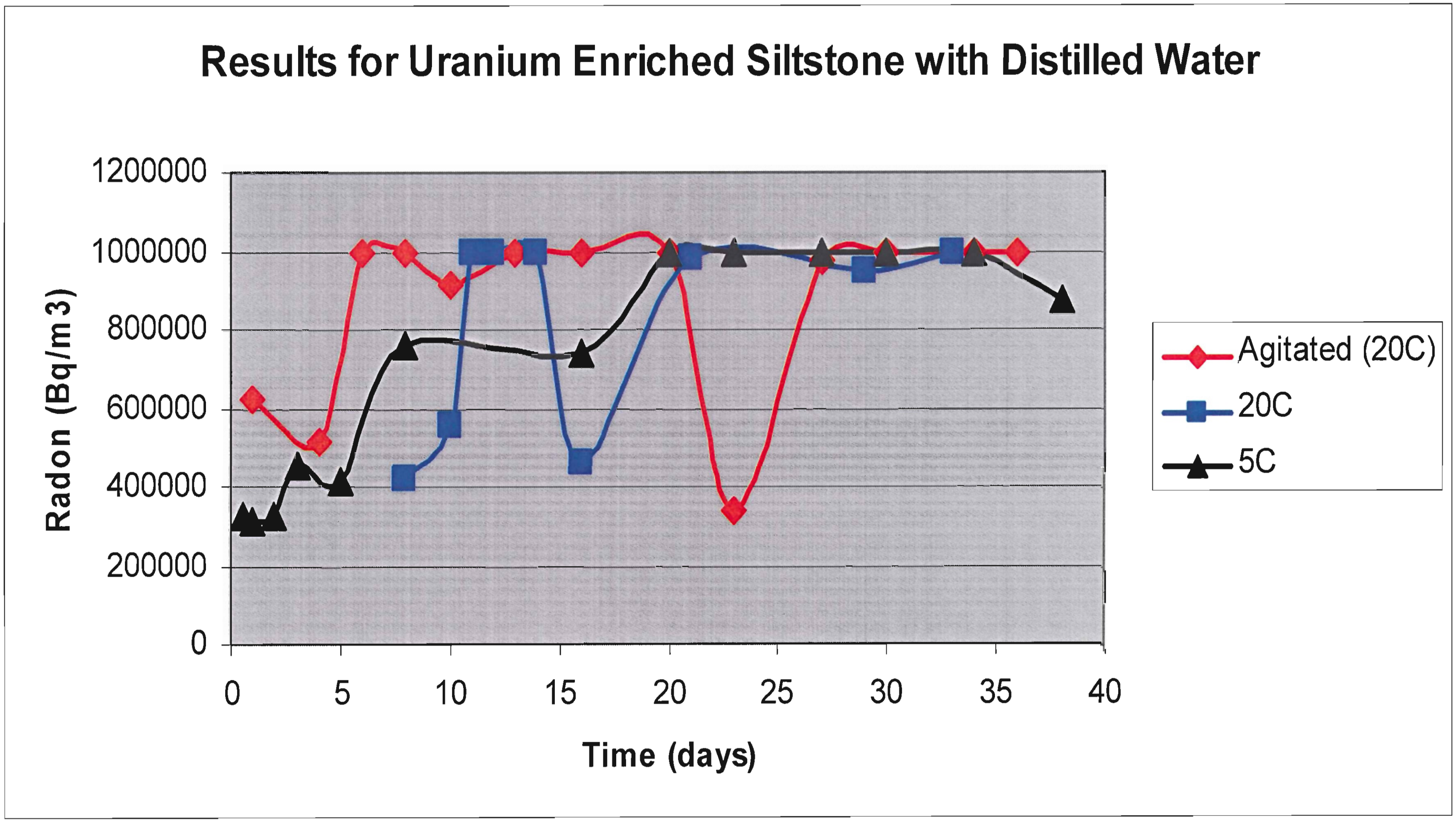

Figure 4.3(g) Uranium Enriched Siltstone – Distilled water suites

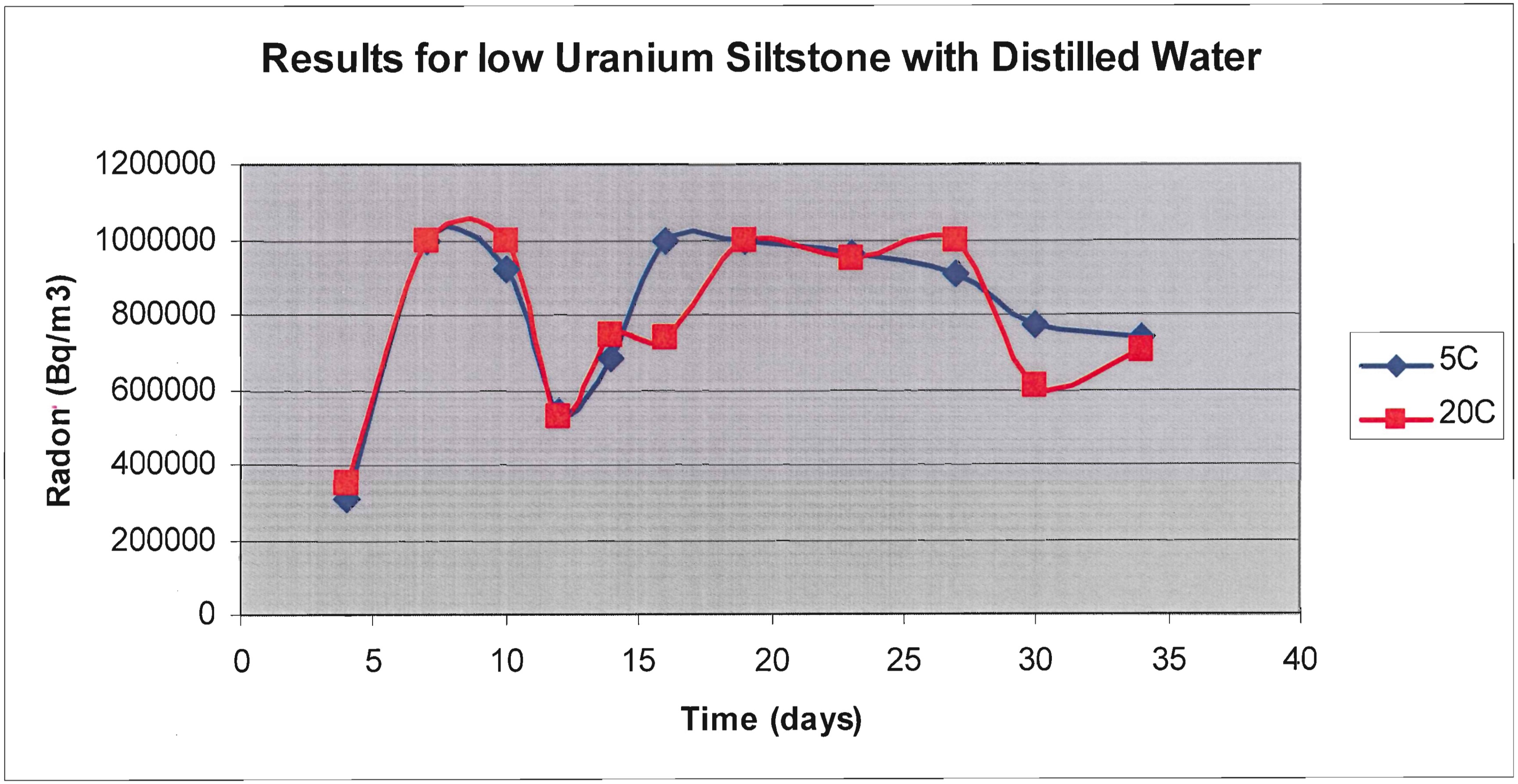

Figure 4.3(h) Low uranium concentration siltstone

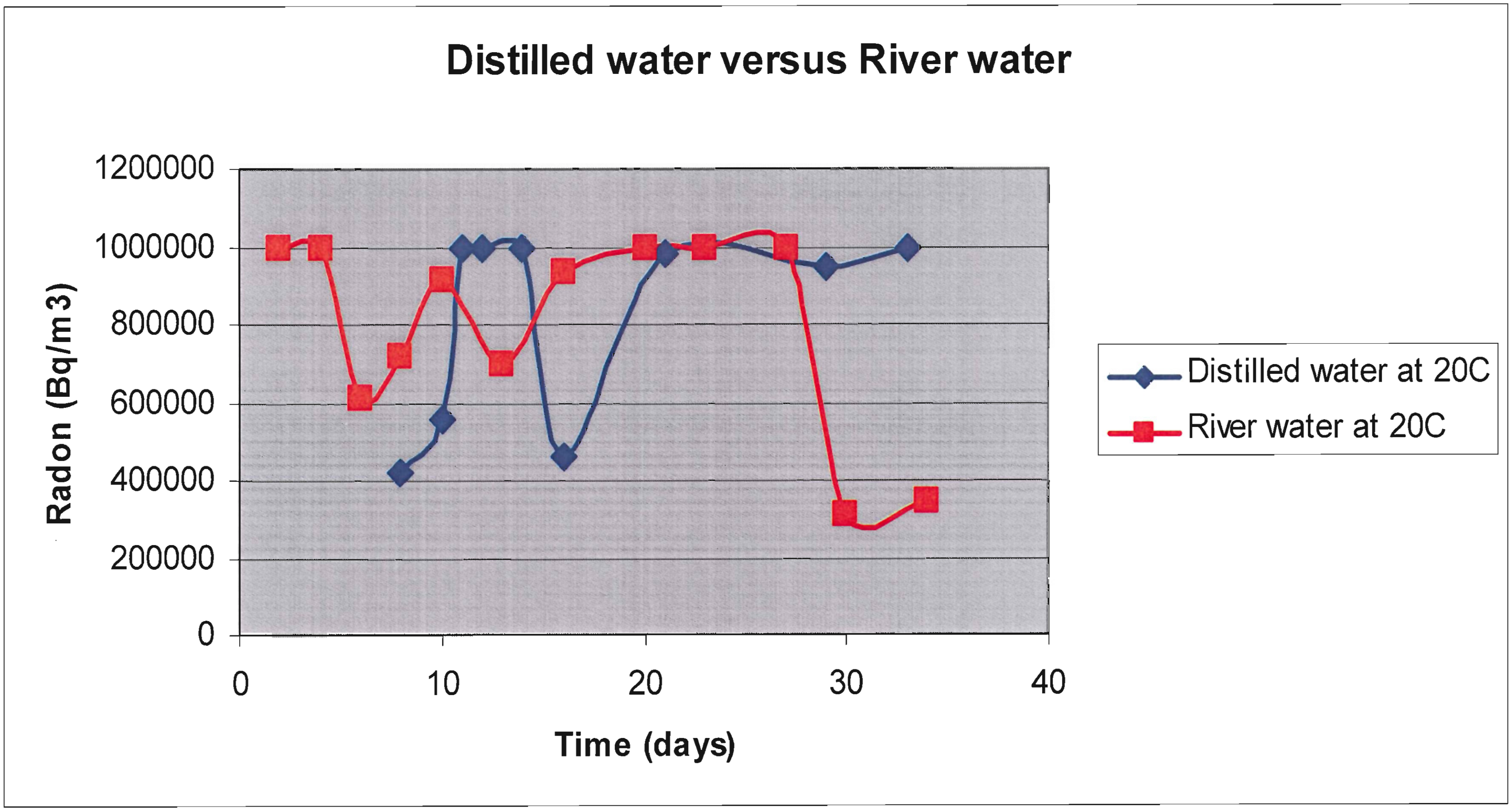

Figure 4.3(i) Uranium Enriched Siltstone – Distilled water versus river water at room temperature

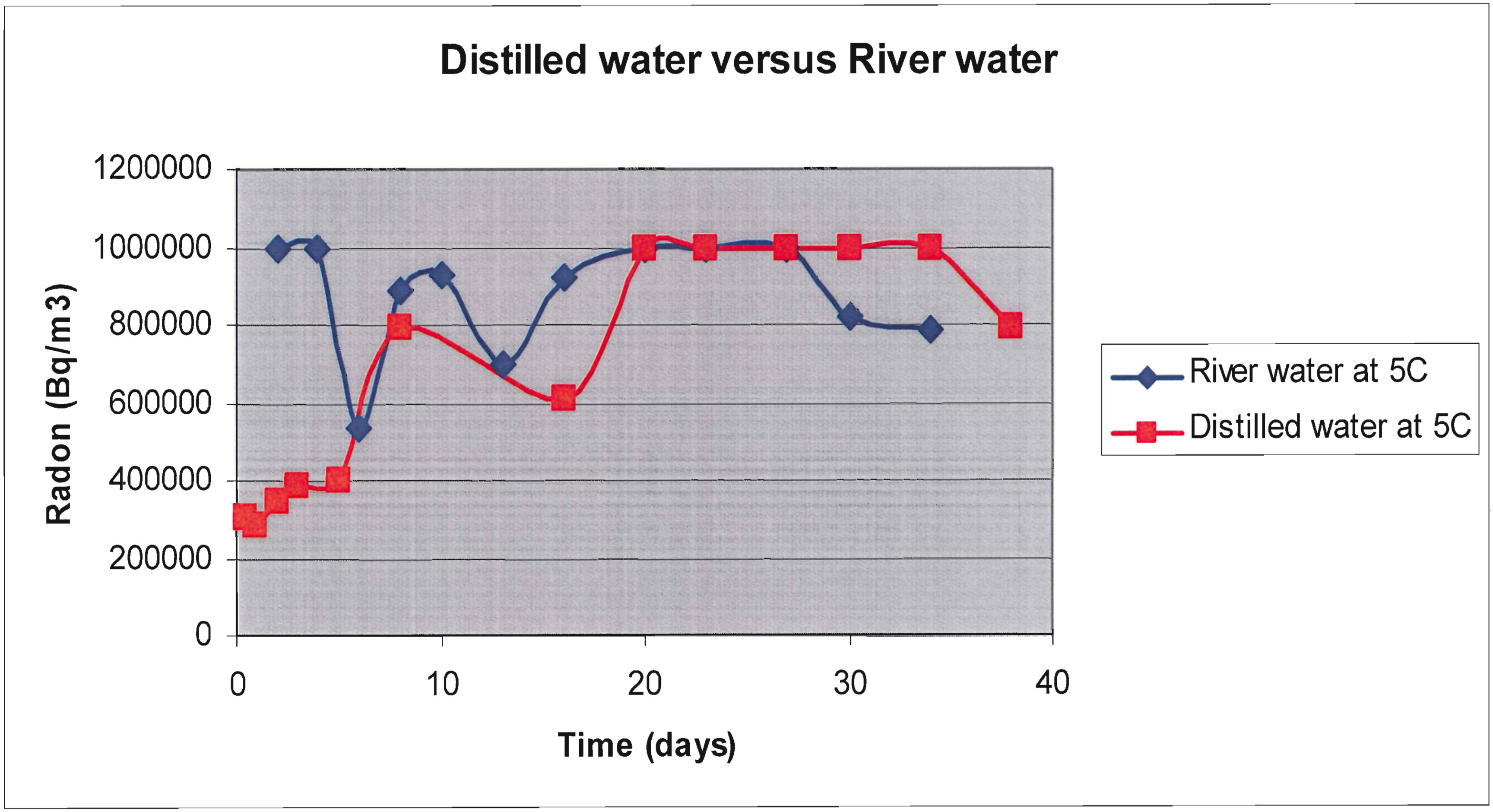

Figure 4.3(j) Uranium Enriched Siltstone – Distilled water versus river water at cold temperatures

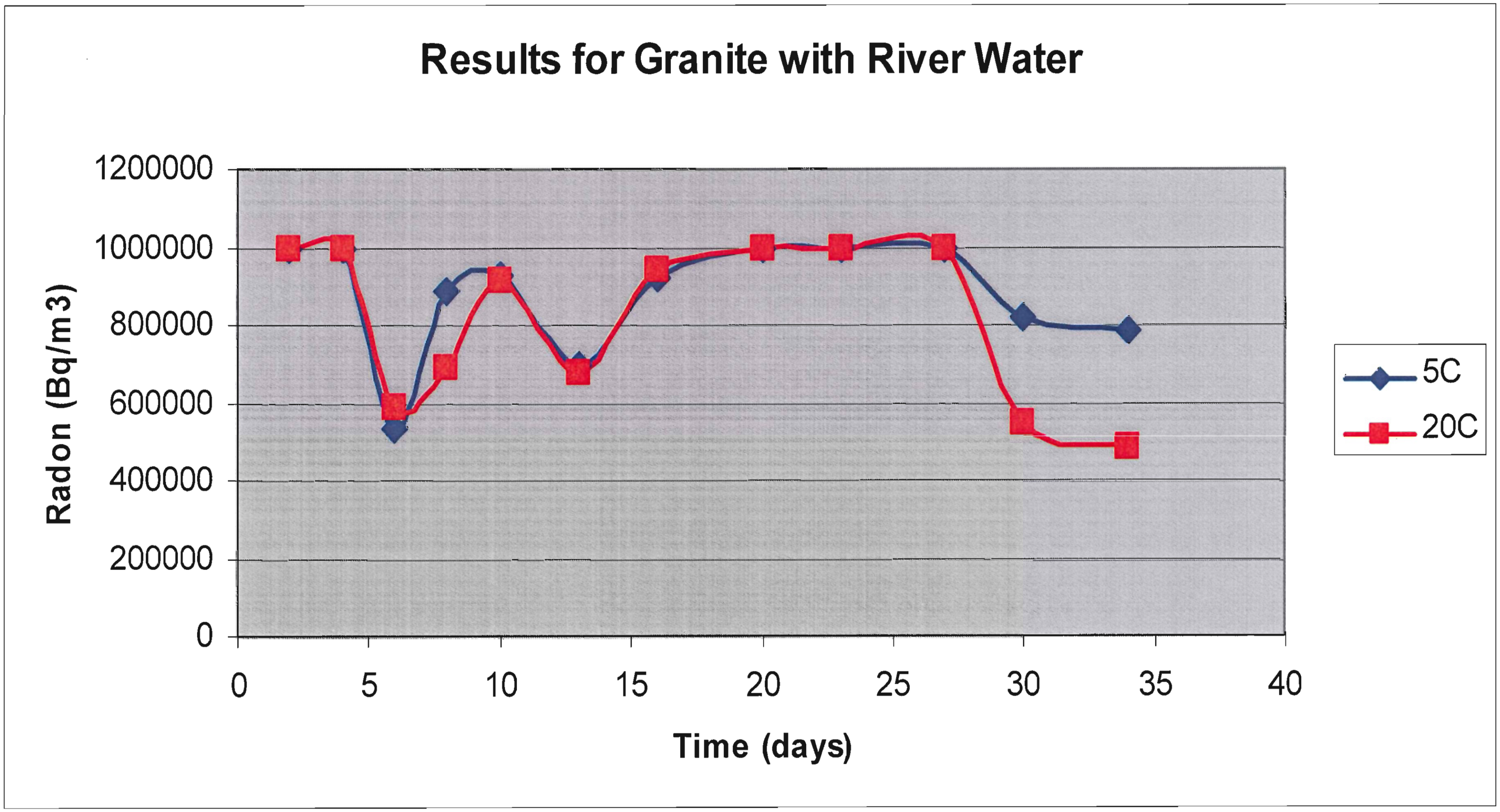

Figure 4.3(k) Reeves Road Granite – River water suites

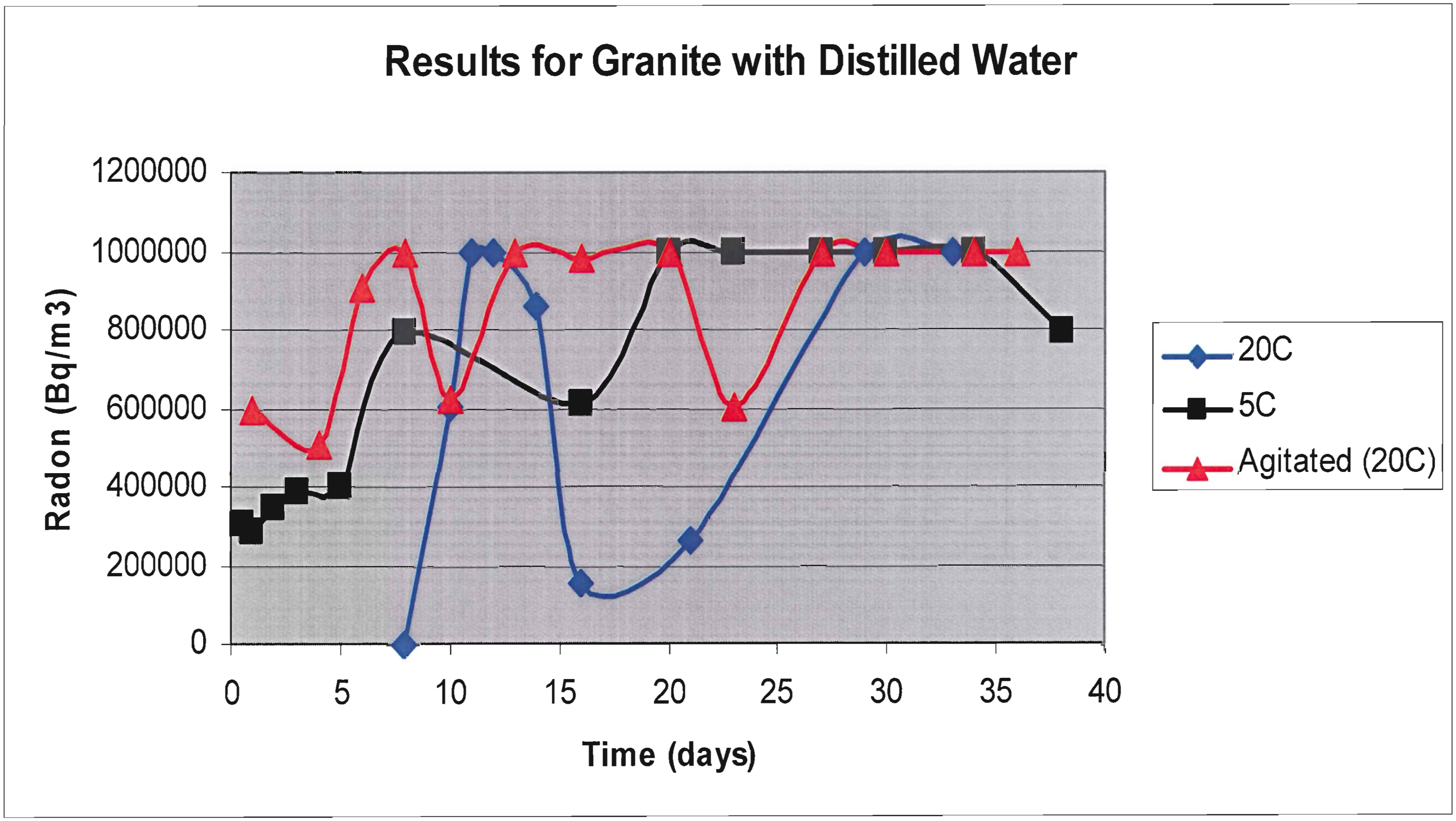

Figure 4.3(l) Reeves Road Granite – Distilled water suites

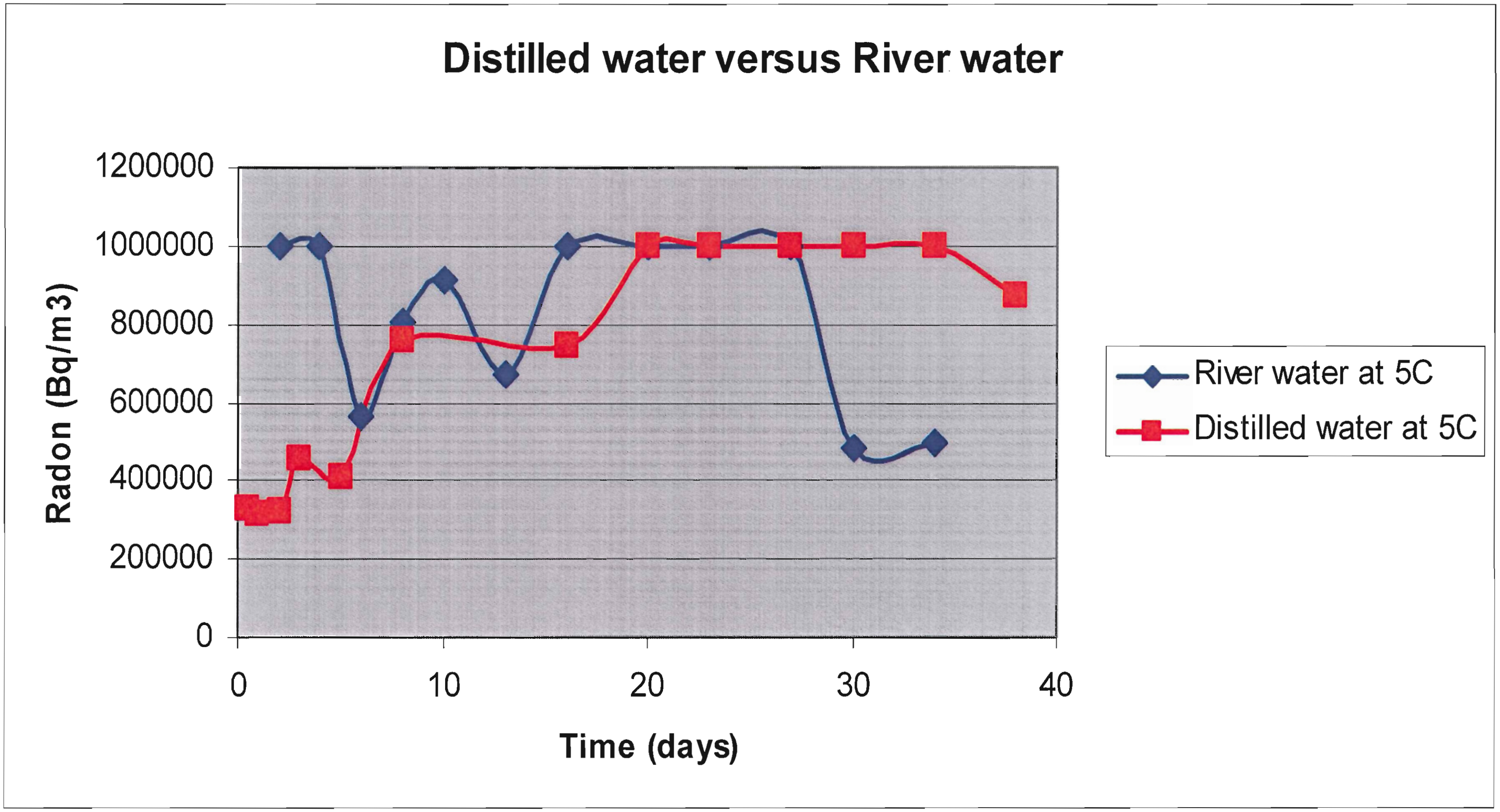

Figure 4.3(m) Reeves Road Granite – Distilled water versus river water at cold temperatures

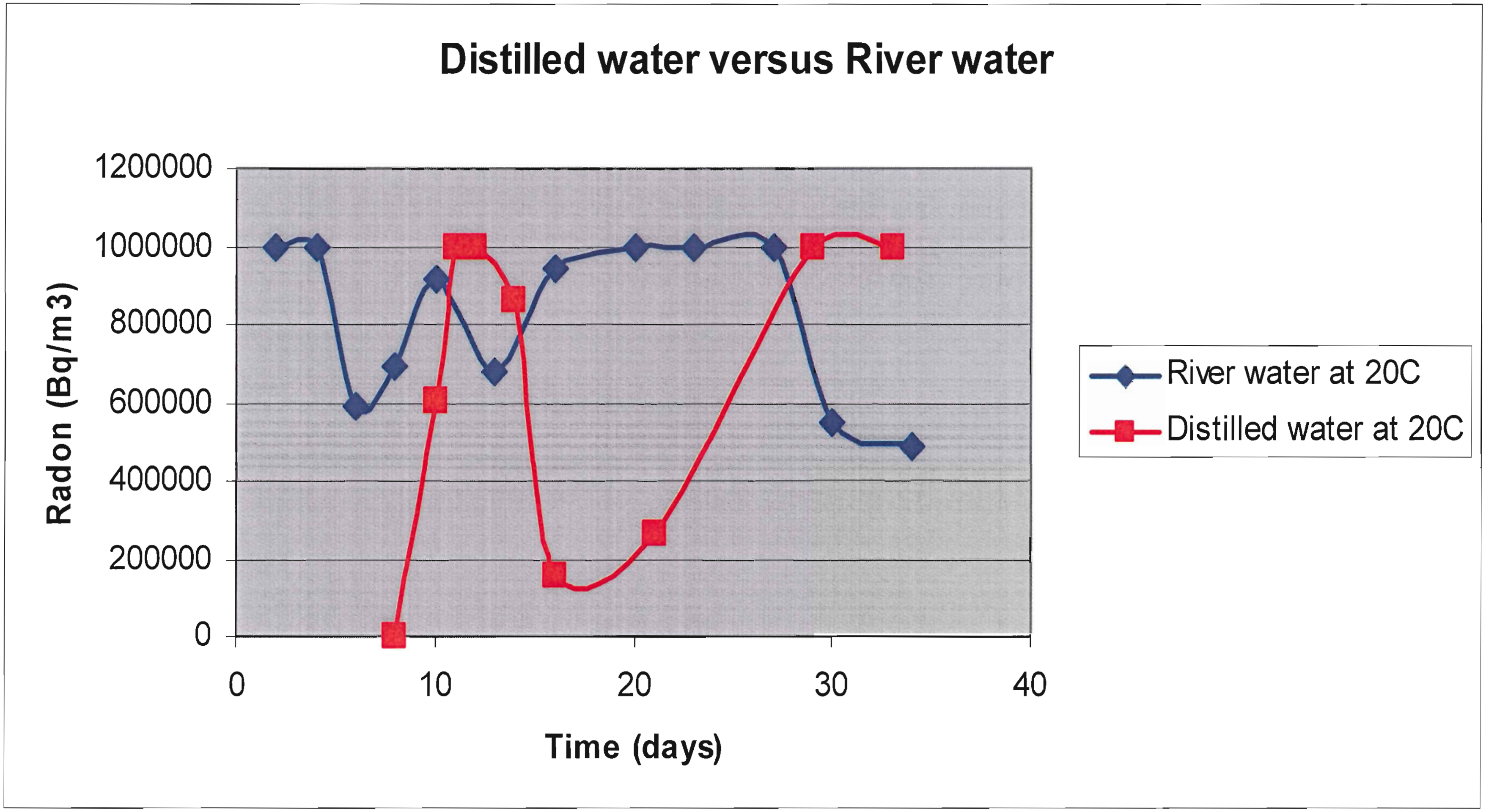

Figure 4.3(n) Reeves Road Granite – Distilled water versus river water at room temperature

4.4 General Water Chemistry

There were six water samples submitted to Maxxam Analytic Inc., in Bedford, NS for general chemistry and available dissolved metals. The river water was submitted prior to the leaching experiment to determine the initial general chemistry and initial dissolved metals (Table 4.4a). The distilled water was not submitted prior to the leaching experiments; it was assumed that the distilled water was pure and contained no dissolved metals. Maxxam require the water samples to be cold (between 4°C and 10°C) and therefore the water samples chosen for analysis preceding the leaching experiments were all suites that were kept cold. The two leached river water samples that were analyzed by Maxxam were from the cold suite using uranium-enriched sandstone and uranium-enriched siltstone. The three leached distilled water samples that were analyzed by Maxxam included uranium-enriched sandstone, uranium-enriched siltstone, both of the Green Street and the low uranium granite of Reeves Road. The water was decanted from the samples once peak radon levels had been reached and were immediately submitted to Maxxam Analytic Inc., in Bedford for analysis. The analysis was completed to determine the amount of uranium and other elements leached from the weathering rock into the water.

The leaching experiments attempted to simulate typical eroding processes such as rainstorms and seasonal temperature changes. By testing the water following the leaching period, it is possible to determine if uranium (or indeed, radium) is being leached from the source rock into the water then subsequently decaying into radon within the water.

Table 4.4a Atlantic RCAp-MS dissolved metals in water – Analysis of river water prior to leaching experiments.

	Units	RIVER WATER (INTIAL)	RDL
Calculated Parameters			
Anion Sum	me/L	0.1	N/A
Bicarb. Alkalinity (calc. as CaCO3)	mg/L	ND	1
Calculated TDS	mg/L	12	1
Carb. Alkalinity (calc. as CaCO3)	mg/L	ND	1
Cation Sum	me/L	0.27	N/A
Hardness (CaCO3)	mg/L	8	1
Ion Balance (% Difference)	%	46	N/A
Langelier Index (@ 20C)	N/A	NC	N/A
Langelier Index (@ 4C)	N/A	NC	N/A
Nitrate (N)	mg/L	ND	0.05
Saturation pH (@ 20C)	N/A	NC	N/A
Saturation pH (@ 4C)	N/A	NC	N/A
INORGANICS			
Total Alkalinity (Total as CaCO3)	mg/L	ND	5
Dissolved Chloride (Cl)	mg/L	3	1
Colour	TCU	23	5
Nitrate + Nitrite	mg/L	ND	0.05
Nitrite (N)	mg/L	ND	0.01
Nitrogen (Ammonia Nitrogen)	mg/L	ND	0.05
Total Organic Carbon (C)	mg/L	3.3	0.5
Orthophosphate (P)	mg/L	ND	0.01
pH	pH	6.51	N/A
Reactive Silica (SiO2)	mg/L	3.3	0.5
Dissolved Sulphate (SO4)	mg/L	ND	2
Turbidity	NTU	ND	0.1
Conductivity	uS/cm	28	1

Elements (ICP-MS)			
Dissolved Aluminum (Al)	µg/L	81	10
Dissolved Antimony (Sb)	µg/L	ND	2
Dissolved Arsenic (As)	µg/L	ND	2
Dissolved Barium (Ba)	µg/L	6	5
Dissolved Beryllium (Be)	µg/L	ND	2
Dissolved Bismuth (Bi)	µg/L	ND	2
Dissolved Boron (B)	µg/L	ND	5
Dissolved Cadmium (Cd)	µg/L	ND	0.3
Dissolved Chromium (Cr)	µg/L	ND	2
Dissolved Cobalt (Co)	µg/L	ND	1
Dissolved Copper (Cu)	µg/L	ND	2
Dissolved Iron (Fe)	µg/L	ND	50
Dissolved Lead (Pb)	µg/L	ND	0.5
Dissolved Manganese (Mn)	µg/L	ND	2
Dissolved Molybdenum (Mo)	µg/L	ND	2
Dissolved Nickel (Ni)	µg/L	ND	2
Dissolved Selenium (Se)	µg/L	ND	2
Dissolved Silver (Ag)	µg/L	ND	0.5
Dissolved Strontium (Sr)	µg/L	8	5
Dissolved Thallium (Tl)	µg/L	ND	0.1
Dissolved Tin (Sn)	µg/L	ND	2
Dissolved Titanium (Ti)	µg/L	ND	2
Dissolved Uranium (U)	µg/L	ND	0.1
Dissolved Vanadium (V)	µg/L	ND	2
Dissolved Zinc (Zn)	µg/L	ND	5
Elements (ICP-OES)			
Dissolved Calcium (Ca)	mg/L	2	0.1
Dissolved Magnesium (Mg)	mg/L	0.6	0.1
Dissolved Phosphorus (P)	mg/L	ND	0.1
Dissolved Potassium (K)	mg/L	0.4	0.1
Dissolved Sodium (Na)	mg/L	2.5	0.2

ND = Not Detected
RDL = Reportable Detection Limit

Table 4.4b Atlantic RCap-MS dissolved metals in water – Analysis of leached water.

	Units	RIVERWATER (URANIUM-ENRICHED SANDSTONE)	RIVERWATER (URANIUM- ENRICHED SILTSTONE)	DISTILLED (GRANITE)	DISTILLED (URANIUM-ENRICHED SANDSTONE)	DISTILLED (URANIUM-ENRICHED SILTSTONE)	RDL
Calculated Parameters							
Anion Sum	me/L	0.15	0.15	0	0	0.05	N/A
Bicarb. Alkalinity (calc. as CaCO3)	mg/L	ND	ND	ND	ND	ND	1
Calculated TDS	mg/L	14	19	1	2	7	1
Carb. Alkalinity (calc. as CaCO3)	mg/L	ND	ND	ND	ND	ND	1
Cation Sum	me/L	0.2	0.29	0.04	0.04	0.09	N/A
Hardness (CaCO3)	mg/L	3	6	ND	ND	ND	1
Ion Balance (% Difference)	%	14.3	31.8	100	100	28.6	N/A
Langelier Index (@ 20C)	N/A	NC	NC	NC	NC	NC	N/A
Langelier Index (@ 4C)	N/A	NC	NC	NC	NC	NC	N/A
Nitrate (N)	mg/L	ND	ND	ND	ND	ND	0.05
Saturation pH (@ 20C)	N/A	NC	NC	NC	NC	NC	N/A
Saturation pH (@ 4C)	N/A	NC	NC	NC	NC	NC	N/A
INORGANICS							
Total Alkalinity (Total as CaCO3)	mg/L	ND	ND	ND	ND	ND	5
Dissolved Chloride (Cl)	mg/L	3	3	ND	ND	ND	1
Colour	TCU	12	14	ND	ND	ND	5
Nitrate + Nitrite	mg/L	ND	ND	ND	ND	ND	0.05
Nitrite (N)	mg/L	ND	ND	ND	ND	ND	0.01
Nitrogen (Ammonia Nitrogen)	mg/L	ND	ND	ND	ND	ND	0.05
Total Organic Carbon (C)	mg/L	3.2	3.6	1	1.2	2.2	0.5
Orthophosphate (P)	mg/L	ND	0.24	0.04	ND	0.11	0.01
pH	pH	5.8	6.25	6.27	5.92	5.67	N/A
Reactive Silica (SiO2)	mg/L	3.8	5.7	0.5	0.9	2.2	0.5
Dissolved Sulphate (SO4)	mg/L	3	3	ND	ND	3	2
Turbidity	NTU	11	4.7	2.5	18	27	0.1
Conductivity	uS/cm	24	32	3	4	10	1

Elements (ICP-MS)	Units	RIVERWATER (URANIUM-ENRICHED SANDSTONE)	RIVERWATER (URANIUM- ENRICHED SILTSTONE)	DISTILLED (GRANITE)	DISTILLED (URANIUM-ENRICHED SANDSTONE)	DISTILLED (URANIUM-ENRICHED SILTSTONE)	RDL
Dissolved Aluminum (Al)	ug/L	120	170	27	21	100	10
Dissolved Antimony (Sb)	ug/L	ND	ND	ND	ND	ND	2
Dissolved Arsenic (As)	ug/L	ND	ND	ND	ND	ND	2
Dissolved Barium (Ba)	ug/L	10	11	ND	ND	ND	5
Dissolved Beryllium (Be)	ug/L	ND	ND	ND	ND	ND	2
Dissolved Bismuth (Bi)	ug/L	ND	ND	ND	ND	ND	2
Dissolved Boron (B)	ug/L	5	16	ND	ND	10	5
Dissolved Cadmium (Cd)	ug/L	ND	ND	ND	ND	ND	0.3
Dissolved Chromium (Cr)	ug/L	ND	ND	ND	ND	ND	2
Dissolved Cobalt (Co)	ug/L	3	1	ND	ND	ND	1
Dissolved Copper (Cu)	ug/L	3	3	ND	ND	ND	2
Dissolved Iron (Fe)	ug/L	ND	ND	ND	ND	ND	50
Dissolved Lead (Pb)	ug/L	ND	1	ND	ND	1.2	0.5
Dissolved Manganese (Mn)	ug/L	49	16	ND	4	13	2
Dissolved Molybdenum (Mo)	ug/L	ND	ND	ND	ND	ND	2
Dissolved Nickel (Ni)	ug/L	ND	ND	ND	ND	ND	2
Dissolved Selenium (Se)	ug/L	ND	ND	ND	ND	ND	2
Dissolved Silver (Ag)	ug/L	ND	ND	ND	ND	ND	0.5
Dissolved Strontium (Sr)	ug/L	ND	6	ND	ND	ND	5
Dissolved Thallium (Tl)	ug/L	ND	ND	ND	ND	ND	0.1
Dissolved Tin (Sn)	ug/L	ND	ND	ND	ND	ND	2
Dissolved Titanium (Ti)	ug/L	ND	5	ND	ND	11	2
Dissolved Uranium (U)	ug/L	5	22	0.4	1.6	7.8	0.1
Dissolved Vanadium (V)	ug/L	ND	ND	ND	ND	ND	2
Dissolved Zinc (Zn)	ug/L	ND	ND	ND	ND	ND	5
Elements (ICP-OES)							
Dissolved Calcium (Ca)	mg/L	0.5	1.5	ND	ND	0.1	0.1
Dissolved Magnesium (Mg)	mg/L	0.3	0.4	ND	ND	ND	0.1
Dissolved Phosphorus (P)	mg/L	ND	0.2	ND	ND	0.1	0.1
Dissolved Potassium (K)	mg/L	1	1.6	0.2	0.4	1	0.1
Dissolved Sodium (Na)	mg/L	2.9	3.3	0.7	0.6	1.2	0.1

4.4.1 Sandstone

There were two sets of sandstone sample rock used; one sample was uranium enriched sandstone with a uranium concentration of 109 ppm and the other sample was sandstone, having a lower uranium concentration of 39 ppm (Parsons, 2007). Despite the difference in the initial uranium concentration in the samples, radon was detected in the leaching waters at levels of >1,000,000 Bq/m^3 in both sets of samples. The radon levels within each suite seemed to change over a similar timeframe. Samples that underwent more intense artificial weathering through daily agitation had radon levels that were somewhat erratic (Figure 4.3a) whereas samples that were not agitated and were maintained at a constant temperature increased and decreased on similar patterns.

(Table 4.3, Figure 4.3(a-e))

4.4.2 Siltstone

There were also two sets of siltstone samples that were used in the leaching experiments. One of the siltstone samples was uranium enriched with a uranium concentration of 212 ppm, whereas the other siltstone sample used had a lower uranium concentration of 57 ppm (Parsons, 2007). As in the sandstone samples, the difference in the uranium level in the rock had little effect on the radon levels measured in the leached water, as both water samples contained radon levels in excess of 1,000,000 Bq/m^3. As in the sandstone samples, agitation was used in one suite to simulate erosion and speed up weathering. The results measured were somewhat erratic, unlike the samples in which the temperature was kept constant, and the resultant waters showed similar patterns of increasing and decreasing values over time. (Table 4.3, Figure 4.3(f-j))

4.4.3 Granite

The granite sample from the Morton Reeve's Road had a lower uranium concentration (4ppm) compared to all four samples of the other two rock types. However, the level of radon that was measured in the water from the leaching was equivalent to that of the two previous rock types (Table 4.3, Figure 4.3(k-n)). The granite sample yielded comparable radon levels to the other rock types. The granite suites also had comparable fluctuations in results when the samples were agitated and similar patterns in the samples that were under controlled temperature conditions. The colder samples exhibited more rapid response to radon release, however the peaks levels are similar in both cases. (Figure 4.3(m))

4.5 Summary

All three rock types were consistent in having elevated radon-in-water levels indicating leaching from the sample source rocks. The data collected has identified some key points that will be discussed further in Chapter 5. The significant points are as follows:

(1) In the leaching experiments initial uranium concentrations in the rock have no correlation with the final peak radon in water levels.

(2) The agitated suite had more erratic fluctuations in radon results compared to suites where no agitation was conducted.

(3) Suites using river water had more consistent results between 5°C and 20°C temperature then distilled water suites under the same conditions.

(4) Low uranium concentration samples that are both siltstone and sandstone from Location 1 of the Horton Group had very consistent results at both 5°C and 20°C.

(5) When leached water was removed from source rock and measured after three half lives, the level of radon was not reduced but remained generally the same.

(6) The Reeves Road granitic sample had comparable results (radon levels >1,000,000 Bq/m^3) to that of the Horton Group samples of high uranium concentration.

(7) Dissolved metals water analysis of the distilled water following leaching from all three rock types showed leached water contained some dissolved uranium.

(8) The river water had an initial radon reading of >1 000 000 Bq/m^3 yet water analysis showed no uranium was detectable in the water.

(9) When comparing distilled water suites to river water suites there were fluctuating levels of radon over the duration of 30-40 days.

(10) Numerous suites consistently dropped in radon levels between 20-30 days then steadily increased from 30-40 days regaining the peak levels.

(11) After more than eight half lives, radon levels are consistently high in most suites therefore radium is present.

(12) In all cases, peak radon levels reached values >1,000,000 Bq/m^3 more than 6 times the suggested safe level of radon in water, as defined by the US EPA.

5.0 Discussion

5.1 Field Observations

The main field observations that are important are:

(1) The potential for uranium and/or radium to leach into the surface water and groundwater from the intensely weathered source rock, and the potential for redistribution of the uranium and/or radium and their subsequent daughter products in the surface and groundwater is evident. The rock samples used in this study are variably weathered with the Reeves Road granite (saprolite) experiencing the most intense weathering. The samples ranged in uranium concentration from a high of 212 ppm in one of the siltstones to a low of 4 ppm in the granite. However, the leaching process showed that in all suites, elevated levels of radon were measured and all suites reached the same peaks of at least 1 000 000 Bq/m^3 (upper detectable limit of the RAD7). Because radon has a half-life of 3.8 days, if radon was not being replenished by decay of radium from the rocks, the water would not continue to have high levels of radon over the time period of the experiment. If the uranium and/or radium is being leached into the surrounding water, it can subsequently be redistributed when the water then moves (i.e. streams or groundwater). Further supporting evidence indicating that radon is indeed being replenished by either radium dissolved in the water or by radium in the particulate sediment, is identified by the initially high levels of radon measured in the river water: the initial radon test completed on the river water indicated

radon levels in excess of 1 000 000 Bq/m^3 yet the **dissolved** metals analysis showed that there was no detectable uranium. Therefore the source of the radon in the river water was upstream and the radon was being redistributed by the stream.

(2) Initial uranium concentrations of the rock are not necessarily good predictors of the levels of radon in water expected. Ball and Miles (1993) also found that the uranium concentration in the rock did not correlate with the radon levels measured in the resultant water, in some cases high uranium concentrations did not result in high radon levels. Whereas in this study, rocks with elevated uranium concentrations do show elevated radon in water concentrations, it is also possible to get elevated levels of radon in water from rocks with relatively low concentrations of uranium. As seen in the results, uranium-enriched siltstone with a uranium concentration of 212 ppm yields a peak radon level of >1,000,000Bq/m^3, but so also did the siltstone with a uranium concentration of 6 ppm yield a peak radon level of >1,000,000Bq/m^3 as did the granite with a uranium concentration of 4ppm. From a health perspective, this suggests that not only rocks with high uranium content may produce high levels of radon in water.

5.2 Radon in water

The main observations of radon levels in the water are as follows:

(1) Agitation of suites to simulate weathering processes results in greater fluctuations in radon levels measured compared to the suites that have had no agitation (Figure 4.3(a,g,l) and Table 4.3h). This more erratic pattern is attributed to the agitation of the samples. When the samples are agitated, the surface area increases, and radon that was trapped is free to escape. In addition, radium is leached into the water and the radon starts to decay but there is a delay in the detection of radon once the radium is released as the radium decays in sufficient quantities to produce elevated radon levels. The cycle is then repeated with the next agitation causing the rock sample to break down further. By the end of the 40 day cycle the average grain size of the rock sample had significantly decreased from 1 centimeter to a few millimeters which created more surface area in turn allowing more radon to be leached into the water.

(2) When leached water was decanted and removed from the source (the sediment) for three radon half lives, the level of radon was not reduced significantly but remained generally the same. The leached water had been extracted after 26 days and when the radon

levels were peaking (>900 000Bq/m3). This decanted water was stored without air space trapping any radon in the water for the three half lives. This water was tested after 4, 8, and 16 days of being removed from the source and each time, still resulted in high radon levels. One possibility is that the leached water or fine particulate matter within the water contains radium which is the immediate parent of radon. With radium in the water (or particulate matter), the water can be removed from the uranium source and still decay into radon. But with no source of radium the water should result in measurements of low levels of radon in the water within a few days.

(3) The Reeves Road granite sample with low uranium concentration had comparable radon in water results to those of the Horton Group samples of high uranium concentration (Table 4.3 and Figure 4.3). This observation suggests that due to the intensity of weathering that the granite (saprolite) has undergone, the uranium that once may have been high within the granite was weathered away leaving its decay daughter radium remaining as a source of the high radon levels. In other words, the uranium series isotopes at this site are in disequilibrium.

(4) The concentration of uranium and radon in the Horton Group is not specific to one lithology as seen in the high concentration of both radon and uranium in both sandstone and siltstone. Parsons (2007) identified a strong correlation between the uranium concentration and other trace elements such as fluoride, chlorine and sulphate which leads to the idea that the uranium within the rock units may be mobilized with these elements, consistent with that of roll front deposition (Skinner, 1999). Results from this study are inconclusive to these data.

(5) Water analysis of dissolved metals of the distilled water following leaching recorded the dissolution of uranium when exposed to each of the three rock samples, although only the river water leached from uranium-enriched siltstone was above the guideline level of 20μg/L, similar to results obtained by Parsons (2007). On leaching, the sediments in the distilled water produced water with some dissolved metals, including dissolved uranium (Table 4.4). The siltstone had an original uranium concentration of 212 ppm and the leached water yielded uranium levels of 22μg/L. The Reeves Road granite had an original uranium concentration of 4 ppm and the leached water yielded uranium levels of 0.4μg/L.

When comparing distilled water suites to the river water suites, the radon levels fluctuated more in the distilled water over time.

The difference in the results could be attributed to the metals and background levels of radon in the river water compared to the distilled water that was initially pure. The river water started out with an initial radon level of >1,000,000 Bq/m^3 therefore indicating a source of radon within the water amplifying the radon measurements. Whereas the dissolved metal contents of the final waters produced from the leaching experiments were analyzed, total metals were not analyzed, so it is possible that fine particulate matter in the water could have adsorbed metals.

(6) When changing the temperature of the distilled water suites from 20°C to 5°C the radon levels reached were comparable but the time to peak radon measurements was faster in the colder suites. Radon gas is more soluble in cold water than in hot water, at elevated temperatures the warmer water more readily releases radon therefore more radon is left in the colder water when measured. The river water suites had more consistent results between the temperature changes which could be due to the elevated background levels of radon initially measured in the river water.

(7) Numerous suites consistently dropped in radon levels between 20-30 days then steadily increased from 30-40 days regaining the peak levels the reason for this is unclear, however one possibility is that

there is a lag time between initial radon decay at the water-sediment interface and radon replenishment from radium within the sediment.

(8) Even after eight radon half lives, radon levels are consistently high in most suites. With such high radon measurements, radon leaching from rocks to water is a concern. Canadian Drinking Water Quality Guidelines (Health Canada, 2008) have no radon in water guideline for drinking water but the EPA is proposing new guidelines in the US that suggest levels of radon that exceed 4,000 pCi/L (148,000Bq/m3) in water are harmful to drink. In all suites the results from the leaching process yielded extreme radon levels that should not be consumed without further testing to determine the health risks that the radon poses since radon gas has been found to cause lung cancer when inhaled. Although Health Canada does not have guidelines for radon in water, radon in hot water outgases readily, and can contribute to increased levels of radon in indoor air, particularly in areas such as bathrooms in a home. As well, studies into the impact of radon in drinking water on other organs in the body, such as stomach cancer, are ongoing.

6.0 Conclusion

Based on the results that were collected from the radon in soil gas, leaching experiments of the rock samples and the uranium data gathered about the Horton Group and South Mountain Batholith I have concluded the following about radon, radium and uranium in the study area:

(1) Radon is currently a problem in the areas of focus of this thesis.

(2) Radon is being leached from the rock in sufficient quantities and over a time frame that suggests there is radium present in the sediments, and this radium is also involved in the leaching process.

(3) The concentration of uranium in the source rock is not the sole determining factor when identifying a radon problem in the water and soil gas. Even in areas with low uranium levels it is possible for radon to leach into the surrounding water systems and soil horizons. In other words, the uranium content is not the determining factor but rather its availability to dissolve.

(4) Radon is being leached into the surrounding area due to the weathering of the source rock and is potentially being redistributed through surface water and groundwater.

(5) The suites that were kept cold at 5°C had elevated levels of radon prior to the warmer suites (20°C) but all suites reach the same peak of >1,000,000 Bq/m^3 which was the upper detection limit of the RAD7 radon detector. Even with this limitation, levels of 1,000,000 Bq/m^3 or greater are considered problematic. This indicates that temperatures over the range studied are not a determining factor.

(6) After eight half lives of radon, the radon levels that are measured are consistently high suggesting that the amount of radon being leached from the weathered rock is a concern.

6.1 Recommendation for Future Work

This thesis has identified that radon migrates into waters and soils from selected outcrops of granite of the South Mountain Batholith and the Horton Group sandstones and siltstones as these rocks are weathered. Future work in the area should include:

(1) A better understanding of all the elements in the uranium-238 decay series with more studies on the potential disequilibrium of the series in the waters draining the rocks of the region.

(2) Previous studies were completed in the late 1970s and early 1980s on the well waters of the study area which should now be updated to determine any changes in the distribution of uranium, radium and radon levels in the well water of the areas.

(3) A study should be conducted on a larger scale surrounding the study area to determine the attributes of weathering and the scale and extent of the uranium occurrences. The impact of weathering on the distribution of uranium not recognized prior to 1999 (O'Beirne-Ryan, 1999) should be studied. Also, further work on regions of known saprolite development is warranted.

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APPENDICES

Appendix A1

Units of Measurement	
Bq/m^3	The International System of Units (SI) for activity of radioactive material. One Becquerel is equal to one disintegration per second
pCi/L	A unit of measurement used by the United States to describe the intensity of radioactivity in a sample of material. One curie is equal to thirty-seven billion disintegrations per second.

Conversion Factors
$37\ Bq/m^3 = 1\ pCi/L$
1 µg/l = 0.001 ppm