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Uranium mill tailings: nuclear waste and natural laboratory for geochemical and radioecological investigations

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Abstract

Uranium mill tailings (UMT) are a high volume, low specific activity radioactive waste typically disposed in surface impoundments. This review focuses on research on UMT and related earth materials during the past decade relevant to the assessment of: (1) mineral hosts of radionuclides; (2) the use of soil analogs in predicting long-term fate of radionuclides; (3) microbial and diagenetic processes that may alter radionuclide mobility in the surficial environment; (4) waste-management technologies to limit radionuclide migration; and (5) the impact of UMT on biota.
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1. Introduction

Uranium mill tailings (UMT) are the crushed ore residues from the extraction of uranium (U) from ores. Except where ores have high carbonate content, the commercial extraction of U from ores generally involves leaching with sulfuric acid (H_2SO_4). The effluent (termed “raffinate” or “barren solution”) and tailings from the mill are discharged as a slurry to a waste-retention pond or to mined-out underground workings for disposal. As the ores typically are low grade, essentially all of the tonnage of ore processed at the mill is disposed of as tailings. The present inventory of UMT in the United States (US) is about 240 million tons. The final

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product of the milling process is a U concentrate called "yellowcake". U extraction efficiencies at commercial mills range from about 65 to 95% (Mudd, 2000). Because solubilization of ^{226}Ra from ore minerals in both sulfuric acid and sodium carbonate is low, the tailings remain a radiological hazard. Safe management of tailings has been the focus of regulatory and environmental research attention since the 1950s.

This report represents the third in a series of critical reviews by the author on the isolation of UMT and their component radionuclides from the biosphere. The first two reports (Landa, 1980, 1999) covered the literature through about 1992. The present report, as the previous ones, emphasizes geochemical and biogeochemical processes that influence the migration of radionuclides and other contaminants, and the linkage to radioactive waste-management and environmental monitoring practices. Some general observations on shifts and trends in research during the last decade are noted below:

(a) In the US, under the Uranium Mill Tailings Remedial Action (UMTRA) program of the US Department of Energy, approximately 50 million tons of UMT at 22 sites were moved and covered, or stabilized in place, between 1983 and 1998. The research and development phase of this program during the 1980s generated much information on radon emanation and transport and cover design guidance for radon control. There has been a marked decrease in the past decade in the number of studies focused on radon release from UMT. The shift of the UMTRA program to an emphasis on groundwater has spurred research on sorptive and microbially mediated controls on contaminant transport.

(b) Uranium mining and milling constitute the first steps of the nuclear fuel cycle. In the US, other activities associated with the front end of the fuel cycle, such as enrichment and fuel fabrication, have received increasing attention as new cleanup programs have been launched by the US Department of Energy (DOE) at sites such as Fernald, Ohio. Because of the raw materials handled at these sites, the contaminant of primary concern is U. At the same time, there has been greater emphasis placed in recent years on U as a contaminant of concern at UMT sites. This shift stems from several scientific, programmatic, and regulatory fronts. The UMTRA groundwater program has identified appreciable U plumes at several sites. In support of UMTRA and the other DOE-site cleanup programs, much research has been funded on U migration, and immobilization by subsurface microbial processes, phytoremediation, and permeable barriers. The UMTRA sites at Gunnison and Rifle, Colorado, and Shiprock, New Mexico, have served as field laboratories for these efforts. During this same era, U was added to the US Environmental Protection Agency's National Primary Drinking Water Regulations; a maximum permissible concentration of 30 $\mu g\ l^{-1}$ was established in 2000. Most recently, toxicological interest in depleted U munitions related to military operations in the Persian Gulf region and Kosovo has spurred new investigations of the environmental fate of U.

(c) Other industrial activities, not associated with the nuclear fuel cycle, have created large inventories of technologically enhanced, naturally occurring, radioactive material (TENORM). ^{226}Ra typically has been the radionuclide of concern here. The past decade has seen a large increase in TENORM studies worldwide (Goldstein et al., 1999). These wastes, which include radium ore extraction residues, oil field pipe scale, and phosphogypsum, include some good analogs for UMT. Research studies that have focused on the environmental fate of these related wastes can provide valuable information in UMT management.

(d) With shifts in U production to Australia and Canada as the world's major producers of yellowcake, there has been more research on contaminant migration in these environments. Also, with the relaxation of political restrictions, more reports in the open scientific literature are coming from eastern Europe and the former Soviet Union (FSU) on past U mining and milling practices and current conditions at sites there.

(e) Increasing attention has been focused on the occurrence of anaerobic conditions in UMT and their impacted aquifers, and the influence of anaerobic microbial processes on the mobility of U and radium associated with UMT.

(f) The past decade has seen a major increase in reports on radioecological impacts of U milling activities on microorganisms, plants, invertebrates, and higher animals. The easing of political barriers is creating the opportunity to access the wealth of radioecological studies done on the fate and impact of radionuclides associated with U ore processing in the FSU (Taskaev et al., 2003).

2. Mineral hosts of radionuclides

2.1. Amorphous silica

Most attention in recent years on mineralogic controls on contaminant mobility in UMT has focused on iron oxyhyroxide and alkaline earth sulfate phases. Silicate minerals make up the major component of most U ores, and colloidal silica has been noted briefly in several studies as a potential radionuclide sorbent in these UMT systems. Johnston (1990) did laboratory-scale, H_2SO_4 milling of Olympic Dam (Australia) ore and noted formation of large amounts of gelatinous silica during leaching. Langmuir (1997; personal communication) noted the silica content of a pH 1.2, filtered UMT effluent to be grossly supersaturated with respect to SiO_2 (amorphous). When this solution was allowed to stand, dissolved silica dropped from 946 to 180 mg l^{-1} (as SiO_2), and the concentrations of dissolved ^{226}Ra and ^{230}Th dropped by more than 90%. Coprecipitation with amorphous silica seems a likely cause of this drop of concentrations. In another filtered acid (pH 0.9) UMT effluent, dissolved silica dropped from 220 to 140 mg l^{-1} on standing. Thus, precipitation of amorphous silica may be a common, but little recognized, phenomenon in UMT leach solutions.

Nirdosh et al. (1987) prepared acid-cured quartz by treating finely ground, boiling-HCl washed quartz at 80 °C with 0.1 M H_2SO_4 for 30 h; free acid was removed by filtration and deionized water-washing. Sorption trials used soluble ^{226}Ra added in 0.1 M HCl. This H_2SO_4-treated material showed far greater retention of ^{226}Ra than untreated quartz (Fig. 1), or quartz treated for 30 h with 0.15 M HCl. The surface of the acid-cured quartz is likely composed of amorphous silica because of the grinding and acid treatment. Protonated silanol ($SiOH_2^+$) groups on this surface probably retain Ra by a combination of mechanisms:

(a) Specific adsorption of the weakly hydrated Ra^{2+} cation at the net positively charged surface present around pH 1 (specific adsorption of Ra at the goethite surface also has been noted by Bassot et al. (2000)), and
(b) Precipitation/coprecipitation of radium by sulfate counterions at this positively charged surface.

These radium retention properties are likely to be of importance in both the fresh (i.e., in-mill circuit) and aging (i.e., environmentally deposited) UMT. The

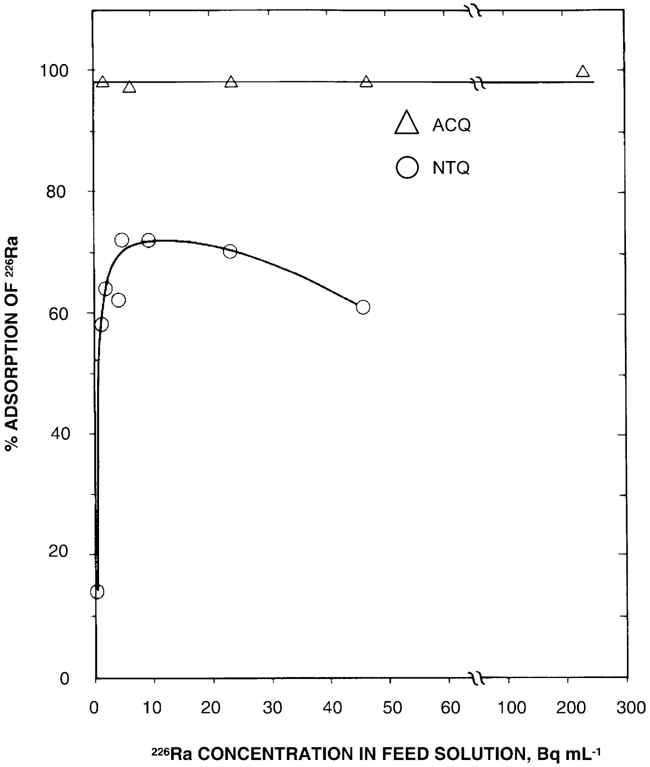

Fig. 1. Adsorption of ^{226}Ra on non-treated quartz (NTQ) and H_2SO_4 cured quartz (ACQ) as a function of radium concentration in the feed solution. Sorption conditions: 24 h; 24 °C; pH 1; solid/liquid ratio = 5 g/10 ml (modified from Nirdosh et al., 1987; with permission to use from the corresponding author).

sorptive and scavenging properties of amorphous silica surfaces and precipitates appear worthy of additional study.

2.2. Hosts of ^{230}Th

Krizman et al. (1995) have investigated the waste products at a H_2SO_4-leach U mill in Slovenia, where the raffinate solution is disposed of (after U removal) separately from the crushed ore residues. The latter tailings are lime-neutralized and disposed at one site ("Borst"). After solvent extraction of the U from acidic leachate phase, the barren solution is neutralized with lime. This neutralization produces a precipitate termed "red mud" that is disposed together with subgrade mining spoils at a second site ("Jazbec"). This unique disposal option was chosen because of the tendency of the red mud to retain moisture, and concerns regarding the geotechnical stability of materials disposed at the hillside Borst site. The distribution of radionuclides among these various materials is shown in Table 1. The high ^{230}Th content of the red mud is clear. In the five-year period of mill operation, about 593,000 tons of tailings and 48,000 tons of red mud were produced; the mud contained about 58% of the ^{230}Th in the processed U ore. The red mud presumably is a ferric hydrous oxide. The environmental fate of the coprecipitated ^{230}Th will likely be linked to that of the iron. For example, microbial iron reduction that might occur in anaerobic zones of the red mud buried under the spoils might mobilize ^{230}Th.

^{230}Th is responsible for the majority of the dose equivalent from inhalation intake of UMT. Reif (1994) has investigated the kinetics of ^{230}Th solubilization from acid-leach (Rifle, Colorado) and alkaline-leach (Ambrosia Lake, New Mexico) UMT (<38 μm sieve fraction of each) in simulated lung fluid. The alkaline-leach UMT showed much greater release, yielding about 30% in 1 month versus about 2% for the acid-leach tailings. These data suggest that large differences in bioavailabilty via other pathways may also exist for ^{230}Th in UMT, depending upon the processing history of the U ore.

The long-term radiotoxicity of UMT is controlled by the half-life of ^{230}Th. The segregated ^{230}Th (half life = 80,000 years) buried at Jazbec and the larger inventory of commingled ^{230}Th buried with the leached ore at conventional UMT sites are the parents of ^{226}Ra (half life = 1600 years). Thus, removal of the ^{230}Th will

Table 1
Radionuclide concentrations (Bq kg^{-1}) in ore and waste products at a former uranium mill in Slovenia. The average ore U_3O_8 content was 0.084% (range = 0.07–0.11%) and secular equilibrium with ^{230}Th and ^{226}Ra was assumed (modified from Krizman et al., 1995; with permission to use from the *Journal of Environmental Radioactivity*)

	^{238}U	^{230}Th	^{226}Ra
U ore (0.084% U_3O_8)	8780	8780	8780
Lime-neutralized uranium mill tailings at Borst site	995	3930	8630
Red mud at Jazbec site	495	65,100	190

prevent in-growth of ^{226}Ra. Nirdosh (2002) has shown how ^{230}Th might be removed from H_2SO_4 U mill leachate or barren solution by solvent extraction in order to limit ^{226}Ra production in the UMT, and thus limit the long-term radiological hazard.

2.3. Reaction products of acid, in-situ leaching (ISL)

In-situ leaching represents an alternative to conventional U milling in which extractant solutions are pumped into the in-place ore body; no tailings are generated at the surface. In contrast to the use of alkaline extractants, such as sodium carbonate at most ISL operations in the US, the extractant in China and the FSU has usually been H_2SO_4 for ISL of U deposits. Recent reports on these acid ISL operations (Mudd, 2001a,b) have revealed several features of geochemical note.

Plugging of ore bodies with jarosite [$KFe_3(SO_4)_2(OH)_6$] has been a problem (i.e., limiting permeability to leaching solutions and hence, uranium recovery), especially where pyrite content of the ore is high, and where ferric sulfate has been used as an oxidant for U(IV). Jarosite can be an important host for environmental contaminants such as lead, mercury, copper, zinc, silver, chromium, arsenic, and selenium, which are incorporated into the jarosite structure by ionic substitution at the K^+, $(SO_4)^{-2}$, or Fe^{+3} sites. Furthermore, jarosite has been shown to concentrate ^{226}Ra, with up to 2960 Bq g^{-1} (80,000 pCi g^{-1}) in magnetic separates of pyritic flotation concentrates from Elliot Lake, Ontario, UMT (Kaiman, 1978). Except for comminution, the residual ore body after ISL is essentially a subsurface tailings body. During the post-mining period at an ISL site, when either active restoration measures or passive flushing by upgradient groundwater are underway, lower sulfate and higher pH, or the action of sulfate-reducing (Ivarson et al., 1976) and iron-reducing microorganisms (Nadeau et al., 2000) may lead to jarosite dissolution and the release of ^{226}Ra and other contaminants. Other alkaline earth sulfate minerals such as barite ($BaSO_4$), which are hosts of coprecipitated ^{226}Ra, may likewise be mobilized, making restoration to pre-mining water quality problematic.

The presence of large quantities of a mineral phase—i.e., an amount sufficient to plug an economically important aquifer—makes characterization of subsurface mineralogy relatively easy (e.g., by X-ray diffraction (XRD)). When mineral quantities are below conventional detection limits by XRD, mineralogical characterization is typically left incomplete, despite the fact that such information is critical to simulation of contaminant transport. Amorphous materials present additional problems to XRD characterization. The limitations imposed by a lack of mineralogic data on modeling the natural attenuation of an acidic plume in the aquifer underneath a UMT pond in the western US have been highlighted by Zhu and Burden (2001) and Zhu et al. (2001,2002). They advocate detailed microscopic and spectroscopic examination of mineral surfaces to establish site densities of hydrous ferric oxide (HFO), amorphous aluminum hydroxide (AAH), and other reactive phases in aquifer solids, and to elucidate the nature and form of surface acidity in contaminated zones. With respect to AAH, the presence of a phase such as jurbanite ($AlOHSO_4 \cdot 5H_2O$), produced by the reaction of the H_2SO_4 plume with AAH, can profoundly influence the breakthrough curves of SO_4^{2-} and H^+ (Zhu and Burden, 2001). The flushing of affected portions of the aquifer with uncontaminated, upgradient groundwater causes the dissolution of jurbanite:

$$2AlOHSO_4 \cdot 5H_2O \; + \; HCO_3^- \Rightarrow Al(OH)_3 + Al^{3+} + 2SO_4^- + H_2CO_3 + 4H_2O.$$

It is important to know the relative roles of sulfate storage on phases like jurbanite and jarosite, as compared to sulfate sorbed on HFO, in order to simulate both the seepage of the acidic UMT pond solution into the aquifer, and then the flushing of the affected zone by uncontaminated, upgradient groundwater after the source of the contamination is terminated.

At some acid ISL operations in the Czech Republic, Ukraine, and perhaps other locations in the FSU, nitric (HNO_3) and hydrofluoric (HF) acid were added to the leaching solution (Rudy, 1997; Mudd, 2001a, b). These process variations clearly make the leaching less selective for U and open the possibility of release of larger inventories of contaminants. ^{226}Ra release from the leached residues is likely to increase with the addition of HNO_3. ^{230}Th mobility might be decreased by the addition of HF due to the insolubility of ThF.

2.4. Carbonate and phosphate minerals

The long-term retention of U by calcium carbonate minerals is a basic tenet underpinning U series dating of cave deposits, carbonate shells, and corals. However, on the basis of spectroscopic studies, some have argued that there may be no stable structural configuration for the uranyl ion in the calcite lattice. Studies by Kelly et al. (2003) of U-rich calcite from a speleothem deposit using X-ray absorption spectroscopy have shown that uranyl does indeed have a stable lattice position in natural calcite; the uranyl ion substitutes for a calcium and two adjacent carbonate ions. Biogenic aragonite in the shells of two species of freshwater mussels likewise appears to retain U following exposure of the organisms to a soluble spike (Markich et al., 2002). Recent investigations of coprecipitation of uranyl by biogenic calcite (Fujita et al., 2000) and abiotically produced (Reeder et al., 2000) calcite and aragonite are thus of interest when considering what aquifer minerals may be hosts for U migrating from UMT. Aragonite precipitated from groundwater samples collected at the Tuba City, Arizona, UMT site (U concentration = 44 $\mu g\ l^{-1}$) and allowed to stand in the laboratory at 24 °C for 6 months showed a U content of 0.04% (Abdelouas et al., 1998). The authors suggest that uranyl precipitation was associated with CO_2 produced from organic matter oxidation during denitrification occurring in the samples. Binding of uranyl by carbonate minerals also seems to be an important U immobilization mechanism in calcareous soils (Shahandeh and Hossner, 2002). Calcium carbonate also may coprecipitate ^{226}Ra (Reeder et al., 1999). Rhodocrosite ($MnCO_3$) may precipitate in groundwater at select UMT sites (e.g., the Grants, New Mexico, site; see Abdelouas et al., 2000); its ability to coprecipitate U and Ra has not been determined. Precipitation of carbonate minerals and their coprecipitation of U, Ra, and other contaminants should be recognized as a possible retention mechanism in aquifer materials at UMT sites.

Column studies by Flury and Harsh (2000) showed that apatite can react with acidic, U-bearing mine drainage water from the Midnite Mine in Washington to yield a U(VI) phosphate mineral, probably autunite [$Ca(UO_2)_2(PO_4)$]. Other research groups (Brendler et al., 1995; Ohnuki and Samadfam, 2001) have proposed the addition of dissolved phosphate or P-bearing solid phases to UMT to decrease U mobility. These amendments would appear to offer promise in aerobic disposal environments.

2.5. Contaminants associated with the back-end of the nuclear fuel cycle

As U milling is the earliest processing step at the front end of the nuclear fuel cycle, the radioactivity present in UMT should be restricted to nuclides in the naturally occurring U (^{238}U), thorium (^{232}Th), and actinium (^{235}U) decay series. The finding of elevated concentrations (i.e., above fallout levels) of fission products such as tritium and technetium-99 (^{99}Tc) in stream and groundwater samples in the vicinity of the Ellweiler U mill in Germany during decommissioning (Koss et al., 1992) was therefore a surprising finding. Between 1972 (when U ore processing, begun in 1961 at the site in the then West Germany, had ended) and 1989 (when the mill closed), residual materials (including excavated soil, slurries, and $UO_2(NO_3)_2$ solutions with depleted U) were processed at the mill for U recovery. The fission products present indicate that some of this material was from the back-end of the fuel cycle and had passed through a reactor.

The possible processing of back-end materials at other U mills or the direct burial of such materials in UMT disposal areas at other sites is not known, but should be considered in design of environmental monitoring programs and remedial action investigations at older sites. The practice remains under consideration by regulatory authorities at the present time. In terms of the direct disposal option, regulatory agencies have identified UMT impoundments as potentially cost-effective places to dispose materials from the reclamation and clean-up of other nuclear fuel cycle facilities (US Nuclear Regulatory Commission, 2000). In terms of the processing and then disposal option, it should be noted that the fate of fission products in U milling circuits has not been investigated. An awareness of the possible presence of such radionuclides will be of importance where alternate feeds (i.e., materials other than natural U ore) are allowed to be processed by U mills and the residuals disposed in mill-tailings impoundments. Among the areas of interest would be the partitioning of the fission products to the various commercial products (yellowcake), waste solutions and waste solids, and the fate of the fission products in the UMT disposal environment. The presence of mixtures such as U and Tc at such sites presents possibilities of interactions not previously considered at such sites; for example, the reduction of mobile Tc(VII) to immobile Tc(IV) by biogenic U(IV) (Lloyd et al., 2002).

3. Soil analogs

Rutherford and van Loon (1982) noted the similarities and contrasts between soils and the tailings from the mining and milling of ores. This view of tailings as a technologically modified, soil parent material can be of value in formulating conceptual models of the long-term fate and transport of radionuclides in UMT. Soil profile development will occur in UMT in response to climatic conditions and other soil-forming factors. The retention, or lack thereof, of radionuclides by components of these horizons (e.g., clay minerals, iron and manganese oxides, humic substances) will be key factors in plant uptake or release to groundwater and other environmental pathways considered when calculating population doses in long-term scenarios.

Geologic analog studies (e.g., the Oklo natural reactor in Gabon) have a well-established place in consideration of radionuclide migration from high-level radioactive waste repositories. Similarly, pedogenic analog studies can provide valuable information on radionuclide migration from UMT disposed in the surficial environment. For the analog approach to be successful, the element of concern must be released in the soil and directly influenced by pedogenic processes (Braun and Pagel, 1994; Aide and Pavich, 2002). Situations where U and Th (or rare earth elements that might serve as surrogates of Th and U(IV)) are locked up in primary minerals that are highly resistant to weathering would not represent good analogs to UMT, where considerable solubilization of ^{230}Th occurs in acid-milling circuits.

Recent studies by Evans and coworkers on soils in the northeastern US represent a case in point. Spodosols developed on hydrothermally altered Conway granite (known for its high content of U and Th), or glacial till in which the Conway granite was the largest lithologic component, were investigated. Using gamma-ray spectroscopy, U- and Th-series nuclides were shown to be deficient (with respect to parent material) in soil horizons, showing field evidence of removal of solutes and colloids by groundwater percolation. Statistical correlations suggested that leaching of both ^{228}Ra (or its precursors in the ^{232}Th decay series) and ^{226}Ra (or its precursors in the ^{238}U decay series) was enhanced in lower soil pH horizons (Evans et al., 1997). Selective extraction techniques were then used to assess the host phases of U and Th in the profiles. Based on ammonium oxalate and citrate dithionite extraction data, Th appears to translocate from the A horizon to the B horizon, presumably eluting as host iron oxides dissolve in the A horizon, and accumulating as new, hosting-iron oxides precipitate in the B horizon. Sodium pyrophosphate extraction data suggest that organic matter, as well as iron oxides, are important hosts for U in these soils (Morton et al., 2001). Insights gained by studies of natural soils can help to predict the movement of the larger quantities of these same radionuclides present in UMT. Migration rates will be a complex function of both downward leaching and recycling to the surface by plant uptake of bioavailable fractions of the mobile radionuclides.

Work being done as part of the US Department of Energy's Long-Term Surveillance and Monitoring Program (http://www.gjo.doe/programs/ltsm/) involves using the soil analog concept to evaluate ways in which pedogenic processes (e.g., aggregation of fine particles, formation of secondary minerals, development of structural planes and of root and earthworm channels that may act as preferential flow pathways) will modify the physical and hydraulic properties of engineered covers on UMT over time. Weighing lysimeters have been installed in vegetated, native soils with textural properties similar to cover materials used at nearby UMT sites. Profile features and vegetation have been preserved to approximate properties influencing water balance (infiltration, evapotranspiration) that might be expected to develop in the engineered cover material after long-term exposure to environmental conditions. By monitoring these analog soils with different vegetative covers over several years, it may be possible to estimate the amount of water that might percolate through the cover under different climatic conditions. This leakage represents water available for groundwater recharge and leaching of the UMT (Waugh and Richardson, 1997). Analog studies by Waugh (1999) near a remediated UMT site in Pennsylvania suggest that establishment of deeply rooted vegetation has the potential to severely degrade the low hydraulic conductivity of a compacted soil cover over its 200–1000 year design life.

4. Microbial processes

4.1. Sulfate and iron/manganese reduction

Because of the H_2SO_4 used to extract U from the ores at acid-leach mills, ^{226}Ra is believed to occur as a sparingly soluble sulfate phase, probably coprecipitated with Ba and other alkaline earth metals (Goulden et al., 1998). Sulfate-reducing bacteria have been identified in U heap leaching piles (Schippers et al., 1995) and UMT (Hard et al., 1999). Fortin and Beveridge (1997) note that sulfate reduction in sulfidic mine tailings is most likely limited by the supply of organic carbon. In UMT, organic carbon supplies may include solvents used in the milling circuit and discharged to the tailings impoundment (such as kerosene) and carbonaceous ores such as lignites. Cover and co-disposal practices at UMT such as the use of a wood chip cover (at the Dawn U mill in eastern Washington; US Department of Energy, Office of Environmental Management, 1999) and of municipal waste (Helling et al., 1998) can add bioavailable carbon sources to the underlying UMT. Sewage sludge and other organic amendments may also be used during decommissioning activities to promote establishment of a vegetative cover and to limit oxygen penetration and pyrite oxidation.

Laboratory studies by Landa et al. (1986) showed enhanced leaching of ^{226}Ra from acid-leach UMT by the action of sulfate-reducing bacteria. Recent studies by Martin et al. (2003) have shown the process to be operative in water-covered (>2 m water depths), acid-leach/lime-neutralized UMT at Elliot Lake, Ontario, Canada. Despite the input of oxygen from photosynthesis by a dense, standing crop of mat-like submerged aquatic vegetation (*Chara* sp.; “muskgrass”), anoxic conditions occur at the tailings-water interface due to oxygen consumption by microbial respiration of decomposing biomass. Sulfate-reducing conditions are evident at about 4 cm above the benthic boundary and persist into tailings. The flux of Ra and Ba into the overlying water is much higher in the regions associated with *Chara* as compared to shallow regions (<1.5-m water depths), where only sparse filamentous vegetation is present. Indeed, the observed increases in ^{226}Ra concentrations in the tailings pond water over a five-year period at this site seem to be related to the gradual increase in the spatial extent and density of this benthic flora. Martin et al. (2003) suggest that vegetation control measures (such as water-lowering to take advantage of ice scouring of the pond bottom during the winter) should be considered in order to minimize ^{226}Ra release from the submerged tailings to the overlying pond water. It is of interest to note that living *Chara* is known to concentrate radium strongly from water (Smith and Kalin, 1989). Thus, the presence of *Chara* not only indirectly influences redox conditions in the underlying tailings, but also probably directly affects the flux of ^{226}Ra to and from them.

UMT tend to be high in iron and manganese due to native concentrations in ores, iron introduced to processed ore by abrasion of iron rods in the grinding circuit, and additions of MnO_2 and ferric salts to the leaching circuit as oxidants for tetravalent U. Selective leaching trials have shown iron and manganese oxides to be important hosts of ^{226}Ra in acid-leach UMT (Landa, 1982; Somot et al., 1997). The dissolution of these oxides by the action of the Fe(III)-reducing bacteria can release radium to contacting waters (Landa et al., 1991).

Fe and Mn oxides/hydrous oxides can form in UMT by a variety of mechanisms:

(a) Where tailings waste streams are neutralized by the addition of process reagents such as MgO, CaO, or $CaCO_3$, or by the combination of acid- and alkaline-leach UMT (Landa, 1987) prior to discharge, these Fe–Mn precipitates may form rapidly and scavenge dissolved and colloidal contaminants, much as in an iron-coagulation treatment in a water treatment plant.
(b) Where tailings waste streams are not neutralized by such reagent additions, a "self-neutralization" process occurs in which residual acidity is consumed by reaction with tailings solids, such as clay minerals present in the ore (Fordham, 1993).
(c) Pre-oxidized, sulfidic, subaqueous UMT represent an important subclass. In these UMT, a period of surface disposal is followed by flooding and underwater storage. During the initial, aerial period, dissolution of sulfides leads to high ferrous iron concentrations in the porewaters. At the air-surface interface during aerial exposure, and at the water (with high dissolved oxygen content)-surface interface upon flooding, a layer of hydrous ferric oxide forms; this may age to goethite (Catalan et al., 2000; Martin et al., 2003). This upper few centimeters of tailings can be highly dynamic, with the retention or release of radionuclides and metals to the water column responding to the influences of environmental factors such as photosynthetic activity, turbulence-driven resuspension, and accumulation of organic sediments.
(d) As pH rises due to the processes described above, jarosite precipitated at an earlier, lower pH stage in the leaching circuit or after tailings discharge may dissolve and the iron reprecipitate as hydrous ferric oxide (for an analogous soil case, see Fanning et al., 2002).

4.2. Uranium reduction

Lovley et al. (1991) demonstrated the enzymatic reduction of soluble U(VI) to U(IV), which precipitated as uraninite (UO_2) (Gorby and Lovley, 1992). Since then, a large body of work has been carried out documenting the range of microorganisms involved, physiological constraints on the process, and applications to bioremediation of U-contaminated groundwater by in-situ and pump-and-treat operations. These studies have recently been reviewed by Lloyd and Macaskie (2000). UMT sites have been the focus of many of the field studies looking at both natural and accelerated bioremediation.

The presence of indigenous bacteria capable of U(VI) reduction was verified in groundwater sampled at three UMT sites (both acid- and alkaline-leach mills) studied by Abdelouas et al. (2000). The addition of organic carbon (optimally as ethanol) and phosphate (as sodium trimetaphosphate) was required to initiate the reduction of the U present in the groundwater downgradient of the mills. Investigations on sediment and water from the U-contaminated aquifer at the Shiprock, New Mexico, UMT site by Finneran et al. (2002) also suggest that the addition of a carbon source (optimally, acetate or glucose) would stimulate the rate of U removal by microbial reduction. Molecular characterization of the microbial community at the Shiprock, New Mexico, UMT site using DNA and phospholipid fatty acid biomarkers extracted from groundwater is underway in an effort to discern the relative importance of iron-reducing bacteria (Holmes et al., 2002) and sulfate-reducing bacteria (Chang et al., 2001) in in-situ U reduction.

Where microbial reduction of U(VI) occurs in anaerobic portions of aquifers, uraninite is deposited. In later years, these subsurface zones may be contacted by oxygenated groundwater. Thus, the possible reoxidation of biogenic uraninite is a concern in terms of the long-term success of bioremediation of U-contaminated aquifers. Suzuki et al. (2002) induced anaerobic conditions in sediment-water mixes from the Midnite U mine in Washington and showed that the average particle size of the biogenic uraninite produced by native microflora was only about 1.5 nm. Such small particles may be prone to rapid reoxidation, although studies by Abdelouas et al. (1999) on groundwater and sediment from the UMT site at Tuba City, Arizona, suggest that preferential oxidation of other aquifer components, such as the iron sulfide mackinawite, may limit reoxidation of biogenic uraninite. Another contaminant-mobility issue noted by Suzuki et al. (2002) is that facilitated transport of these U(IV) nanoparticles may be possible unless they are flocculated by solutes in the groundwater or physically trapped by extracellular polysaccharides.

The initial studies of microbial U reduction focused on soluble U(VI) species. Recent studies have shown that U(VI) sorbed species and the U(VI) lattice components in minerals may also be reduced to U(IV). For example, Haas et al. (2002) showed that U(VI) sorbed to kaolinite was readily reduced by *Shewanella putrefaciens*, but that the reduction of U(VI) sorbed on montmorillonite was much less effective. Frederickson et al. (2000) demonstrated that *S. putrefaciens* could reduce the U(VI) oxide metaschoepite ($UO_3 \cdot 2H_2O$) to the U(IV) oxide uraninite. Thus, microbial reduction of U should be considered a possible reaction at multiple stages in groundwater flow path investigation and simulation at UMT sites, even when sorption or coprecipitation occur. In addition to enzymatic U(VI) reduction by bacteria, abiotic, surface-catalyzed U(VI) reduction by Fe(II) has been reported by Liger et al. (1999). Surfaces such as nanoparticulate hematite can serve as both the sources of Fe(II) (by reductive dissolution) and the sorption surfaces for U(VI) at oxic/anoxic transition zones in aquifers at UMT sites and other settings. The authors suggest that at near-neutral pH and in the absence of excessive desorption of U(VI) by the formation of strong aqueous carbonate and hydroxy carbonate complexes, this reaction may be kinetically competitive with enzymatic reduction.

4.3. Nitrogen cycle

4.3.1. Nitrification

Ammonia is used in large quantities at U mills to precipitate the extracted U as ammonium diuranate (yellowcake). Nitrate-containing reagents were generally used in limited quantities, yet one sees nitrate contamination in surface and groundwater at many UMT sites. The nitrates appear to have formed by microorganisms using the abundant ammonia as a substrate. Populations of both autotrophic, ammonia-oxidizing bacteria (AAO) and denitrifying bacteria have been reported at UMT sites. Using molecular methods, Ivanova et al. (2000) have characterized the population of AAO in groundwater sampled in the contamination plume downgradient from the UMT at Shiprock, New Mexico. (Uranium milling ceased at this site in 1968; US Department of Energy, 1998). Groundwater at this site had concentrations of up to 7800 mg l^{-1} nitrate and 630 mg l^{-1} ammonia. Ivanova and coworkers estimated the AAO population size by measuring the concentration of the functional gene-marker encoding the ammonia monooxygenase (*amo*A), the enzyme mediating the first step in nitrification. A good correlation ($r = 0.76$) was found between the nitrate concentration and the quantity of *amo*A gene present in the groundwater (Fig. 2), supporting the view that the AAO mediate nitrification in this system.

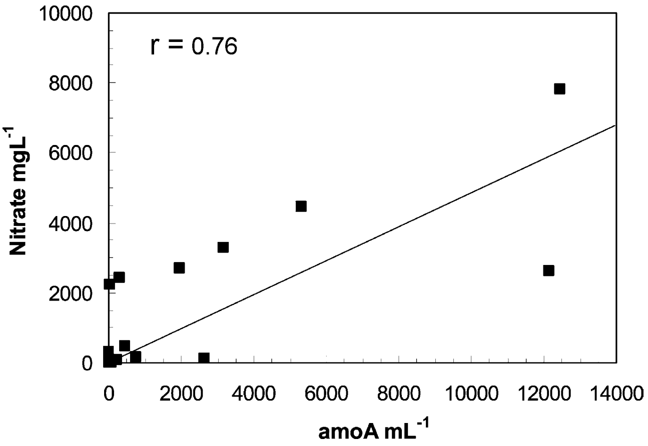

Fig. 2. Scatter plot of *amo*A and nitrate concentrations in groundwater sampled within contamination plume originating at UMT disposal site at Shiprock, New Mexico (modified from Ivanova et al., 2000; with permission to use from the *Canadian Journal of Microbiology*).

Besides its inherent role as a nutrient and groundwater contaminant, nitrate present within the tailings can influence radionuclide release from these materials. Anaerobic, nitrate-dependent, microbial oxidation of ferrous iron can increase pyrite oxidation (Straub et al., 1996; Landa et al., 2000) within the tailings, resulting in increased U and Th leaching and suppressed Ra release from solid phases such as $BaSO_4$. In addition, transport of nitrate from the tailings can potentially decrease rates of U reduction in the subsurface by providing anaerobic bacteria with a more energetically favorable terminal electron acceptor than U(VI) (Ivanova et al., 2000).

4.3.2. Denitrification

Longmire and Thomson (1992) have presented evidence for denitrification at the Maybell, Colorado, UMT site. In contrast to a dispersed chloride plume, nitrate concentrations decreased from two to four orders of magnitude immediately downgradient of the UMT impoundment. Microcosm studies using core materials from 10 to 40 m depth slurried with groundwater amended to nitrate levels close to the maximum groundwater concentration observed at the site all showed rapid denitrification, with an average nitrate removal of 40% in two weeks. No remedial action is planned for the Maybell site, and denitrification is one of the geochemical processes being relied on here for natural attenuation of nitrates (US Department of Energy, 2003). Barton et al. (1994) extended these studies and demonstrated by culturing techniques the presence of denitrifying bacteria in high-nitrate groundwaters at four UMTRA sites. Abdelouas et al. (1998) showed the presence of two species of denitrifying bacteria (*Psuedomonas aeruginosa and P. stutzeri*) in groundwater sampled at the alkaline-leach, Tuba City, Arizona, UMT site. In anaerobic microcosm experiments with C and P amendments to optimize reaction rates, the nitrate concentration was lowered by denitrification from about 1400 mg l^{-1} to <0.1 mg l^{-1} in 10 days at 24 °C.

In contrast to the Maybell site described above, the buildup of nitrate and lack of nitrite at Shiprock (Ivanova et al., 2000) suggest that denitrification is limited at that site. As nitrate is a contaminant of concern at many UMT sites, these contrasting results between the Maybell and Shiprock sites are of real interest. Recent landfill studies by Senko et al. (2002) have shown the intermediate products of dentrification (nitrite, nitrous oxide, and nitric oxide) were able to oxidize U(IV) and thereby mobilize previously reduced U. Environmental factors, such as availability of carbon, that might be controlling rates of denitrification in the subsurface sediments at UMT sites, would appear to be an area of considerable importance when considering U mobility.

The application of soil analog studies to UMT sites has been noted earlier. Information on microbial processes occurring at UMT sites may similarly be gleaned from investigations at other waste-disposal sites, including conventional landfills. Recent examples include investigations at the Norman, Oklahoma, landfill research field site that have demonstrated the dissolution of alkaline earth sulfate phases (e.g., $Ba(Ra)SO_4$) by the action of sulfate-reducing bacteria (Ulrich et al., 2003), and the microbial reduction and subsequent remobilization of U(VI) (Senko et al., 2002).

4.4. Fungi

Kalin and Stokes (1981) reported on the occurrence and metal content of macrofungi at abandoned UMT sites in the Elliot Lake and Bancroft areas of Ontario, Canada. The authors noted the consistent association of two of the fungi with balsam poplar roots and suggested that mycorrhizal associations might aid in the establishment of vegetation on UMT by enhancing nutrient uptake and/or restricting uptake and translocation of heavy metals. Recent work by Rufyikiri et al. (2002) has shown that the arbuscular mycorrhizal (AM) fungus, *Glomus intradices*, can assimilate U from solution culture and translocate it to excised carrot roots. Future research may determine if the U hyperaccumulates in the AM-colonized roots, or if enhanced U translocation to shoots occurs. This research has great promise in designing phytoremediation strategies for UMT and U-contaminated soils.

Zhdanova et al. (2003) have shown the tendency of some soil fungi to grow toward radioactive sources ("radiotropism"), colonize, and promote the dissolution of hot particles (HP) from the Chernobyl reactor accident and the Semipalatinsk nuclear weapons test site. Bioaccumulation of radionuclides, in this case ^{137}Cs and ^{152}Eu, was demonstrated, even in the absence of hyphal contact with the HP, suggesting a role of exometabolites in the dissolution of the HP constituents. It has been suggested that mycorrhizal fungi may also increase the release of calcium from apatite in soils (Blum et al., 2002). Stieff (1984) noted the presence of ^{226}Ra HP in UMT at Monticello, Utah. Landa et al. (1994) showed the presence of ^{210}Pb-bearing $PbSO_4$ HP that precipitated from laboratory-scale U milling solutions. The radiotropic behavior of select soil fungi perhaps may have a role in determining the long-term physical and chemical stability of such sparingly soluble HP in UMT piles and the surrounding depositional environments.

5. Diagenesis

Studies by Somot and colleagues of acid-leach UMT in France and Africa have revealed significant diagenetic changes in periods of two to three decades. Selective extraction trials suggest that gypsum is a major host of radium in fresh, acid-leach/lime-neutralized UMT (Somot et al., 2000). Following discharge from the mill and with exposure to infiltrating rainwater, this relatively soluble gypsum dissolves, releasing the coprecipitated ^{226}Ra to interact with surrounding tailings components. For aged tailings, the predominant ^{226}Ra hosts seem to be iron oxyhydroxides and barium and lead sulfates (Somot et al., 1997, 2000; Pagel and Somot, 2002). The French investigators have studied water-saturated tailings at Jouac in which red UMT derived from granite ore overlie green UMT derived from a gneiss ore. The green unit is characterized by low dissolved oxygen concentration, an absence of nitrates, high dissolved iron in the porewater, and siderite and ankerite (a calcium, ferrous iron carbonate mineral) in the solid phase. Acidophilic, Fe(III)-reducing and sulfate-reducing microorganisms are present, but appear to be nutrient-limited. Within the red tailings, radium released by the dissolution of gypsum is sorbed by iron oxyhydroxides, limiting migration. In the green unit, radium released by dissolving iron oxyhydroxides appears to be coprecipitated by barium sulfate, the barium coming from the in-pile weathering of metamorphic silicate minerals. Radium release by the action of SRB appears to be unimportant in this unit, and porewater ^{226}Ra concentrations are low (Pagel and Somot, 2002; Somot et al., 2002). Thus, we see that multiple, sequential reactions (sorption by iron oxyhydroxides, coprecipitated by barium sulfate) tend to keep radium immobile.

Recent studies in France have focused on the role of iron oxyhydroxides in this process. Bassot et al. (2000) have studied the sorption/desorption of radium on goethite; specific sorption below the zero point of charge was noted. Studies in progress by Lefebvre (2002) at the University of Paris are examining the fate of radium during crystallization of amorphous iron oxyhydroxides to crystalline phases such as goethite. Such diagenetic processes are underway in aging UMT and may influence the long-term behavior of radium in these systems.

6. Covers and subsurface barriers

For UMT exposed to the atmosphere, wind erosion is a concern. Thomas (2000a, c) has observed a freeze-drying phenomenon in tailings discharged during winter at the Key Lake UMT site in northern Saskatchewan. These non-cohesive particles are readily carried by late winter winds and deposited on neighboring soils and vegetative surfaces. At older UMT sites in the US, cover design focused primarily on a low permeability, compacted soil layer, protected from erosion by riprap, to limit radon emissions and provide for longevity of burial. In the mid-1980s, with implementation of groundwater quality standards for UMT sites, concerns increased regarding the need to limit infiltration of meteoric water into the buried UMT, and new cover designs evolved (Smith, 1999). A "water balance" approach has been employed in the cover design at Monticello, Utah, and uses a multilayer, 170-cm thick topsoil zone above a 90-cm-thick, geotextile/sand capillary barrier and compacted soil cap above the UMT. The topsoil is designed to store water while plants are dormant for later root uptake and transpiration during the growing season. The system thus helps to maintain a vegetative cover in this semiarid region, while limiting infiltration of moisture into the tailings. Gravel incorporated into the surface layer helps to create an erosion-limiting desert pavement over time. A discrete cobble layer is used within the topsoil to limit animal intrusion. The capillary barrier helps to maintain moisture in the topsoil, and the compacted soil layer at the base acts as a radon barrier (Waugh and Richardson, 1997).

Investigations by Lewis and MacDonell (1990) suggest that deeply rooted plants can act as conduits for radon (Rn) from buried UMT. Such transport can compromise design criteria for suppression of Rn release by cover materials. ^{222}Rn release from corn and sunflowers grown on limed, acid-leach UMT was shown to be proportional to leaf area. Radon appears to be transported via mass flow of water from the roots up the xylem. However, unlike transpiration of water, no day versus night differences in Rn release were observed, suggesting that release of Rn to the atmosphere occurs by diffusion through the leaf cuticle rather than through the stomata. Continued advances in modeling of Rn fluxes from UMT are reflected in the work of Ferry et al. (2002), whose TRACI model calculates moisture-content profiles and water and air permeability in UMT and cover materials under varying meteorological conditions. The TRACI model does a good job of predicting the effectiveness of Rn control, based on the thickness and textural properties of the cover material.

Geochemical barriers for contaminant removal have been proposed to line the bottoms of UMT impoundments and have been emplaced in groundwater plumes as permeable reactive barriers (PRB) at U mill and mining sites. Morrison and Spangler (1992) surveyed a wide range of low-cost, solid sorbents (including fly ash, sawdust, peat, lignite, coal, zeolites, ferric oxides, phosphate rock) and precipitants (including hydrated lime) as possible liner materials for a new UMT repository at Monticello, Utah. U and Mo were the target contaminants, and synthetic UMT pore fluid (pH 7.8) was the matrix solution tested. Sphagnum peat moss proved to be effective for removal of both U and Mo. Its potential role as a geochemical barrier has been demonstrated in the field at the Rayrock UMT in the Northwest Territories, Canada. Core samples showed an 8-cm thick peat zone that occurs naturally under 50 cm of unlimed, acid-leach UMT to be a good sorbent for both U and Ra (Veska and Eaton, 1991). Most attention with respect to PRB has focused on U removal by iron- and phosphate-bearing phases; the subject is discussed in detail in the recent volume by Naftz et al. (2002). Komarneni et al. (2001) synthesized a mica with high selectivity for radium uptake. Distribution coefficients for ^{226}Ra uptake from 0.5 M NaCl for this material was about from 850 to 1040 ml g^{-1}, as compared to from 12 to 44 ml g^{-1} for natural montmorillonite samples. This mica, synthesized by reacting kaolinite with NaF, may have applications in PRB at U mining and milling sites.

7. Biota

7.1. Bioaccumulation of radionuclides and metals by plants and animals

Baumgartner et al. (2000) have investigated the uptake of trace elements from Sudan grass and salt bush (a native species in the American southwest) irrigated with groundwater contaminated by U mill operations at Tuba City, Arizona. Above-ground tissue showed elevated concentrations (above background levels) of Se, U, and Mo (Sudan grass only). As might be expected when applying U in a soluble form, concentration ratios were higher than typically observed from contaminated soils. Selenium has been a frequent contaminant of concern when dealing with the impacts of U mining. Recent studies have demonstrated Se uptake by vegetation on U-mine overburden (Hossner et al., 1992; Sharmasarkar and Vance,

2002) and on land irrigated with wastewater from in-situ U mining operations (Ramirez and Rogers, 2002).

The bioaccumulation and toxicological effects of contaminants (nutrients, trace elements, radionuclides) associated with UMT on vertebrate and invertebrate species will be dealt with briefly here, with a focus on implications for environmental monitoring.

In the Key Lake U mining district of northern Saskatchewan, in-situ toxicity testing (using larval fathead minnows as the test organisms) was conducted in two lakes receiving treated U-mill effluent. Minnow mortality was significantly elevated in these two lakes compared to a control lake upstream of mining and milling discharges. The likely toxicant appears to be Se, which is accumulated in dietary sources (e.g., plankton) consumed by the minnows (Pyle et al., 2001). Peterson et al. (2002) have looked at trace element contamination in benthic macroinvertebrates collected from Montezuma Creek, a stream that flows through the Monticello, Utah, UMT site. Mean concentrations of As, Mo, Se, and V were two to four times higher at the two sampling sites immediately downstream of the tailings site as compared to upstream and control stream locations.

Bioaccumulation of radium in bone is well established in the human medical literature. Mirka et al. (1996) have measured ^{226}Ra in surface waters of the Elliot Lake, Ontario, U mining and milling area, as well as bones from muskrats trapped near these water-quality stations. Muskrats have a limited feeding and movement range (generally less than 150 m for individuals), and therefore their body composition has the potential to mirror local water-quality conditions. These investigations showed a good correlation ($r = 0.74$) between the total ^{226}Ra concentration in water and the ^{226}Ra concentration in muskrat bones, suggesting that these bones are a good biomonitor of environmental ^{226}Ra concentrations. The ^{226}Ra content of mink and otter bones in animals sampled in this same area did not decline from 1986 to 1997, despite the fact that active U mining/milling operations wound down and then ceased in the watershed during this period (Dewit et al., 2002), indicative of the long-term nature of the UMT as a ^{226}Ra source.

Snail shells may also act as bioaccumulation sinks for radium at UMT sites, again reflecting the concentration by ionic substitution in calcium-rich tissues (Gaso et al., 1995). While U is less recognized as a "bone seeker", it also tends to accumulate in calcium-rich tissues. This behavior reflects the ability of the uranyl ion to substitute for Ca in skeletal hydroxyapatite. Laboratory feeding studies (trout pellets spiked with uranyl acetate) on lake whitefish by Cooley and Klaverkamp (2000) suggest that fish scales on a variety of resident species may serve as readily sampled tissues for assessing bioavailable concentrations of U in aquatic habitats impacted by U mining and milling.

Rippon and Riley (1996) looked at the ecological effects of a UMT impoundment dam breach. Their investigation employed toxicity-testing protocols to assess the impact on biota in downstream floodplain environments. A multi-step experimental approach was adopted to address a variety of environmental processes and exposure scenarios. Tailings fines (<64 μ fraction from the Rum Uranium Mine in the Northern Territory, Australia) were washed extensively (to a constant electrical conductivity) prior to testing to simulate the leaching of readily soluble salts that would occur with stream transport prior to deposition in low-energy environments. Exposure of organisms to suspended UMT (juvenile mussels used here as test organism), and to overlying water in likely deposition sites in the floodplain (species of hydra and water fleas used here as test organisms), was examined. Various ratios of tailings to floodplain sediments were evaluated; other variables included a comparison of aeration or no aeration during a two-month incubation period of these tailings/sediment mixtures with native water. The final wash water showed markedly elevated concentrations of NH_4, SO_4, Ca, Mn, and U compared to native waters, reflecting solute contributions from the UMT. While only minimal toxic effects on the test species were noted, the authors stressed the need to examine the effects of aging of tailings on their toxicity.

The Olympic Dam operation in South Australia processes ore for both Cu and U recovery. The mill here includes an anode furnace, where a natural gas-fired process is used to refine the extracted Cu. Volatilization of ^{210}Po in this smelting unit and its release to the environment with stack gases may be responsible for elevated concentrations of this radionuclide in the tissues of herbivorous lizards collected at the tailings disposal area (Read and Pickering, 1999). Bunzl et al. (1995) have observed that a silt loam surface-soil sample collected near the exhaust ventilation shaft of a U mine showed a much greater fraction of readily exchangeable (1 M $MgCl_2$) ^{210}Pb than another silt loam about 200 m away. The attachment of this radon decay product to the surface of mine dust particles coming from the exhaust shaft was suggested as the likely cause. The volatilized ^{210}Po emitted from the anode furnace stack will likely also attach to the surfaces of stack and atmospheric dust particles and be readily bioavailable upon deposition in soils and water bodies.

7.2. Radiation effects studies

Studies in aquatic and terrestrial environments have looked at radiation doses to macroinvertebrates that burrow in sediments and soils impacted by UMT and related NORM wastes. While limited in number, these studies suggest that deleterious effects associated with ionizing radiation may be possible in such environments. Warwick et al. (1987) showed a greater incidence of mouth-part deformities in larvae of the midge *Chironomus* spp. collected in Lake Ontario sediments at Port Hope Harbour, Ontario, the site of a radium recovery plant and a U refinery whose early waste-management practices are believed to have contaminated the sediments. The radiation dose rate to chironomids in these sediments was estimated to be about 1 mGy day^{-1}. Other contaminants such as heavy metals are also enriched in the sediments, so that non-radiological toxic effects cannot be ruled out (Hart et al., 1986). Platford and Joshi (1988) have calculated doses from ^{226}Ra of up to 500 mGy year^{-1} (1.4 mGy day^{-1}) to worms in the sediments of a Saskatchewan lake receiving U mine/mill tailings.

At a site in the Komi Republic of northwestern Russia (Taskaev et al., 2003), soils are contaminated with radium-rich brines and have ^{226}Ra concentrations of about 26 Bq g^{-1} (700 pCi g^{-1}) and dose rates to soil invertebrates of about 10 mGy day^{-1} (1 rad day^{-1}). Krivolutzky (1987) has shown decreases in population size and reproduction rates, and histological changes in mid-gut epithelium in earthworms at this site compared to uncontaminated control sites. This work, taken together with the work of Warwick et al. (1987), suggests that somatic effects are possible in invertebrates in the dose range seen at UMT sites.

Thomas and Liber (2001) have presented a detailed description of a method for estimating internal and external dose to benthic invertebrates from the principal radionuclides in the ^{238}U decay series, emphasizing the strong dependence of calculated doses on assumptions made regarding weighting factors used for alpha radiation. They estimated doses to larval midges and adult amphipods of about 550 mGy $year^{-1}$ (1.5 mGy day^{-1}) at sites downstream of the Rabbit Lake U mining and milling operations in northern Saskatchewan.

Two recent studies have used molecular biomarkers to measure DNA damage in UMT environments. Au et al. (1995, 1998) examined blood samples collected from 24 humans living within 1.6 km of two U mining and milling sites and one U mining-only site in southern Texas (“exposed” population). These samples were compared with a control group of 24 individuals located from about 13 to 16 km south (predominantly upwind) of the mining/milling areas. Uranium concentrations in surface soils and household dust served as surrogates for exposure. The soils and dust samples in the exposed areas had U concentrations about 6 to 25 times those in the control sites. Radon concentrations in the homes of the exposed and control individuals did not differ. More (but not a statistically significant increase) chromosome aberrations were seen in the exposed individuals than in the controls. When the blood cells were challenged with gamma radiation (1 Gy), a statistically significant increase ($p < 0.05$) appeared in total (all types) and deletion-type chromosome aberrations in the exposed group compared to the controls. The gamma challenge assays suggest abnormal DNA repair capabilities in the cells of the exposed individuals.

Ptacek et al. (2002) measured DNA damage in a perennial grass species [*Calamagrostis epigejos* (wood small-reed)] growing on acid-leached UMT at a site in the Czech Republic. The tailings had a ^{226}Ra concentration of about 11,800 Bq kg^{-1} (318 pCi g^{-1}). The gamma dose rate (1 m above tailings) was 1000 pGy s^{-1} (0.1 mGy day^{-1}). A Comet assay, which measures DNA strand breaks, crosslinks, and incomplete repair sites, was used in cells from the leaves of plants growing atop the UMT and control plants from soils in the Prague area. Positive controls consisted of plants acutely exposed to 30 Gy of ^{60}Co gamma irradiation. No significant increase in DNA damage was observed in the plants sampled at the UMT site as compared to those in the control site. There is a clear need for more data to assess the biological impact of chronic irradiation of organisms at or near UMT sites.

7.3. *Simulation of radiation transfers and dosimetry*

Environmental modeling and risk assessment will be dealt with only briefly here. The International Atomic Energy Agency, under its Biospheric Model Validation

Study—Phase II (BIOMOVS II) program, completed a major testing of models designed to predict the environmental transfer and bioaccumulation of radionuclides and non-radioactive trace contaminants. One of the test scenarios looked at long-term (10,000 years) contaminant migration and impacts from UMT. Biogeochemical processes considered in the modeling included: dispersion of soluble contaminants in groundwater plumes, sorption of soluble contaminants by soils and aquifer materials, erosion and deposition of particulate-associated contaminants in lake bed sediment (aerobic and anaerobic), plant uptake of bioavailable contaminants, and soil ingestion. The members of the ^{238}U decay series and As, Ni, and Cr were studied; details may be found in BIOMOVS II (1995) and Camus et al. (1999). For such long-term dosimetric assessments, the need to consider in-growth of ^{210}Pb ($T_{1/2} = 22.3$ years) and^{210}Po ($T_{1/2} = 138$ days) from the parent ^{226}Ra ($T_{1/2} = 1600$ years) in biosphere components such as surface soils was stressed. The BIOMOVS II working group also noted that simplifying assumptions, such as a uniform physical half-life in soil (i.e., all removal processes except radioactive decay) for all U-series radionuclides, should be avoided. More recent project activities have examined the long-term stabilization of UMT (BIOMOVS II, 2003). Proposed IAEA activities under the Environmental Modeling for Radiation Safety (EMRAS) program will look further at U/Th mining and milling wastes and related, naturally occurring, radioactive material (NORM) wastes, such as oilfield pipe scale and zirconium extraction tailings.

Thomas (2000b) suggests that measures of bioavailable radionuclide concentrations rather than total radionuclide concentrations be used in calculating concentration ratios for food-chain transfer models at UMT sites. For example, laboratory extraction of soils from impacted sites with ammonium acetate (at near-soil pH) extraction for the soil-to-plant pathway or with solutions simulating conditions in the gastrointestinal tract for the soil-ingestion pathway may be of value when considering the soil-vegetation-small mammal food-chain. Studies downstream of the Rum Jungle UMT site in Australia have shown that the shells of freshwater mussels can serve as archives for bioavailable U and other metal contaminants in stream water (Markich et al., 2002). Annual laminations in the shells and the use of secondary ion mass spectrometry for analysis allowed for monitoring over the lifetime of the test organisms, here about 15–20 years. Thus, these shells can serve as retrospective monitors of water chemistry. While it seems likely that ^{226}Ra also substitutes for Ca (both alkaline earth metals) in the shell aragonite, analytical techniques are not available to measure its zonation in the annual laminations.

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