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Decommissioning of Low-grade Uranium Mines in Canada Current Status and Long-term Radium Mobility

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Abstract

Low ore grade and uranium market conditions have lead to the closure of all low-grade uranium mines in Canada. With the exception some past historic and abandoned mine sites, all other inactive and recently closed-out mines and associated waste management sites have been rehabilitated and decommissioned. Acid mine drainage and contaminant migration, including uranium decay series radionuclides and trace heavy metals, from waste management areas are major environmental concerns at many of these sites. Hence, long-term prevention, control and treatment measures are required.

In the past, many older and inactive surface-deposited waste management sites, having acidic drainage problems, were rehabilitated with vegetation covers. This has controlled surface erosion and greatly improved site aesthetics. However, acidic drainage continues and at these sites long-term effluent collection and treatment is required. The waste management areas at the newer, recently closed mines have been rehabilitated with shallow, in-situ water covers, having a minimum depth of approximately 1 m, to minimize acid generation and release of radon gas and its progeny. Acid generation at these sites has decreased significantly, and at most sites only a limited effluent treatment is required for pH control and radium removal.

The mobility of Ra-226 from pyritic uranium tailings was evaluated for both on-land and underwater disposal scenarios. During the acid generation phase and/or in the presence of available sulphate ions, the dissolution and mobility of radium was very low due to sulphate ion solubility control. With the cessation of acid generation, and/or upon depletion of sulphate ion control, including microbial sulphate degradation, the solubility of radium was enhanced. Hence, further investigations are needed to improve the understanding of the long-term stability and control aspects of decommissioned uranium mine waste management sites.

Introduction - Canadian Uranium Mining History

Canada is the world's leading producer of uranium, producing approximately 11,500 tonnes of uranium annually (Vance, 2002). The uranium mining in Canada started in the 1930's with the opening of the first radium/uranium mine by Eldorado Gold mining company at Port Radium in the Great Bear Lake region of Northwest Territories (Figure 1). Pitchblende was mined here first for the radium dial paint industry (1934-1939), and then for uranium (1943-1962) for the Canadian nuclear research program. Uranium exploration, prospecting and mining activities increased many fold in the 1950's to meet the increased uranium demand for the US nuclear and western defence programs. This resulted in the opening of many small and medium-sized mines in the Elliot Lake-Blind River and Bancroft regions of north-central and eastern Ontario, and in the Uranium City-Lake Athabasca region of northern Saskatchewan (Figure 1). During this period, a total of 23 mines and 19 uranium-milling plants were in operation. The first phase of Canadian uranium production peaked in 1959 when more than 12,000 tonnes of uranium was produced, yielding more export revenue than any other mineral exported from Canada that year.

Uranium ore bodies mined in the Elliot Lake-Blind River region were low grade, containing approximately 0.1% U. The ore, hosted in a quartz-pebble conglomerate, contained some sulphide mineralization, mostly that of iron and some trace heavy metals. In the Uranium City-Lake Athabasca region the mines were again low grade but the ore was primarily pitchblende veins hosted in different types of rock. Most of the uranium mines in both of these areas were closed and the facilities abandoned in the early 1960's due to the cancellation of the uranium delivery contracts by the US Department of Energy. The remaining mines in Elliot Lake and Bancroft regions of Ontario, and in the Uranium City region of northern Saskatchewan, were consolidated in the late 1960's to early 1970's and uranium production stabilized to meet the increasing demand of the nuclear power generation industry.

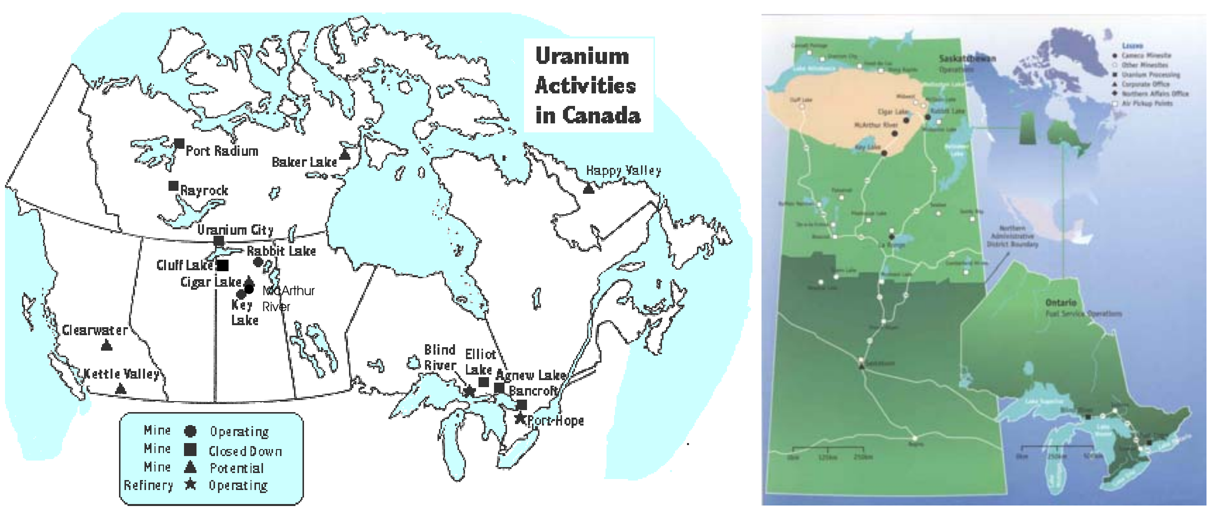

Figure 1. Locations of the past and present uranium mines in Canada.

Economic realities of low-grade ore and uranium market conditions led to the cancellation/non-renewal of several long-term uranium delivery contracts for the uranium mining companies in the Elliot Lake region. Consequently, all low-grade uranium mines in Ontario and northern Saskatchewan closed during the early 1980's to mid 1990's. Presently, uranium mining operations are concentrated in the Athabasca sandstone region of northern Saskatchewan, containing high-grade ore deposits averaging between 0.4 to 23% U (Figure 1).

Excepting some older inactive and abandoned mine sites in northern Saskatchewan and eastern Ontario, all other inactive and recently closed low-grade uranium mines have been rehabilitated and decommissioned or are in a process of being decommissioned. This paper reviews the status of decommissioning of uranium mine, mill and associated waste management sites in Canada, and outlines potential long-term environmental issues associated with uranium mine decommissioning.

Uranium Mine/Mill Rehabilitation and Decommissioning

The Government of Canada is responsible for ensuring that the long-term storage and management (including disposal) of radioactive wastes from uranium mining, milling, and refining and reprocessing facilities, and nuclear power generation plants are carried out in a safe, environmentally sound, comprehensive, cost effective and integrated manner.

The *Nuclear Safety and Control Act* (NSCA) requires that all uranium producing, processing and waste storage facilities, including active, inactive and abandoned sites be licensed by the Canadian Nuclear Safety Commission (CNSC). The licensing of inactive and abandoned (past historic) sites is required to enable the CNSC to ensure appropriate financial guarantees, where deemed necessary, covering the decommissioning and subsequent management of the waste sites (CNSC 2001).

Site Classification

The low-grade uranium mine, mill, processing and waste management sites that have been inactive or recently closed are classified according to the following two categories:

1. Past Historic sites:
    a. Inactive and abandoned sites
    b. Rehabilitated, decommissioned, and managed inactive sites
2. Newer mine sites

The past historic sites are those that operated and closed during the early period of uranium mining, from the 1930's to 1960's, and have been inactive for the past thirty years or so. Many of these mines operated during the period when environmental regulations were non-existent, were operated by small or junior mining companies and abandoned upon cessation of mining activities. For those sites whose owners/operators are not easily traceable, the management responsibility has been reverted to the government. Many of these sites are in an immediate need of proper rehabilitation and decommissioning. These sites remained unlicensed.

The past historic and inactive sites that are owned and operated by their original or new private-sector owners have all been properly rehabilitated and decommissioned. These sites are on care and maintenance, and are managed by their owners/operators according to the past-historic site licensing requirements.

The newer sites are those that ceased operation in the early to mid 1990's. These sites have been properly rehabilitated and decommissioned in accordance with the new CNSC licensing requirements. Presently these sites are in a transition phase of monitoring, and care and maintenance.

Inactive and Abandoned Historic Waste Sites

These sites include Gunnar and Lorado mine/mill and several other smaller mine sites in the vicinity of Uranium City - Lake Athabasca region of northern Saskatchewan, Port Radium and Ray Rock mine/mill sites in the Great Bear Lake region of Northwest Territories, and Dyno, Bicroft and Faraday mine sites in the Bancroft region of eastern Ontario (Figure 1). These sites have been abandoned for the past 40 years or so, and are in the need of proper rehabilitation and decommissioning. In order to comply with the new CNSC licensing requirements, joint plans and funding arrangements are being developed by the respective government departments for the long-term management of these sites. A brief description and summary of any rehabilitation/decommissioning work performed at some of these sites are provided below:

Gunnar Mine/Mill Site
The Gunnar mine/mill site is located on the north shore of Lake Athabasca, approximately 26 km southwest of Uranium City (Figure 1). The mine operated in the mid to late 1950's and has been abandoned since. Figure 2 shows a general view of the abandoned Gunnar mine/mill complex, main tailings basin, waste rock pile and open pit. The site has approximately 4.5 million tonnes of acid-leached tailings that were discharged un-neutralized, at a pH of around 1.7-2.0, in a nearby Mudford Lake, which filled the lake.

Figure 2. General view of Gunnar mine/mill complex, tailings, open pit and waste rock pile.

A major proportion of the Gunnar tailings are contained in the Gunnar Main area covering approximately 45 ha (Figure 2). Some tailings, mostly fines, overflowed the Mudford Lake basin and settled in Langley Bay, and in a small area in between Gunnar Main and Langley Bay (Gunnar Central). The tailings in Langley Bay divide the bay into a closed out western arm, and the remaining part is open to Lake Athabasca. Some tailings in the bay are underwater (Kalin 1982).

The tailings have little pyrite and are non-acid generating. The surface of the tailings in the Gunnar Main and Central areas is moderately acidic to nearly neutral, having pH's of 4.0-7.9, and is mostly dry with some vegetation cover dominantly in the Gunnar Central area. The site also has a large waste rock pile, a flooded open pit that connects to underground workings and some old buildings and structures (Figure 2). Surface discharge from the site flows untreated through various channels to Lake Athabasca.

<u>Lorado Mine/Mill Site</u>

The Lorado mine/mill site is located near Hanson Bay, on the west side of Beaverlodge Lake, approximately 10 km southwest of Uranium City (Figure 1). The mill at the site operated from 1957-1960, custom milling sulphide rich ores from several mines (6 major and 25 smaller mines), and produced approximately 0.4 million tonnes of acid leached tailings. The tailings are acid generating, containing up to 10% pyrite, and were deposited un-neutralized in a nearby Nero Lake, turning the lake acidic. Initially, pyrite was floated in the milling process to produce the required sulphuric acid for leaching. Tailings produced during this period were deposited on the surface, forming two beaches at the west end of Nero Lake, as shown in Figure 3. The exposed tailings produce acidic drainage that drains towards Nero Lake (Kalin 1982).

Figure 3. General view of the Lorado tailings and Nero Lake.

The Nero Lake water is acidic having a pH of approximately 3-4 and a total Ra-226 concentration of 1 Bq/L. Untreated drainage from Nero lake flows partly by surface overflow and partly by subsurface seepage through a berm to Hanson Bay, an embayment of Beaverlodge Lake, where a plume of metal precipitates could be clearly seen.

For both of these sites, rehabilitation, decommissioning and long-term management plans and funding arrangements are currently being developed jointly by the respective government departments.

Rehabilitated and Managed Historic Waste Sites

These uranium mine/mill and waste management facilities, which operated during the 1950's to 1960's and some up to the early 1980's, have been all rehabilitated and are managed by their current owners/operators. Nine of such sites are located in the Elliot Lake area, which include: Nordic, Lacnor, Pronto, Milliken, Spanish American, Sheriff Creek, Canmet and Stanrock. In addition, two small wetland sites containing partially submerged tailings at Panel Wetlands and Lower Williams Lake have been included as parts of extended waste management facilities of nearby Panel and Denison waste management sites. All uranium mine tailings in the Elliot Lake area are acid generating, containing approximately 5-10% pyrite. Other rehabilitated and managed sites include: Agnew Lake mine, located approximately 40 km northwest of Sudbury, Ontario, Madawaska mine near Bancroft, Ontario, and Beaverlodge mine near Uranium City, northern Saskatchewan.

Excepting Agnew Lake and Madawaska mines, all other mine and waste management facilities are licensed by the CNSC as historic waste sites, and are on care and maintenance basis and managed accordingly. The Agnew Lake site was rehabilitated and decommissioned during the early 1980's. After ten years of site monitoring and management, it was granted closure in the early 1990's and has now been reverted back to the Provincial Crown. The Madawaska mine site was also rehabilitated, to a certain extent, during the 1980's. Both these sites are presently unlicensed.

The detailed site description and summary of rehabilitation and decommission activities conducted at two of the inactive sites in Ontario, namely Nordic and Stanrock mines at Elliot Lake, and one in northern Saskatchewan, Beaverlodge mine, at Uranium City are provided below. These sites are illustrated for their diversity in past management history and/or ore types.

Nordic Mine/Mill and Waste Management Sites

The Nordic mine, located approximately 5 km east of the city of Elliot Lake, operated from 1957 to 1968 and produced approximately 12 million tonnes of acid generating tailings containing ~5-10% pyrite. An acid leach process was used in milling and the tailings were neutralized to a pH of ~ 8.5 prior to their discharge in an above grade waste management facility. Most of the tailings are contained in the larger Nordic Main area with a smaller proportion deposited initially in the Nordic West Arm extension of the main tailings area. The surface deposited tailings area covers approximately 107 ha and is impounded by perimeter dams along the southeast, south and west sides. The tailings site was extensively used in the past for environmental research in the areas of vegetative reclamation, acid generation, and contaminants migration and uptake via various environmental pathways. The tailings surface was successfully revegetated and rehabilitated in the mid 1970's, and the site has been one of the best revegetated sites in Canada for the past 25 years or more, as shown in Figure 4.

A full rehabilitation of the site was done in the mid 1990's. The mine/mill complex and ancillary facilities at the site were removed. The shaft and all opening to underground workings were sealed and capped with cement pads. All contaminated materials at the mine/mill site were relocated to the waste management area, excavated areas backfilled and covered with a layer of clean sand or till and the site was vegetated. Dams and effluent treatment facilities at the site were upgraded in 1998-1999.

Figure 4. Rehabilitated Nordic mine and waste management facilities, Elliot Lake, Ontario.

The site is presently managed on a care and maintenance basis (BHP Billiton 2001). Surface run-off and seepage from this site and from the upstream located Lacnor waste management facility are collected and treated at the Nordic effluent plant. The treatment sludge is allowed to settle and is stored in the nearby North Nordic Lake sludge management facility.

Stanrock Mine/Mill and Waste Management Sites - Vegetative Reclamation

The Stanrock mine, located approximately 20 km northeast of the city of Elliot Lake, operated from 1957 to 1968 and produced approximately 8 million tonnes of acid generating tailings containing ~ 5-10% pyrite. The milling also used an acid leach process and the tailings were neutralized prior to their disposal in a surface based waste management facility. The site covers an area of approximately 71 ha and also contains tailings discharged from a nearby Canmet mine. Originally, the tailings were impounded by extensive dams, constructed of coarse cycloned tailings, on the east, south and west sides. This resulted in breach of dams and spillage of some tailings outside the containment area in the past. Excepting some experimental vegetation trials and other surface stabilization and rehabilitation studies, the surface of the tailings remained exposed and un-reclaimed. The site was kept pending for disposal of additional tailings from future mining operations. Figure 5 shows the Stanrock tailings site before and after decommissioning.

The mine/mill complex at the site was removed and the area rehabilitated during the early 1990’s. The waste management site was rehabilitated during 1997 to 1999 by constructing new, till-cored engineered dams immediately downstream of the original containment dams. These low permeability dams were designed and constructed according to new and applicable dam construction codes, and keyed onto the grouted basement rock (bedrock). This was done to facilitate the rise of the water table to near the tailings surface and above the un-oxidized tailings zone in the basin, thus minimizing further oxidation of the tailings, and at the same time providing radon exhalation control. A new effluent treatment facility was constructed at the site in 1998 and the tailings surface was vegetated during 1998-2000 (Ludgate et al. 2000). Water cover at the site was considered during the planning stage of decommissioning, but it was deemed to be unfeasible and too expensive given the topographical conditions at the site.

Figure 5. Stanrock waste management facility before and after rehabilitation, Elliot Lake, Ontario.

The site is currently managed on a care and maintenance basis. The effluent from the site is collected and stored in a holding facility for treatment on a seasonal basis, or on demand as and when required, depending upon effluent discharge flow and loading conditions.

<u>Beaverlodge Mine/Mill and Waste Management Sites</u>

The Beaverlodge mine is located approximately 5 km east of Uranium City. It operated from 1952 to 1981 and produced approximately 10 million tonnes of tailings and 4.8 million tonnes of waste rock from one major and 11 satellite mines. During the course of mine operation a total of 12 open pits were also established at the site. The ore had low acid generation potential and an alkaline, carbonate leach process was used in the milling. Approximately 40% of the total tailings were placed underground as backfill, and the remaining were placed in two natural lakes, Fookes Lake and Marie Lake, located on the upstream side of Beaverlodge Lake towards the north. A majority of the tailings were deposited in Fookes Lake as a surface deposition facility and tailings beach developed on both lakes (Conor Pacific 1999, Phillips et al. 2000, and Phillips 2002). Figure 6 shows the Beaverlodge mine waste management facility before and after decommissioning.

The site was decommissioned from 1982 to 1985 during which all mine/mill and ancillary facility buildings were demolished, contaminated materials placed underground and all openings to underground workings were sealed with waste rock or capped with concrete. The open pits were backfilled with waste rock and/or flooded, and the remaining waste rock surface was recontoured to conform to the local topography. The exposed tailings in the two lakes were covered with a 0.6-m layer of waste rock and the manmade water controlling structures were removed to bring the water levels in the two lakes to within 1 m of their natural outlet levels (Figure 6). Additional reclamation work consisted of covering any exposed tailings areas, which developed on top of the waste rock cover after initial reclamation, with a 0.3-m layer of fine-grained filter sand overlain by an additional 0.3 m layer of sand and gravel (Conor Pacific 1999).

Figure 6. Beaverlodge mine waste management facility before and after rehabilitation, Uranium City, northern Saskatchewan.

The site is managed on a care and maintenance basis and no active treatment is provided to drainage effluents. Planning is underway for returning portions of the decommissioned site back to the province for institutional control.

Decommissioning of Newer Mine Sites

Mines that ceased operation during the early to mid 1990's, though some of them have been operating since the mid 1950's, are closed out and decommissioned according to the new CNSC licensing requirements. Four of such mines, namely Quirke, Panel, Denison and Stanleigh mines, are located at Elliot Lake, Ontario. While Quirke, Panel and Denison mines ceased operation during 1990 to 1992, the Stanleigh mine was closed in 1996. With the closing of its last operating mine, the uranium mining era in the Elliot Lake region, known popularly as the uranium capital of the world, came to an end. These mine sites have extensive waste management areas comprised of acid generating tailings containing ~ 5-10% pyrite. Acid mine drainage (AMD) is a major issue of environmental concern at these sites.

In the past, the surface deposited tailings and waste rock sites were rehabilitated with a vegetation cover (e.g. Nordic tailings site), but the expected benefits of the cover in controlling acid generation and release of contaminants were not realized. Thus, for controlling acid generation at the newer sites, the waste management areas have been rehabilitated to provide a shallow, in-situ water cover having a minimum depth of approximately 1 m on top of the submerged tailings. Water has a low oxygen solubility of 8-10 mg/L compared to ~ 285 mg/L in air (20% oxygen concentration), and a low diffusion coefficient for molecular oxygen of ~ $1 x 10^{-9}$ m^2/s compared to ~ $1.8 x 10^{-5}$ m^2/s in air. Thus, where site climatic and topographic conditions are favourable, a water cover provides the most economical and natural cover for controlling acid generation in-situ. The details of site rehabilitation and decommissioning of two such mine sites, namely Quirke and Denison, are provided below.

Quirke Mine/Mill and Waste Management Sites

The Quirke mine located approximately 14 km north of the city of Elliot Lake, Ontario, operated from 1956 to 1961, and again from 1968 to 1990. It produced approximately 42 million tonnes of acid generating tailings and 4 million tonnes of waste rock. Upon cessation of mining, the rehabilitation and decommissioning activities at the mine site, including establishment of a shallow water cover on the tailings, started in 1992. All the buildings and ancillary facilities at the mine site were removed, and all mine openings and shafts were sealed and capped with cement pads. All contaminated materials, soils and acid generating waste rock were hauled and deposited in the waste management area. The mine site was rehabilitated by covering with a clean till/sand fill and revegetated.

The tailings and waste rock were placed in a 192-ha waste management area (WMA), located in a valley containing east-west trending ridges, and impounded by low permeability containment dams. The acid generating waste rock was used to construct internal haulage roads and dykes within the waste management facility. The waste management area has an elevation difference of approximately 15 m along the east-west direction, which made it difficult to establish a single elevation water cover. The site was thus divided into five cells by internal dykes in a terraced configuration to provide the required water cover at five different elevations, as shown in Figure 7. The internal dykes have fine tailings and till layers on upstream sides to minimize seepage losses through them. The perimeter containment dams are engineered, clay-core-earthfilled dams keyed onto the grouted bedrock surface at the bottom. At the main eastside dam, where the bedrock was too deep, a vertical bentonite grout curtain as a cut-off wall was also constructed below the dam between the clay core and basement rock, to minimize seepage losses (Senes 1993, Payne 2000 and BHP Billiton 2001).

Figure 7. Rehabilitated Quirke mine/mill and waste management areas, Elliot Lake, Ontario.

The water cover in the WMA is maintained by natural precipitation and runoff from the catchment area of the site, and by diversion of surface discharge flow from an upstream located Gravel Pit Lake. The water level in the Gravel Pit Lake is maintained approximately 3 m above the maximum water elevation in the WMA. This facilitates a gravity inflow from Gravel Pit Lake to the WMA, at the same time maintaining a higher hydraulic pressure head in the lake to minimize seepage of contaminated porewater from the WMA to the lake.

Prior to the establishment of the water cover, the surface of the tailings in the WMA was leveled and agriculture grade limestone was incorporated into the exposed tailings surface to neutralize existing and any potential acidity generated during and after establishment of the water cover. Hydrated lime slurry is periodically added to the various cells, as and when required, to maintain nearly neutral pH conditions in the surface water cover.

The water quality in upstream cells of the WMA is close to discharge quality levels, but flushing of previously accumulated acidity and oxidation reaction products is occurring from upstream to the downstream located cells. The effluent from the last cell is treated for controlling pH, dissolved metals and radium prior to its discharge. Presently the site is in a transition phase of treatment, monitoring, and care and maintenance.

Denison Mine/Mill and Waste Management Sites

The Denison mine, located approximately 16 km north of the city of Elliot Lake, operated from 1957 to 1992, and produced approximately 63 million tonnes of acid generating tailings containing ~ 5-10% pyrite. The tailings were placed in two tailings areas, TMA-1 and TMA-2, located in several former lake basins in a valley. TMA-1 lies immediately south of TMA-2 and is surrounded by east-west trending ridges and five engineered, low permeability dams. TMA-2 is located in a northwest trending valley and its outflow is directed towards TMA-1. The total surface area of the WMA is approximately 271 ha. Figure 8 shows the Denison mine, mill and WMA sites before and after rehabilitation.

The rehabilitation and decommissioning of the mine and WMA started in 1992 and completed in 1996. This consisted of removal and decontamination of sellable assets, removal of buildings and ancillary facilities including headframes, concentrators, workshops and building foundations etc. All contaminated materials at the site were removed and placed underground or buried at designated landfill locations within the WMA. All openings to the underground workings including mine shafts, adits and other mine portals were sealed and capped with concrete. The area around the mill complex was re-graded, covered with a layer of clean till, soil and overburden materials, when and where required, and the site was vegetated with local and agronomical species of grasses and legumes (Ludgate et al. 2000).

Figure 8. Denison mine/mill and waste management sites before and after rehabilitation and decommissioning, Elliot Lake, Ontario.

For providing single elevation water covers in TMA-1 and TMA-2, the tailings in TMA-1 were dredged from a higher elevation on the east side to below the expected final water elevation in the basin, and deposited in the deeper western part of the basin. The downstream perimeter dam in TMA-1 was reinforced and strengthened. The height of the low permeability west dam in TMA-2 was raised to divert the surface water flow from TMA-2 to TMA-1. The tailings above the anticipated final water elevation in TMA-2 were excavated or removed by hydraulic monitoring and placed in TMA-1 and underground. While removing the exposed tailings, lime was incorporated into the surface of the relocated and disturbed tailings in TMA-1 and TMA-2 to neutralize resident and potential future acidity. Figure 8 shows the Denison mine and WMA sites before and after rehabilitation.

The water covers in the two TMA's are maintained at near neutral conditions by periodically adding hydrated lime slurry or sodium hydroxide solution. The final effluent from TMA-1 is treated prior to its discharge on as and when required basis. The quality of the surface water cover in TMA-1 has improved significantly since the completion of the remediation work and only a very periodic treatment, especially during spring runoff, is required for pH control (Figure 9). However, treatment for controlling Ra-226 levels in the discharge effluent is still required. Similar to other decommissioned facilities, this site is presently under a transition phase of treatment, monitoring, and care and maintenance.

A progressive waste management and decommissioning strategy has been adopted for the new high-grade uranium mines in northern Saskatchewan. It consists of depositing the tailings and problematic waste rock in mined-out open pits having a porous envelope to isolate the deposited waste from local groundwater regimes. Upon closure, a layer of clean waste rock and/or till and sand, 1-3 m in thickness, would be placed on top of the deposited waste and submerged under approximately 10-30 m of water cover (Mittal et al., 2000).

Performance of Decommissioned Sites

All WMAs, including the previously decommissioned vegetated and the newly rehabilitated water cover sites are managed in accordance with the facility decommissioning and site management licences. Effluents from all WMAs are collected and treated, on as-and-when required basis, using hydrated lime or any other alkali solution for pH and dissolved metals control, and by addition of a solution of barium chloride ($BaCl_2$) for dissolved radium removal to meet the regulatory discharge water quality standards.

The water-covered sites are managed additionally by periodic additions of lime slurry to the established water covers to maintain near neutral pH conditions. This practice is followed to provide an in-situ treatment within the water cover for decreasing the treatment and sludge handling loads at the effluent treatment plant.

The performance of the rehabilitated sites was assessed by collecting and analyzing the annual total alkali consumption data, in terms of equivalent limestone requirements per unit area for the integrated basin including the effluent treatment plant, and any additional treatment provided to the vegetated or water cover basins. The performance of the Denison WMA having single elevation water covers, in terms of its annual equivalent limestone requirements/ha, as well as a comparative performance of the vegetated Nordic and water cover Quirke, Panel, Denison and Stanleigh sites are shown in Figure 9.

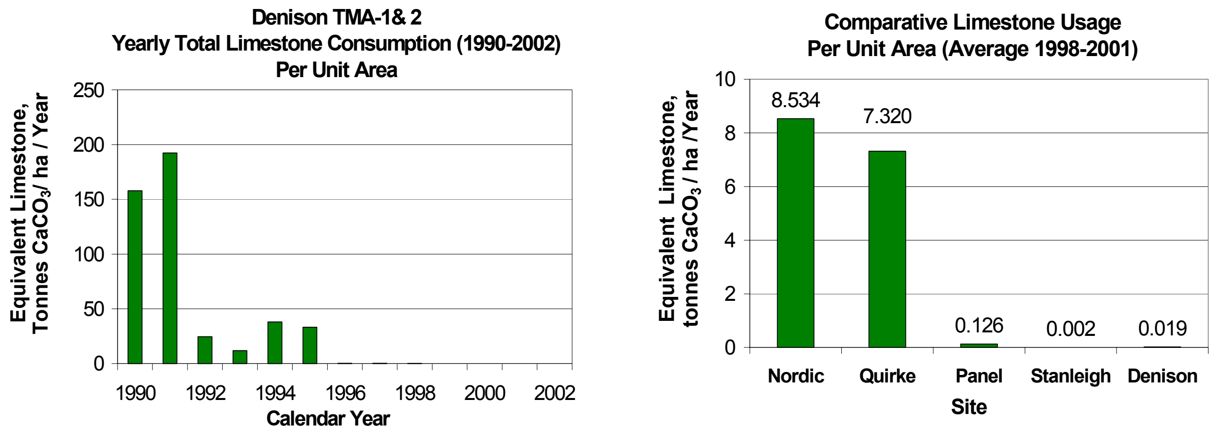

Figure 9. Performance of water covered and vegetated uranium waste management sites in the Elliot Lake area - area normalized, annual-equivalent limestone consumption rate for the various sites

The results clearly demonstrate the effectiveness of water covers in controlling acid generation. The post decommissioning annual equivalent-limestone-consumption rate/ha at the Denison WMA has decreased to less than 0.03% and 0.15%, respectively, of that required during operational and rehabilitation periods.

The yearly average equivalent-limestone-consumption rate (1998-2001) for the vegetated Nordic WMA was high at 8.53 tonnes $CaCO_3$/ha/y. The acid generation rate at the Denison, Panel and Stanleigh WMAs has decreased to less than 1.6% of that at the Nordic WMA during the same monitoring period.

The post rehabilitation equivalent-limestone-consumption rate for the Quirke WMA was initially high at approximately 7.3 tonnes $CaCO_3$/ha/y during the 1998-2001 monitoring period. It has decreased to ~1.5 tonnes $CaCO_3$/ha/y during 2002-2003 (Coggan, 2005). Because of its unique hydrological configuration and past exposure history, the Quirke WMA is still flushing out the stored acidity and oxidation reaction products and thus its total alkali requirements are high. With the continued flushing of these products the performance of the Quirke WMA is gradually expected to approach that of Denison, Panel and Stanleigh sites.

Uranium Mine Decommissioning – Long-term Radium Mobility

The decommissioning of uranium mine and waste management sites has been very successful, and all primary objectives related to physical and chemical stability of the decommissioned sites have been fully accomplished. Although acid generation in sulphide bearing uranium tailings and waste rock continues to be a cause of environmental concern at most vegetated sites, it has been significantly reduced with the incorporation of water covers at recently decommissioned sites.

The mobility of long-lived, uranium-decay-chain radionuclides, and particularly that of radium and its progeny, including their parent thorium isotopes, however, would remain the long-term and ultimate cause of environmental concern with uranium mine decommissioning. In this respect, the acid mine drainage related aspects are considered to be of a short term duration compared to the long-term containment requirements of the naturally occurring radionuclides within the uranium waste management facilities. The leaching and mobility aspects of Ra-226, having a half-life of 1600 y, and as one of the key monitoring parameters associated with uranium mining, both with and without acid generation, are considered below.

In the uranium milling process, according to Constable and Snodgrass (1987), most of the radium associated with the uranium ore matrix is first completely dissolved in the hot sulphuric acid leaching process, and then immediately co-precipitated with and/or adsorbed onto the various mineral phases associated with the precipitation of barite ($BaSO_4$), gypsum ($CaSO_4$), oxides/oxy-hydroxides and oxy-sulphates of Fe and Mn, as well as secondary alumino-silicate minerals in the leaching and partial neutralization circuits, and with other metal hydroxides and gypsum in the final neutralization circuit. More than 95% of Ra-226 contained in the uranium ore is immobilized with the tailings solids, leaving a very small fraction in the process solution streams. Because of the fine precipitates, the fine tailings fraction (<75 µm) generally contained higher total radium concentration, as much as twice or more, than the coarse tailings fraction (>75 µm) (Constable and Snodgrass, 1987; Goulden, 1998: and Davé, 1999a and 199b).

Following tailings neutralization and disposal, the residual radium in the tailings porewater is redistributed and decreased gradually as a solid-liquid partition equilibrium is established between the two phases at the prevailing pH – Eh (redox) conditions. The sulphate ion concentration in the tailings pore and contact waters, however, controlled the solubility of the sparingly soluble sulphate co-precipitates containing radium, and hence its leaching and mobility. The dissolved radium may be further redistributed amongst the Fe-Mn oxide/oxy-hydroxide/oxy-sulphate phases and other adsorption sites in accordance with the shifting partition equilibrium (Somot et al., 2002).

In the acid generating uranium mine tailings, the immobilized radium is fairly stable during the acid generation phase, and/or during gypsum dissolution, when the sulphate ions so produced controlled the mobility of radium associated with $BaSO_4$ co-precipitate. However, with continued oxidation and eventual depletion of pyrite as well as gypsum, the mobility of radium is enhanced through the increased dissolution of $BaSO_4$ and other secondary sulphate minerals, such as $PbSO_4$ and jarosite ($KFe_3(SO_4)_2(OH)_6(6H_2O)$), in the absence of the sulphate ion solubility control. Constable and Snodgrass (1987) observed in laboratory column leaching experiments that under unsaturated leaching conditions up to 80% of total Ra-226 contained in the top 10 cm layer of the experimental tailings was mobilized upon depletion of pyrite. Ra-226 was, however, re-precipitated in the tailings substrate below the zone of pyrite depletion. The authors further suggested that $BaSO_4$ was the main radium mobility-controlling mineral, whose solubility was adversely impacted by the depletion of pyrite and/or other soluble sulphate minerals such as gypsum. While the sulphate control on radium mobility was observed for both pyritic and non-pyritic uranium tailings, it was more pronounced in the former case.

Goulden (1998) undertook extensive mineralogical characterization and sequential extraction studies to quantify the association of Ra-226 with the various water soluble, weak acid soluble, cation exchangeable, reducible Fe-Mn oxides/hydroxides and other residual mineral phases in low-sulphide uranium mill tailings from a high grade uranium mine in northern Saskatchewan. The author concluded that while a very low fraction, ~0.35%, of the total Ra-226 contained in the process tailings was associated with the water-soluble mineral components, significantly higher fractions of total Ra-226 at ~54%, ~20% and ~ 15% were associated with ion exchangeable, weak acid soluble and reducible phases of Fe and/or Mn oxides/oxy-hydroxides, respectively. In these cases, both Ra-226 and Ba were extracted concomitantly. A residual 20% of total Ra-226 was associated with the insoluble fraction extractable only in a strongly oxidizing and acidic media. Thus, the mobility of radium may be enhanced in the absence of sulphate ion control due to continued weathering of decommissioned waste management sites, as well as in reducing environments where sulphate reduction may adversely impact the stability of $BaSO_4$ itself.

Laboratory column leaching studies conducted by this author over an extensive seven and a half year study period, simulating both on-land disposal (unsaturated conditions) and shallow water submersion (saturated condition) of pyritic uranium tailings containing ~ 5-7% pyrite, further elucidate the enhanced mobility aspects of Ra-226 in the presence/absence of sulphate ion solubility control as briefly described below. The detailed findings are presented separately in Davé (1999a and 1999b) and Davé et al. (2002).

<u>On-land disposal scenario – unsaturated zone leaching conditions:</u>
The unsaturated zone leaching experiments were conducted for two un-oxidized and un-amended uranium tailings: 1) coarse tailings (~ 96% greater than 75 µm) and, 2) un-segregated mill total tailings (~ 50% less than 75 µm). During the acid generation period, the Ra-226 mobility from both of these tailings was extremely low, at approximately <0.20% of total Ra-226 contained in the two tailings, as shown in Figure 10 and 11, respectively, for the coarse and mill total tailings. However, with continued leaching of the coarse tailings, and upon nearly depletion of pyrite in the top 80% of the tailings height, increased mobility and release of Ra-226 was observed near the end of the study period, when dissolved sulphate ion concentrations in the leachates had decreased to ~ 100 mg/L (Figure 10). In the mill total tailings, where acid generation continued at a very low rate compared to that of the coarse tailings, no such enhanced release of Ra-226 was noted near the end of the monitoring period (Figure 11).

Enhanced release of Ra-226 was also noted, at concentrations as high as ~ 10 Bq/L, during the additional flushing of the above leached coarse tailings under saturated conditions after completion of the leaching study, when the effluent sulphate concentrations were very low at near background levels of < 10 mg/L, as shown in Figure 12. The flushing was undertaken by submerging the leached tailings under a shallow water cover using natural lake water to remove all residual oxidation reaction products, which had accumulated during the unsaturated zone leaching of the coarse tailings, at the same time minimizing further oxidation of the remaining un-oxidized tailings. During this flushing process, an additional 1% of the total Ra-226 contained in the coarse tailings was removed by approximately 35 pore volume exchanges over a two and a half month's period. The total Ra-226 removal in this process was about five times that of the seven and a half year unsaturated zone leaching study. With further leaching of the coarse tailings under unsaturated conditions, after flushing and removal of the oxidation reaction products, the mobility of Ra-226 decreased again due to acid generation and increased sulphate ion concentration.

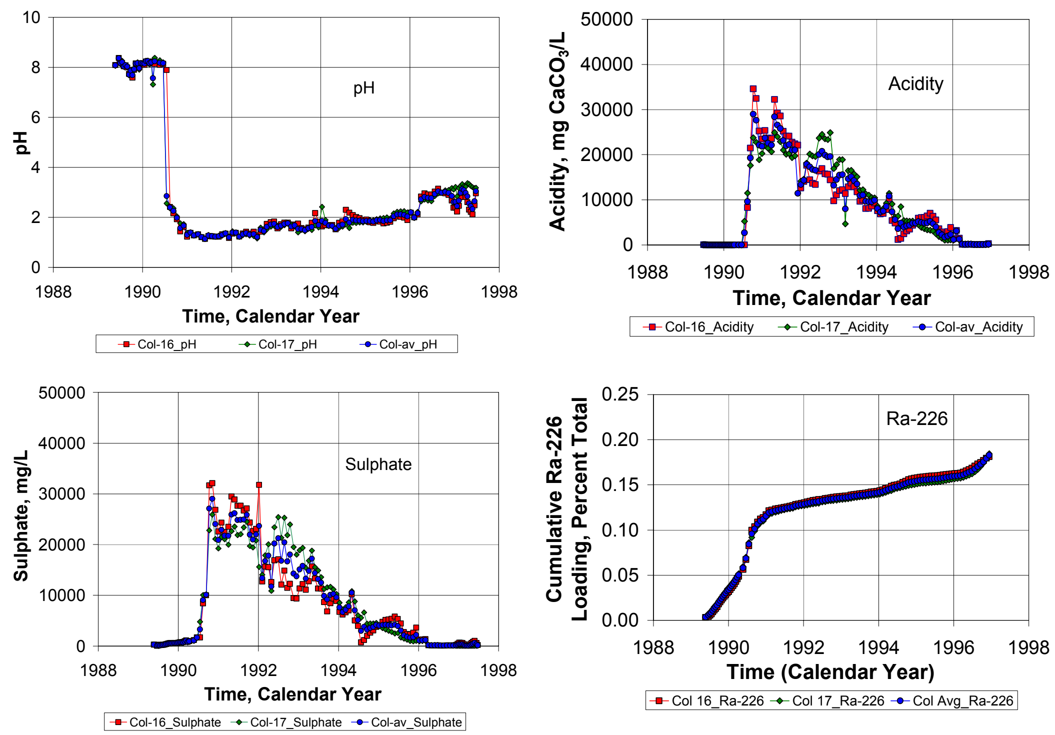

Figure 10. Unsaturated zone leaching conditions – variations of effluent pH, total acidity, SO_4 and cumulative loading of Ra-226 vs. time for coarse pyritic uranium tailings.

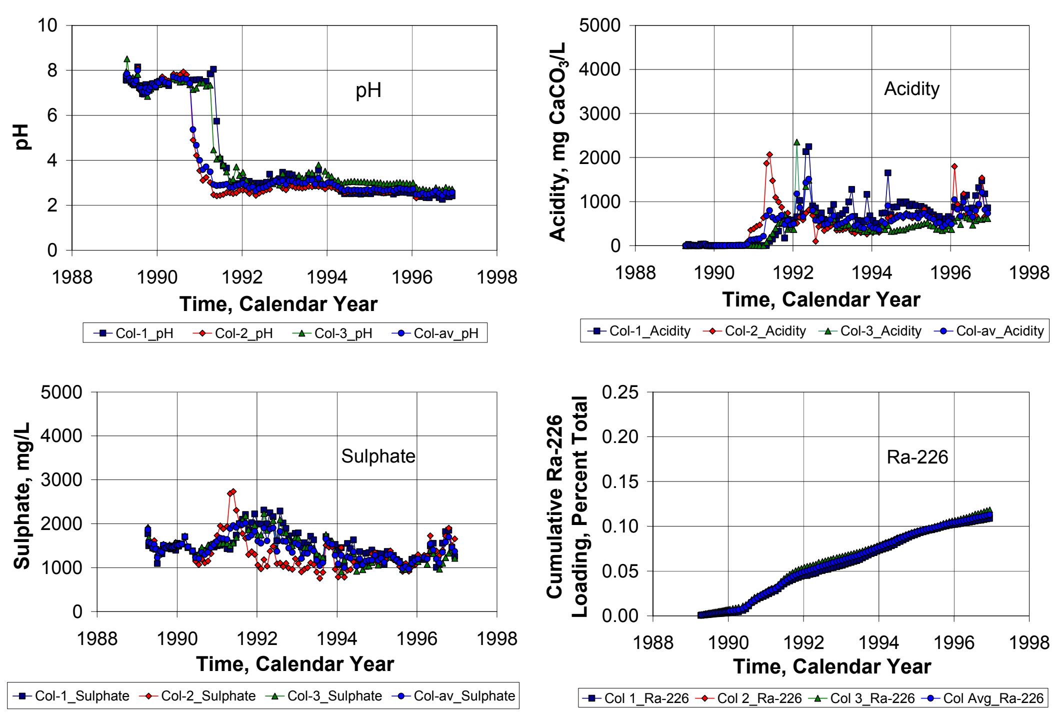

Figure 11. Unsaturated zone leaching conditions – variations of effluent pH, total acidity, SO_4 and cumulative loading of Ra-226 vs. time for mill total uranium tailings.

In both of these unsaturated zone leaching cases, the total amount of Ra-226 mobilized during the acid generation phase was significantly low and controlled by sulphate ion production/release.

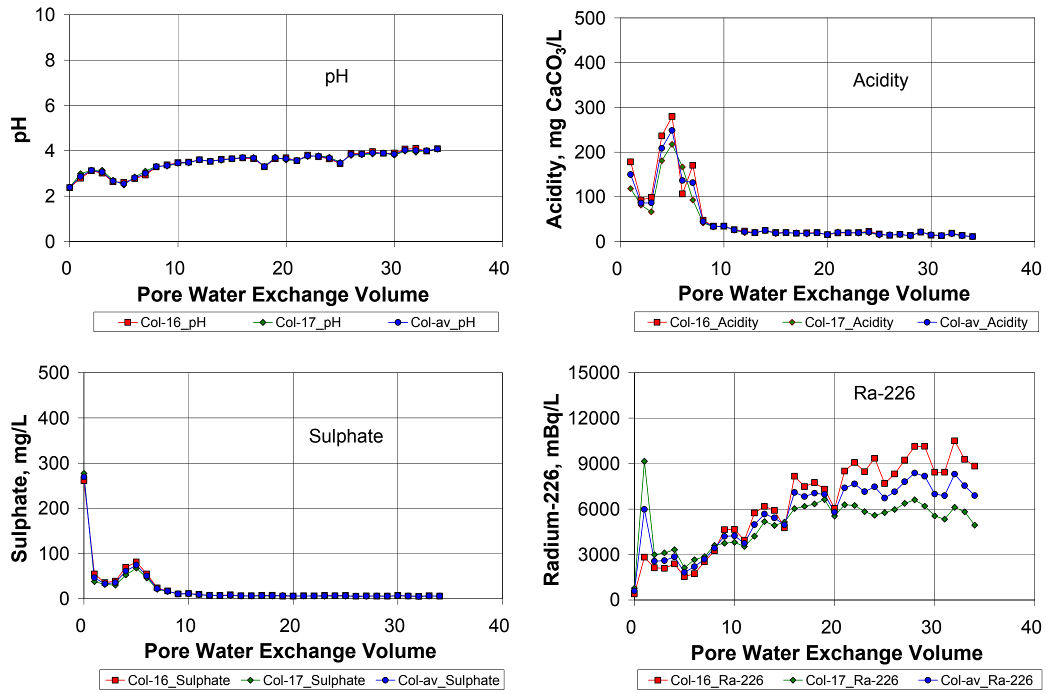

Figure 12. Post leaching, saturated zone flushing of coarse pyritic uranium tailings – variations of effluent pH, total acidity, SO_4 and cumulative loading of Ra-226 vs. time.

<u>Underwater disposal scenario – saturated zone leaching conditions:</u>
The saturated zone leaching experiments were conducted using un-oxidized, coarse pyritic uranium tailings, submerged under shallow water cover conditions to minimize their surface oxidation to a low rate controlled by the availability of dissolved oxygen in the water cover. Separate experiments were undertaken for tailings containing gypsum, and for tailings depleted of gypsum for surface water quality monitoring for a period of approximately fifteen months, and for tailings, both before and after gypsum removal, for long-term porewater quality monitoring for a period of seven and a half years. The first two experiments were conducted using aquarium type lysimeters and natural lake water, and the later one using column lysimeters and de-ionized water to facilitate the long-term porewater drainage requirements.

For the submerged coarse pyritic uranium tailings under shallow water conditions, significant mobilization and release of Ra-226 was observed both in the surface water cover as well as in the porewater drainage after complete depletion of gypsum, as shown in Figures 13 and 14 for surface and pore water quality profiles, respectively. The dissolved Ra-226 concentrations in the surface water cover of the gypsum-depleted tailings increased significantly to 33 Bq/L during the initial period of low dissolved sulphate concentrations, and decreased gradually to ~ 6 Bq/L with increasing sulphate concentration in the water cover (Figure 13).

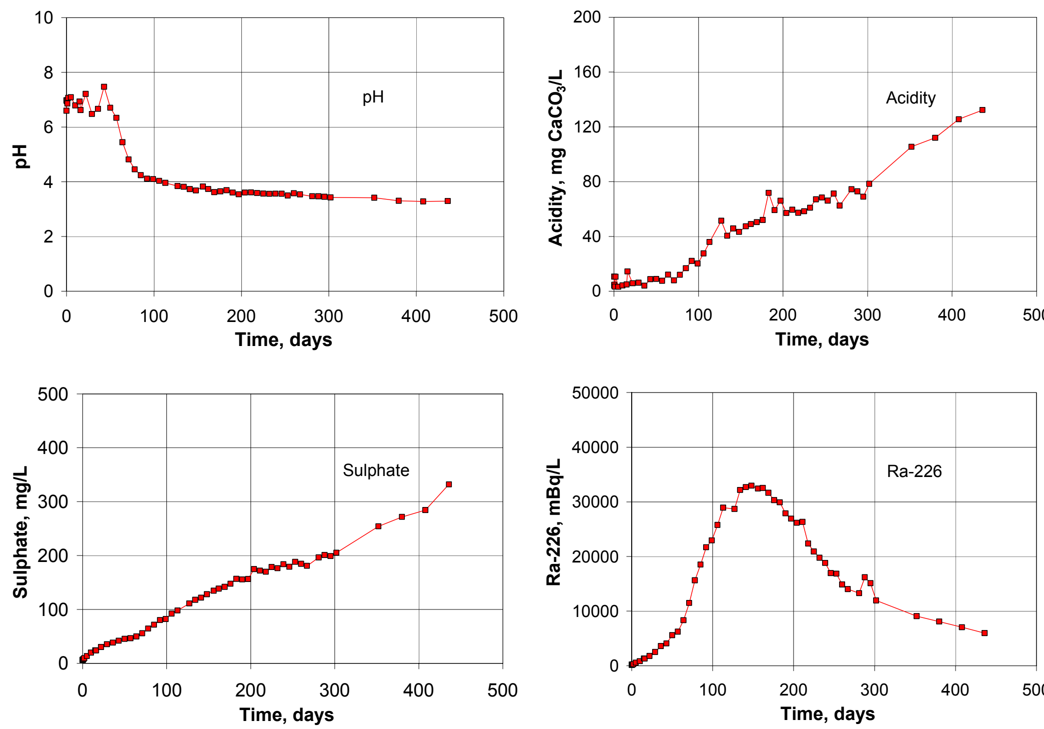

Figure 13. Variations of pH, total acidity, sulphate and Ra-226 concentrations in the surface water cover vs. time for the submerged coarse pyritic uranium tailings.

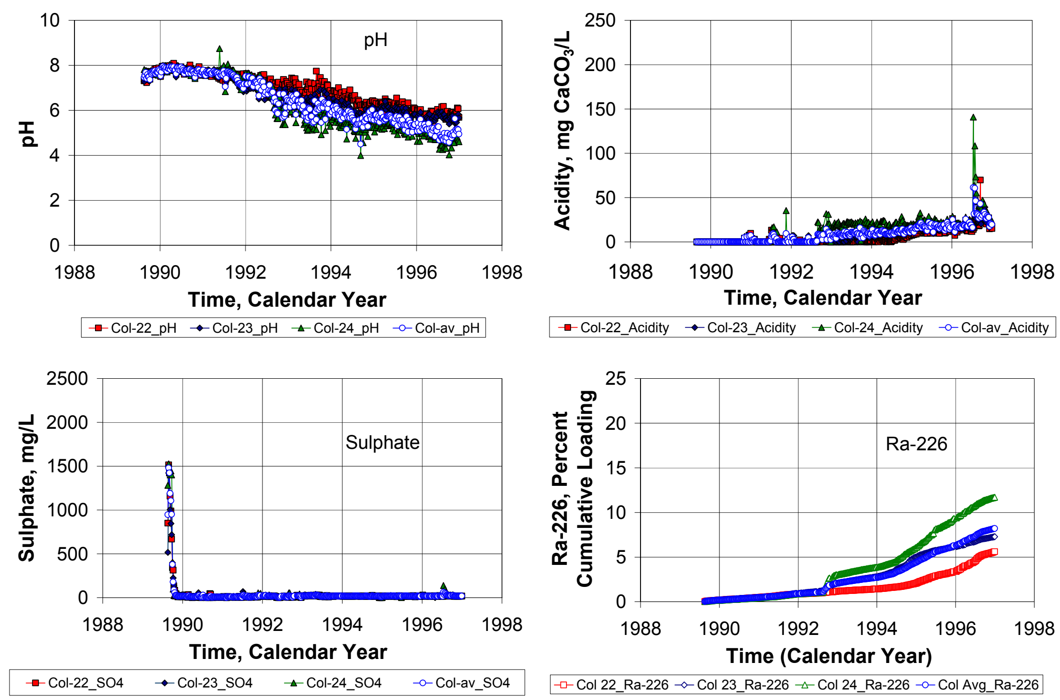

Figure 14. Variations of pH, total acidity, sulphate conc. and cumulative Ra-226 loadings vs. time in the porewater drainage for the submerged coarse pyritic uranium tailings.

For the porewater drainage study under shallow water cover conditions, the mobility and cumulative release of Ra-226 was significantly high at ~ 8% of the total Ra-226 contained in the coarse tailings, compared to ~ 0.2 % for the unsaturated zone leaching of the same tailings during the seven and a half year leaching period (Figures 10 and 14). The enhanced release of Ra-226 in the porewater drainage was concomitant with the breakthrough and increased drainage of dissolved iron in the porewater (Davé et al. 2002). Martin et al. (2003) also observed a significantly increased mobility of Ra-226 in the surface water cover as well in the shallow zone porewater of decommissioned pyritic uranium tailings in both coarse and fine tailings areas. The authors attributed the increased release of Ra-226 in the fine tailings areas to the microbial reduction of $(Ra)BaSO_4$ co-precipitate upon depletion of the sulphate ion solubility control in zones where a dense vegetation cover on the submerged tailings had contributed to anoxia, resulting in vegetative reduction of sulphate just below the benthic boundary.

Collectively, these results show that the increased mobility and release of Ra-226 may be a cause of long-term environmental concern at both on-land and underwater deposited uranium tailings when sulphate ion mobility control has been exhausted. Additionally, these findings may have implications on the long-term radiological stability of the effluent treatment sludge itself, containing $(Ra)BaSO_4$ precipitates along with those of gypsum and other metal hydroxides. While the sludge is stable during on-going treatment operations and as long as the sulphate ion solubility control is present, the loss of the latter under both oxic and anoxic substrate conditions, combined with microbial degradation of sulphate in reducing environments, has the potential to re-dissolve the immobilized radium. Because of the long-term stability aspects of the effluent treatment sludge, the decommissioned uranium mines in the eastern Germany cast the dewatered sludge in a concrete matrix and dispose the cast slabs at a suitable sub-terrain location.

It should, however, be emphasized that the sulphate solubility related control aspects apply only to the fraction of total radium that has been immobilized with the insoluble metal or other sulphates. Radium is also strongly attached to Fe-Mn oxy-hydroxides minerals, which are common to most uranium tailings and treatment sludge, and may play a key role in controlling the additional mobility of radium under both oxic and anoxic environmental conditions (Somot et al., 2002). The existing database on radium mobility, however, is still very limited to fully quantify the long-term radiological stability aspects of decommissioned uranium mine tailings, waste rock and effluent treatment sludge. Thus, further investigations in this area are warranted.

Summary and Conclusions

The rehabilitation and decommissioning of inactive uranium mine/mill and waste management sites in Canada have been very successful through the joint efforts of the mining industry, government and regulatory authorities. A majority of the past historic mine sites have been rehabilitated and managed under the current regulatory framework for such sites. Efforts are underway to develop decommissioning and management strategies for the remaining inactive and abandoned sites. All newer close-out mines in the Elliot Lake area, where acid mine drainage is a major issue of environmental concern, are decommissioned with engineered waste management facilities to provide a shallow, in-situ water cover having a minimum depth of ~1 m above the submerged waste. The water cover has reduced the acid generation at these sites to a low rate, and has minimized the exhalation of radon gas and its progeny from underwater deposited wastes. During active treatment and transition phases, all sites are regularly monitored and are on care and maintenance basis to meet regulatory requirements. Effluents at these sites would continue to be collected and treated until required.

During the acid generation phase, the mobility of Ra-226 was low. It was, however, enhanced upon depletion of the sulphate ion solubility control for both on-land and underwater disposal scenarios of pyritic uranium tailings. This may be a cause of long-term environmental concern and further investigations in this area are warranted.

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