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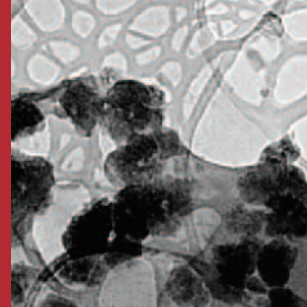
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Uranium Mill Tailings: Geochemistry, Mineralogy, and Environmental Impact

Abdesselam Abdelouas*



Worldwide, the mining of uranium has generated $938 \times 10^6 \text{ m}^3$ of mill tailings. The radioactivity of these tailings depends on the grade of ore mined and varies from less than 1 Bq/g to more than 100 Bq/g. The most common mode of disposal is near-surface impoundment in the vicinity of the mine or mill. The principal radiation risks from uranium tailings are gamma radiation, essentially from radium decay; windblown radioactive dust dispersal; and radon gas and its radioactive progeny, which are known to cause lung cancer. Uranium mill tailings are also often associated with elevated concentrations of highly toxic heavy metals, which are a major source of surface and groundwater contamination. Due to their high sulfide content (a few to tens of wt%), tailings may acidify groundwater, accelerating the release of radioactive and hazardous elements.

KEYWORDS: uranium mill tailings, tailings composition, environmental impact

INTRODUCTION

The nuclear fuel cycle includes uranium mining (ore extraction), milling (physical and chemical extraction of U from the ore), conversion (into gaseous UF_6), enrichment (up to 3.5–5% ^{235}U), fuel fabrication (UO_2 pellets encased in metal tubes), power generation (fission in nuclear reactors), and storage of used fuel (either for reprocessing or deep geologic disposal). Radioactive wastes are produced at each step of the fuel cycle, but the largest volume consists of mine and mill tailings, which contain all of the naturally occurring radioactive elements found in uranium ore. Typically, a light-water reactor (LWR) producing 900 megawatts of electrical power uses 20 metric tonnes (t) of metallic U per year. The fuel is enriched to about 4% ^{235}U from roughly 160 t of U from a UF_6 feed by the expenditure of about 100,000 SWU (separative work units have the dimension of mass, kg-SWU, and are a measure of the amount of work required to produce a specified level of ^{235}U enrichment) (IAEA 2005). Producing 20 t of metallic U fuel ($\sim 22.7 \text{ t UO}_2$) requires the mining of 17,000 t of 1 wt% uranium ore. At present, uranium is mainly produced in Australia, Canada, China, Kazakhstan, Namibia, Niger, the Russian Federation, South Africa, the USA, and Uzbekistan. Canada produces the largest share of uranium (29% of the world supply), followed by Australia (21%) (UIC 2005).

Today, 443 nuclear power plants are in operation with a total net installed capacity of 370 gigawatts (GWe) of electrical output, and 24 nuclear power plants are under construction (IAEA 2005). In 2003, 36,000 t of metallic U were produced, which is about half of the annual utilities requirements (66,000 t U). The balance is obtained from

recycled uranium and plutonium from spent fuel, weapons-grade uranium, and civilian stockpiles. Due to diminishing uranium stockpiles, it is expected that mine production will increase after 2015. Since 2002, market prices have increased by more than 100% ($\sim$ US\$45 per kg U). The increased demand and mining activity will lead to a substantial increase in the volume of uranium mill tailings. Known proven resources recoverable with current technology amount to a total of 4,588,700 t metallic U (OECD 2004).

Uranium is mined by three basic processes: surface (open pit), underground, and solution mining

(in situ leaching). The type of extraction process depends on grade, size, location, and geology, as well as on economic considerations. In 2004, production came from open pit mining (27%), underground mining (40%), in situ leaching (21%), and byproducts (12%) (UIC 2005). Both open pit and underground mining produce wastes, which are comprised of overburden (soil and rock covering the ore deposit, containing trace amounts of the ore and radioactive decay products); subeconomic ores that have too little uranium to be profitable; barren rock containing no ore; and drill cuttings. In situ leach mining causes little surface disturbance and leaves no tailings but can still lead to groundwater contamination. In a uranium ore deposit, secular equilibrium is reached between ^{238}U and ^{235}U and their decay products. The equilibrium may be somewhat disturbed by geochemical processes in the ore deposit. The ^{238}U decay chain (Fig. 1) includes ^{230}Th (half-life of 77,000 years), which decays slowly, producing ^{226}Ra (half-life of 1600 years). The ^{226}Ra decays slowly, producing ^{222}Rn (half-life of 3.8 days), an alpha-emitter, which when inhaled can cause lung cancer. After U extraction from the ore, the mill tailings contain virtually all of the radionuclides in the U decay series, especially those of ^{238}U . Thus, it is important to determine the geochemistry and mineralogy of these radionuclides, in particular ^{230}Th and ^{226}Ra , which have the longest half-lives. Unlike Th and Ra, radon gas reaches the environment (atmosphere) mainly by seepage through fractures and overlying soils.

Worldwide, the total estimated volume of tailings is 938 million cubic meters produced at more than 4000 mines (IAEA 2004). TABLE 1 lists the geographic distribution, age, and number of mines worldwide. The data are incomplete and approximate, but still show that uranium mining and the associated tailings are widespread. Kazakhstan has produced by far the largest volume of tailings ($209 \times 10^6 \text{ m}^3$),

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The uranium-238 decay chain

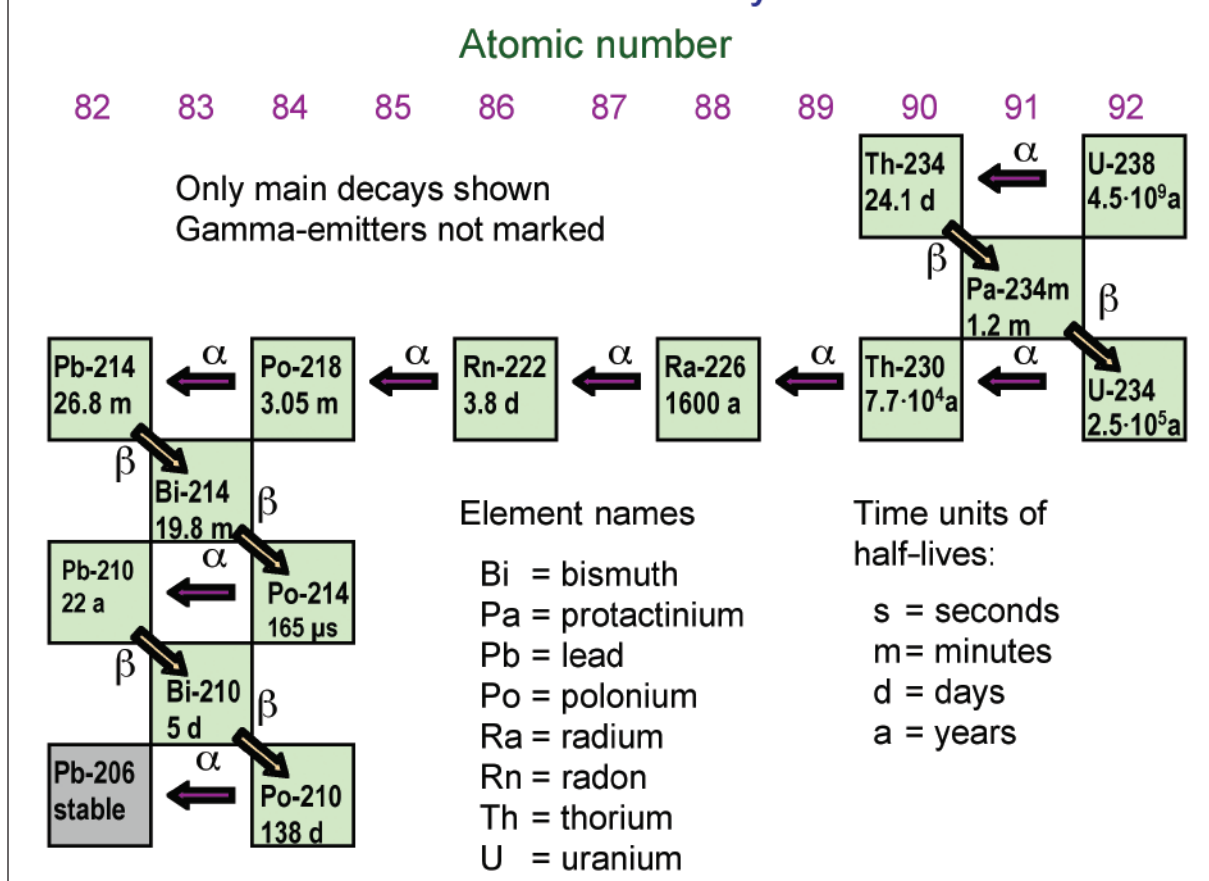


FIGURE 1 The ^{238}U decay chain scheme

which represents 23% of the world inventory of mine and mill tailings. Tailings production in the USA and Canada represents 13% and 3% of the global inventory, respectively. With the advent of a free market for uranium, many smaller, high-cost mines became uneconomic and have closed down (e.g. in France, Portugal, and Hungary). The United States, Germany, and France (Fig. 2) have completed the surface remediation of most of their mill tailings sites, while the former Soviet Union and Eastern Bloc countries, including Ukraine, Kazakhstan, and Kyrgyzstan, still require funding in order to remediate mine and mill tailings sites. The accumulated and estimated costs of decommissioning and cleanup of uranium-producing projects in 14 countries that had produced approximately 63% of the entire cumulative world production of U by 1992 were estimated at US\$12.9 billion (WISE 2002).

The USA has the most extensively documented procedures for dealing with uranium mill tailings. Special "repositories" have been designed to contain mill tailings. They generally consist of lined storage cells with rock-armored embankments designed to last at least 1000 years. U.S. regulations strongly influence the regulatory procedures around the world. The regulations focus on reducing emanations of radon from the tailings; preventing the spread of tailings through erosion of the containment structure; reducing contamination by seepage; and setting requirements for thorough assessment of risk to the public and the environment, including the risk of groundwater pollution. Chapter 40 of the U.S. Code of Federal Regulations (CFR) stipulates (1) a design to control radiological hazards for up

to 1000 years, to the extent achievable, and in any case for at least 200 years; (2) the use of natural materials to construct the disposal cover; (3) that the ^{226}Ra concentration shall not exceed 0.185 Bq/g and 0.555 Bq/g in the top 15 cm of soil and deeper, respectively; (4) that, at all times, the radon decay product concentration (including background) in any occupied or habitable building shall not exceed 111 Bq/m³ (3 pCi L⁻¹), and the level of gamma radiation shall not exceed the background level by more than 175.5 nGy/h (Gy denotes absorbed energy in Joule kg⁻¹); and (5) that the cover shall be designed to perform under steady-state conditions, such that moisture infiltration is limited to a rate, or flux, that will not allow long-term seepage to migrate out of the disposal cell into groundwater and exceed the established groundwater standards for constituents common to uranium mill tailings at a given point of compliance. The U.S. NRC and U.S. EPA groundwater standards for tailings sites and inactive sites are as follows: combined ^{226}Ra and ^{228}Ra : 0.185 Bq L⁻¹, gross alpha-particle activity: 0.555 Bq L⁻¹, combined ^{238}U and ^{234}U : 1.11 Bq L⁻¹ (equivalent to 0.044 mg L⁻¹ for equilibrium), Ag: 0.05 mg L⁻¹, As: 0.05 mg L⁻¹, Ba: 1.0 mg L⁻¹, Cd: 0.01 mg L⁻¹, Cr: 0.05 mg L⁻¹, Hg: 0.002 mg L⁻¹, Mo: 0.1 mg L⁻¹, Pb: 0.05 mg L⁻¹, Se: 0.01 mg L⁻¹, NO₃⁻ (as N) 10 mg L⁻¹.

Under the Uranium Mill Tailings Remedial Action (UMTRA) program, the U.S. Department of Energy (DOE) has completed the surface remediation of 24 sites in 10 states and transferred them to the Long-Term Surveillance Plan (LTSP), which conducts annual inspections and groundwater monitoring at the disposal sites. Under the UMTRA groundwater compliance project, some sites, including Ambrosia Lake (New Mexico), Belfield (North Dakota), and Lowman (Idaho), need no further action, while sites such as

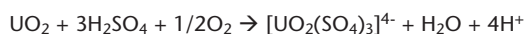
TABLE 1 NUMBER AND SIZE OF CLOSED AND OPERATING URANIUM MINES AND TAILINGS PILES WITH CURRENT STATUS INDICATED (AFTER IAEA 2004)

Continent	Country	U Mining since	Number of mines	Tailings volume ($\times 10^6 \text{ m}^3$)	Status
North America	United States	1898	3900	120	Major rehabilitation program completed 1979–1999
	Canada	1933	24	ca. 30	Old small sites mostly rehabilitated; major current producer from 3 mines and 3 mills
South America	Argentina	1950s	11	–	Mining and milling ceased; planning for rehabilitation in progress
	Brazil	1981	1	2.17	Mining and milling ended 1997; restoration pending
Australia	Australia	1930s	25	48.6	Old small sites mostly rehabilitated; major current producer from 2 mines
Asia	Kazakhstan	1955	19	209	Only in situ leach mining; hard-rock mining ceased
	Uzbekistan	–	4	30	–
	Krgystan	–	4	–	Many mines closed in early 1990s; dumps unremediated
	China, India, Japan	–	13	0.03	–
Europe	Germany	1946	9	161	Production ceased 1990; major WISMUT clean-up in progress
	Ukraine	1950s	6	130	Production continues from one mill; restoration activity, started in 1991, has paused due to economic difficulties
	Russia	1950	21	54.1	Production continues; old mines rehabilitated; rehabilitation and decommissioning plans for operating plants being reviewed
	France	1950s	180	47.3	Mining and milling stopped; old sites being progressively rehabilitated since 1990
	Czech Republic	1948	13	46.8	Reduced production matched to national energy needs; rehabilitation of closed sites underway
	Hungary, Bulgaria, Estonia, Romania, Portugal, Spain, Sweden, Slovenia, Poland, Finland	–	111	59.15	Production ceased or is very low in most countries; rehabilitation is on-going
Africa	Gabon, Namibia, Dem. Rep. of Congo, South Africa	–	43	–	–
TOTALS			4384	938	

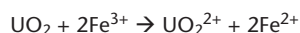
Tuba City (Arizona) or Monument Valley (Arizona) need active remediation. By the end of 1999, the total accumulated remediation cost of the UMTRA project had reached US\$1.48 billion. Active remediation will require another decade of effort.

MINERALOGY AND GEOCHEMISTRY OF URANIUM MILL TAILINGS

The mineralogy and chemistry of tailings depend on the mineralogy of the ore and on the leaching process (acid or alkaline) used for extraction. The presence of sulfide minerals in the ore may produce acid leachates, which facilitate the release of radionuclides and toxic metals from the tailings to the environment. The extraction of uranium from the ore generally involves leaching with sulfuric acid (H_2SO_4). Ore with large amounts of carbonate are preferably leached with carbonate (Na_2CO_3). The sulfuric acid leaching method is represented by the following reaction:



For in situ leach mining, UO_2 oxidation may proceed by reaction with ferric iron, which is a ubiquitous natural oxidant:



Alkaline leaching with sodium carbonate proceeds by the following reactions:



Sometimes, the addition of a synthetic oxidant (ClO_4^- , H_2O_2) is required to catalyze the oxidation of UO_2 . The effluent “raffinate” or “barren solution” and leach residue are neutralized by the addition of CaCO_3 and/or $\text{Ca}(\text{OH})_2$ and usually discharged as a slurry into a waste retention pond or above-ground facility.

Solid phases in the tailings consist of fine-grained primary and secondary minerals. Primary minerals are phases that have resisted the leaching in the mill and remain relatively unchanged (silicate minerals such as quartz, feldspar, and clay, along with remaining sulfides). Secondary minerals (e.g. Fe oxyhydroxides and Ca sulfate) form by the precipitation of ore species and reagents added during processing and neutralization. The fine fraction of the tailings, including colloids, consists mainly of clay, sulfate salts [e.g. gypsum ($\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$), barite (BaSO_4)], and oxyhydroxides of Fe, Al, Mn, and Si. The chemical composition of tailings reflects their mineralogy (TABLE 2). The tailings contain high concentrations of hazardous elements (e.g. As, Pb, V) in addition to radionuclides in the ^{238}U decay series. This paper will focus on U, Ra, Rn, Th, and As due to their toxicity.



FIGURE 2 (A) Aerial view of the open pit at the Ecarpière mine site (France) during operation. The mine was operated between 1957 and 1990. (B) After operation and remediation. Site remediation was completed within 7 years. PHOTOS COURTESY OF NUCLEAR ENERGY INSTITUTE (NEI)

TABLE 2 CHEMICAL COMPOSITION OF URANIUM MILL TAILINGS FROM DIFFERENT LOCATIONS

Analyte	Canada ¹	France ²	Utah ^{3a}	Utah ^{3b}	Slovenia ⁴
SO ₄ (µg/g)	101,000	110,522	61,100	6760	–
Ca (µg/g)	51,800	52,500	63,100	17,700	–
Al (µg/g)	34,100	69,527	–	–	–
Fe (µg/g)	19,200	27,000	18,200	7640	–
As (µg/g)	5640	120.6	74	48	–
Pb (µg/g)	749	150.5	158	72	–
V (µg/g)	205	–	517	2820	–
U (µg/g)	152	126	531	350	~81
Mo (µg/g)	91	–	35	10	–
Ba (µg/g)	77	195.4	1010	777	–
Cr (µg/g)	25	48.5	–	–	–
SiO ₂ (%)	28	52.3	–	–	–
²²⁶ Ra (Bq/g)	74	28.4	26.3	21.2	8.78
²¹⁰ Pb (Bq/g)	18	–	–	–	–
²¹⁰ Po (Bq/g)	14	–	–	–	–
²³⁰ Th (Bq/g)	19.5	–	32.3	26.1	3.93

¹ Donahue et al. (2000)

² Somot et al. (1997)

³ Morrison and Cahn (1991); (A) acid-leached ore, (B) carbonate-leached ore

⁴ Krizman et al. (1995)

Uranium

In nature uranium generally exists in either the U⁶⁺ or the U⁴⁺ oxidation state. The geochemistry of uranium under equilibrium conditions can conveniently be summarized in a pE-pH diagram (FIG. 3). Under oxidizing conditions, U⁶⁺ is present in the form of the uranyl ion UO₂²⁺, which is soluble in water. Uranium's mobility is even higher upon formation of uranyl carbonate complexes, including [UO₂(CO₃)₂]²⁻ and [UO₂(CO₃)₃]⁴⁻, under neutral to alkaline conditions. However, under reducing conditions, U⁴⁺ precipitates as a highly insoluble UO₂ phase. In tailings disposal sites, oxidizing conditions are dominant, but confinement and microbial activity may lead to localized reducing conditions. However, despite oxidizing conditions, uranyl ions still sorb onto Fe oxyhydroxides, which are major components of mill tailings (Somot et al. 1997; Lottermoser and Ashley 2005). Also, coprecipitation with carbonate and phosphate [e.g. autunite, Ca(UO₂)₂(PO₄)₂] may play a significant role in U stabilization in mill tailings disposal sites (Abdelouas et al. 1998, 2000; Ohnuki et al. 2004). Under reducing conditions, anaerobic bacteria in mill tailings sites reduce U⁶⁺ to U⁴⁺, resulting in precipitation of highly insoluble uraninite (FIG. 4) (Abdelouas et al. 2000). Typical U concentrations in tailings pore waters range from less than 0.1 mg L⁻¹ under reducing conditions to a few mg L⁻¹ under oxidizing conditions (Lottermoser and Ashley 2005). Acidification of tailings pore waters, due to oxidation of sulfide minerals initially present in the ore, may cause uranyl desorption from solids, thus maintaining high U concentrations in pore waters.

Radium-226

Tailings typically contain all the ²²⁶Ra initially present in the ore (TABLE 2). For instance, French uranium mill tailings contain between 4000 and 60,000 Bq kg⁻¹ of radium. Radium in mill tailings is generally sorbed and coprecipitated with Fe–Mn oxyhydroxides, gypsum, and barite. Also, coprecipitation with amorphous silica may play an important role in ²²⁶Ra retention (Landa 2004). Investigation of the ²²⁶Ra profile in a flooded (0.3 to 3 m water depth) mill tailings deposit cell showed low concentrations in the overlying surface water under shallow (<1.5 m water depth) oxidizing conditions due to coprecipitation with poorly crystallized barite (Martin et al. 2003). For a water depth of 2 m, reduction of sulfate by sulfate-reducing bacteria resulted in severe barite dissolution. Therefore, microbial activity indirectly contributes to ²²⁶Ra release in pore water by causing barite dissolution.

Radon-222

²²²Rn, an inert radioactive gas with a short half-life (3.8 days), has been identified as an important carcinogen (BEIR VI 1999). A recent pooled analysis of key European studies showed that the risk of lung cancer increases by 16% for each 100 Bq/m³ increase in radon concentration (WHO 2005). Radon concentrations persist in mill tailings because ²²²Rn is a decay product of the longer-lived ²²⁶Ra (half-life = 1600 years). Radon migrates through the soil mainly by pressure- and/or temperature-induced convection and by molecular diffusion in the air- and water-filled pores. Convection by water in unsaturated soil is usually negligible due to the very low water flow velocity. In stabilized tailings, ²²²Rn and other radionuclides are confined by a system of multiple layers that prevents the dispersion of the radioactivity. To prevent radon migration and water ponding in the tailings, a special cap of highly compacted silty clay soil is placed on tailings. The cover systems are designed to last for 1000 years.

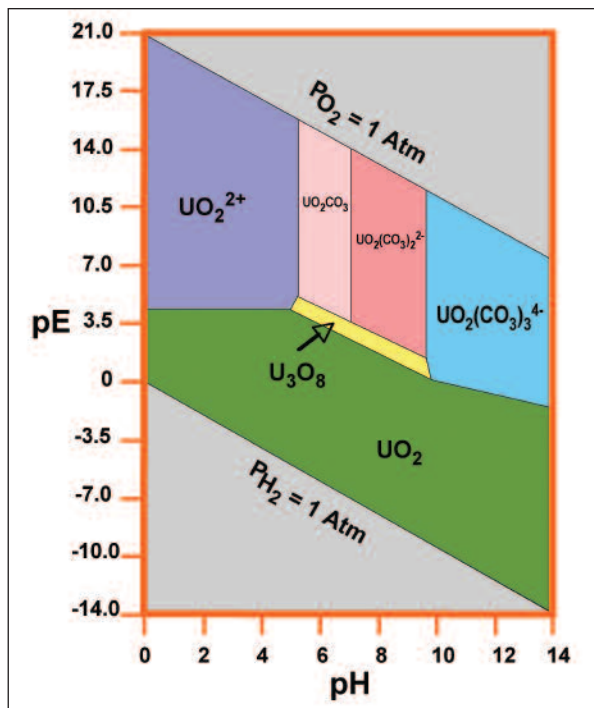


FIGURE 3 pE-pH diagram for the U-C-O-H system at 25°C. The total concentrations of uranium and carbon are 10^{-6} M and 10^{-3} M, respectively.

Thorium-230

Under slightly acidic to alkaline conditions, thorium is highly insoluble [ThO_2 , $\text{Th}(\text{OH})_4$]. However, the solubility of thorium increases in acidic aqueous solutions ($\text{pH} < 4$). Ferric hydroxides formed as a result of neutralization of leachates from U mill tailings can have high concentrations of ^{230}Th (25.1 kBq kg^{-1}) compared to U ore (8.78 kBq kg^{-1}) and mill tailings (3.93 kBq kg^{-1}) (Križman et al. 1995). In acidic tailings liquids, the ^{230}Th concentration may be very high ($1.7 \times 10^{-8} \text{ M}$), but it drops drastically at $\text{pH} \geq 4.6$ (7.2×10^{-12} to $2.4 \times 10^{-13} \text{ M}$) (Lottermoser and Ashley 2005). Acidification of tailings waters may lead to the release of Th into the environment.

Arsenic

Arsenic concentrations in uranium ore deposits may be as high as 10 wt% (Donahue and Hendry 2003). A good summary of the geochemistry and mineralogy of arsenic can be found in *Elements*, vol. 2, no. 2 (2006). Under the oxidizing conditions commonly prevalent in mill tailings, As occurs as arsenate (As^{5+}) species and less often as arsenite (As^{3+}) or organic species. After the addition of lime for pH control in the mill, typical pore waters in tailings have pH values above 7 or 8, are slightly oxidizing, and contain HAsO_4^{2-} with typical concentrations of 5–15 mg L^{-1} , which are higher than the U.S. NRC groundwater standard for tailings sites (0.05 mg L^{-1}) (Langmuir et al. 1999). Under oxidizing to slightly reducing conditions, As adsorbs onto mineral surfaces (e.g. Fe oxyhydroxides), but at high As concentrations (several mg L^{-1}), calcium arsenates precipitate. Under highly reducing conditions, As may precipitate as sulfides and arsenides. The spatial extent of a contaminant plume of As is determined by the hydrology of the surface and groundwaters, the biogeochemistry of the site, and the nature of soils surrounding the disposal site (i.e. the amount of Fe oxyhydroxides).

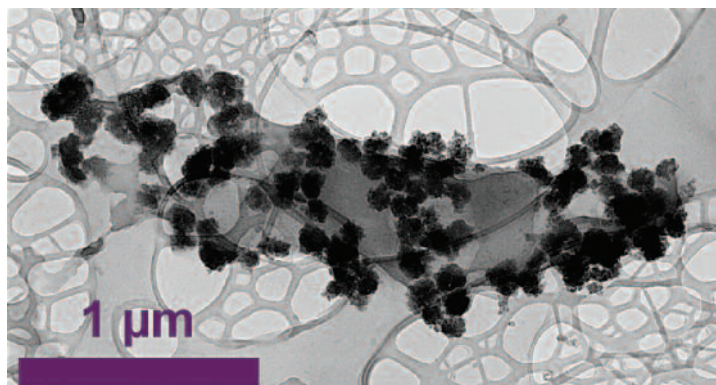


FIGURE 4 Uraninite particles precipitated on an indigenous bacterium grown in amended groundwater from the Tuba City mill tailings site, Arizona, USA. MODIFIED FROM ABDELOUAS ET AL. (2000)

The release of U, Th, Ra, Rn, and As to the environment is clearly related to the physical stability of the tailings disposal facility, including engineered covers, and the chemical stability of mineral phases controlling elemental concentrations in solution. Fe oxyhydroxides are key phases that control radionuclide and hazardous element concentrations in solution. The major concern in mill tailings from a geochemical standpoint is the oxidation of sulfides and the generation of acid pore waters. In low-pH waters, desorption of radionuclides and hazardous elements occurs, increasing their concentration. Seepage of such acid waters, in particular from unlined mill tailings, may affect the surrounding environment, including soils, surface water, and groundwater. For instance, acidity combined with deposited salts and heavy metals can prevent plant growth. In acid water leaching from tailings, dissolved uranium and daughter products can reach levels that are a direct threat to nearby ecosystems. Recently, effort has been focused on better containment of radioactivity by means of managed drainage and retention ponds.

ENVIRONMENTAL IMPACT OF URANIUM MINES AND MILL TAILINGS

Improper disposal (without liners) of mill tailings during the early decades of uranium mining has led to substantial contamination of soils, surface waters, and groundwater. Disposal sites with no effective containment of the tailings are widespread. Furthermore, some tailings, essentially coarse sands, have even been used in building materials, such as concrete (IAEA 2004). In this case, the radiation hazard originates mainly from ^{222}Rn and its daughter products.

Catastrophic or chronic failure of containment structures is a major cause of environmental impact. Chronic failures include dispersal of radioactive dust from dry tailings, erosion of tailings from outer surfaces of the containment structure, and effluent discharge. Hundreds of incidents involving active and inactive tailings dams have been reported; most of them were caused by slope instability, seepage, overtopping, and earthquakes (Strachan 2002). A breached dam wall at a U tailings facility at Church Rock, New Mexico, released 370,000 m^3 of radioactive water and 1000 tons of contaminated sediment, affecting some 110 km of the Rio Puerco (IAEA 2004).

Furthermore, contamination of groundwater with tailings wastes may occur during or after in situ leach-mining operations. Restoration of aquifers leached with acids has proven to be difficult, as pH values are difficult to restore to more neutral values. Migration of the acid leachate with dissolved radionuclides and heavy metals into areas with

poor hydraulic connections to the rest of the production zone (the zone leached for its uranium) can complicate the extraction operation. A common problem encountered in H_2SO_4 in situ leach mining of uranium deposits is the clogging of the well with precipitates, such as jarosite [$\text{KFe}_3(\text{SO}_4)_2(\text{OH})_6$] (Mudd 2001). Jarosite can retain significant amounts of contaminants (As, Pb, Hg, Cu, Zn, Cr, Se, ^{226}Ra), which may be released to the environment during aquifer flushing with previously uncontaminated groundwater.

Release of radon and interaction of mill tailings with surface water constitute a significant proportion of the health hazard. Gatzweiler (2000) estimated additional annual doses of 1–6 mSv/y, compared to the natural background dose of ~2.4 mSv/y, in the vicinity of uranium mill tailings sites in Germany. An estimation of population dose in the vicinity of six old Spanish uranium mines showed doses between 3.2 and 5.1 mSv/y, which are higher than the average national value of 2.6 mSv/y (Quindós et al. 2004). Epidemiological studies revealed a greater incidence of lung cancer than in the general population (Lopez et al. 2001). In addition, tailings have a direct impact when they are used in construction materials, such as when tailings sands are used in concrete for house construction and when houses are built over tailings-derived landfill. For example, very high concentrations of ^{222}Rn (e.g. 200,000 Bq/m³) have been measured at Schneeberg, Germany (Keller 1993). Most countries have adopted radon concentration limits of 200–400 Bq/m³ (5.4–10.8 pCi L⁻¹) for indoor air as an action or reference level above which mitigation measures should be taken to reduce Rn concentrations.

Acid drainage is a major environmental concern with uranium mill tailings. Oxidation of sulfides, which are common constituents in ore deposits, generates acidic waters. Oxidation may occur in the upper part of the tailings (Lottermoser and Ashley 2005), but also in certain cases in the bottom part due to infiltration of oxygen-rich groundwater. Sulfide oxidation is catalyzed by autotrophic bacteria such as *Acidithiobacillus ferrooxidans* and *Acidithiobacillus thiooxidans*. Low-pH waters may dissolve large quantities of secondary phases, including Fe oxyhydroxides and carbonates, containing radionuclides and heavy metals. Also, certain radionuclides and hazardous metals desorb under acidic conditions. Migration of contaminants from an unlined acid tailings repository and evaporation ponds into the local unconfined aquifer has been demonstrated by Lottermoser and Ashley (2005). Monitored bore-holes showed U concentrations up to 2.08 mg L⁻¹, low pH (≤ 4.5), and high concentrations of SO_4 (up to ~7 g L⁻¹), Cl (up to ~1 g L⁻¹), and Mn (up to ~1 g L⁻¹). U, ^{226}Ra , ^{210}Pb , As, Fe, and Mn are mobilized from the upper oxidized part of the tailings pile into surface water seepages. The spreading plume that originates from tailings dams can move at rates of several tens of meters per year (von der Heyden and New 2004). In alkaline mill tailings, migration of U into groundwater is facilitated by the formation of stable uranyl carbonate complexes [$\text{UO}_2(\text{CO}_3)_2^{2-}$ and $\text{UO}_2(\text{CO}_3)_3^{4-}$] as shown by Abdelouas et al. (1998) in groundwater near a tailings pile at an UMTRA site near Tuba City, Arizona. In addition to U, the plume contained high concentrations of nitrate (~1 g L⁻¹) and sulfate (~2 g L⁻¹). A 12 m saturated zone was contaminated by the plume, which was moving at an average rate of 15 m per year.

Geochemical and biological features of an area impacted by failure of the Aznalcóllar acidic pyrite tailings dam located west of Seville, southern Spain have been studied extensively (Hudson-Edwards et al. 2003; Olías et al. 2006). Though the tailings do not originate from uranium ore, geochemical and mineralogical features are similar to uranium mill tailings (e.g. presence of acid-generating minerals,

sulfides, and toxic elements including As, Pb, Cd, and Zn). The spill (5.5×10^6 m³ of acidic water and 1.3×10^6 m³ of heavy metal-bearing tailings) occurred on 25 April 1998, and an area of 4286 hectares along the Agrio and Guadimar river valleys was affected. After removal of most of the contaminated soil in 1999, sediments and waters were sampled (Hudson-Edwards et al. 2003). More than a year after the spill, levels of As, Cd, Pb, and Zn in the contaminated upper part of the Guadimar valley were still much higher (7 to 55 times) than the pre-spill concentrations. River waters located 2.6 km downstream of the mine still showed relatively low pH (4.4) and high sulfate concentrations (~1 g L⁻¹), likely due to oxidation of pyrite deposited in the sediment as a result of the spill. Continued monitoring of river water quality showed a recovery of water quality by 2002 (Olías et al. 2006). Arsenic concentrations in river waters remained low (~5 µg L⁻¹) under the relatively low pH conditions near the mining area due to strong sorption on and coprecipitation with Fe oxyhydroxides. The highest concentrations of As (~0.1 mg L⁻¹) were observed in the lower part of the river far from the tailings area, where As was desorbed from sediments as the pH increased.

In order to study the biological effects of the Aznalcóllar tailings spill, Bonilla-Valverde et al. (2004) used the Algerian mouse (*Mus spretus*) as a bioindicator at various sites in the Guadimar valley and in the Doñana National Park located 60 km south of the Aznalcóllar mine. Several biochemical parameters used as biomarkers responsive to metal and organic pollutants were measured in the liver, and metal concentrations in the kidney were determined. The authors concluded that the increasing response of most biomarkers, in particular antioxidative enzymes, can be ascribed to the prooxidant effects of the tailings metals (Pb, Cd, As) and suggested the progressive pollution of key Doñana ecosystems. Madejón et al. (2006) demonstrated the accumulation of As, Cd, Zn, and other metals in tubers of *Scirpus maritimus* L. from Doñana marshes, a source of food for local geese. In some areas, concentrations of Cd (0.25 mg kg⁻¹) and Zn (61 mg kg⁻¹) in tuber tissues were higher than in tubers from a control area (0.02 mg kg⁻¹ for Cd and 22 mg kg⁻¹ for Zn), while the high concentrations of As (3 mg kg⁻¹) in tubers sampled from areas not affected by the spill were attributed to past mining activities.

Fernández et al. (2005) studied the toxicity of arsenic on the microbial community in soils affected by mine tailings by measuring dehydrogenase activity (DHA). DHA is a very sensitive parameter used to evaluate the toxicity of a compound on microbiological activity; high DHA indicates high microbial activity. As the concentration of water-soluble As⁵⁺ increases, DHA (e.g. the oxidative capacity of the soil) decreases. The arsenic-bearing tailings are indeed negatively affecting the overall oxidative capacity of the soil, thus directly affecting the food chain.

Modeling efforts to assess the long-term impacts of contaminants released from uranium mill tailings, involving multiple pathways, multiple contaminants, and multiple environmental receptors, have been made by an international working group supported by the Swedish Radiation Protection Institute (Camus et al. 1999). Using a realistic scenario based on cover/cap degradation beginning at 200 years after disposal, the authors showed that release of radon and deposition of contaminated dust from the atmosphere are still important contributors to the radiation dose for human beings. Ingestion of leafy vegetables grown in soils contaminated with radioactive dust is the most important pathway for contributing to intake and dose (71–79% of total dose). The rest of the dose is attributed to dust and gas inhalation (12–20%), external exposure from soils (3–9%), and ingestion of beef (4–7%). For radioactivity released in

groundwater, the pathways contributing most to internal dose were ingestion of leafy vegetables grown with contaminated groundwater and direct ingestion of water for all contaminants except for arsenic, for which ingestion of beef was important due to its relatively high beef transfer factor.

SUMMARY

Uranium mine and mill tailings are hazardous due to radioactive and toxic elements that remain from the ore deposits. Most of these “inherited” elements are associated with Fe- and other metal-oxyhydroxides formed during the neutralization process in acid mill tailings. However, these elements may also form separate mineral phases, including autunite (uranium) and Ca arsenate (arsenic). Catastrophic or continuous release of contaminants can have a substantial impact on the environment. The mobility of radionuclides and toxic metals is enhanced by acidification of tailings due to sulfide oxidation catalyzed by microbial activity. Tailings impact ecosystems by negatively affecting the growth of the microbial community, plants, and animals. Adequate disposal of tailings—e.g. by using bottom

liners and caps on tailings designed to last for 200 to 1000 years, in compliance with strictly established regulations—efficiently reduces the release of radionuclides and hazardous elements. While surface remedial action mainly consists of stabilizing the tailings onsite, active remediation of contaminated groundwater is required for complete remediation of certain mill tailings sites (e.g. Tuba City site, Arizona). Technologies including in situ bioremediation are still being developed to improve the remediation of mill tailings sites. In all cases, groundwater monitoring is required for tens of years.

Future research should focus on the role of microorganisms on the behavior of radionuclides and metals in tailings under disposal conditions. A better understanding of the entire microbial community at tailings sites is crucial to the development of effective remediation strategies. Hence, while iron- and sulfur-oxidizing bacteria may cause radionuclide concentrations in surface and groundwater to rise through acidification, metal- and sulfate-reducing microorganisms may help precipitate radionuclides, such as uranium, via reduction of U^{6+} to U^{4+} . ■

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