

**CONTAMINANT DISTRIBUTIONS AT TYPICAL
U.S. URANIUM MILLING FACILITIES
AND THEIR EFFECT ON REMEDIAL ACTION DECISIONS**

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INTRODUCTION

Past operations at uranium processing sites throughout the United States have resulted in local contamination of soils and ground water by radionuclides, toxic metals, or both. Understanding the origin of contamination and how the constituents are distributed is a basic element for planning remedial action decisions. This paper describes the radiological and nonradiological species found in ground water at a typical U.S. uranium milling facility. The paper will provide the audience with an understanding of the vast spectrum of contaminants that must be controlled in planning solutions to the long-term management of these waste materials.

URANIUM MILL TAILINGS REMEDIAL ACTION OVERVIEW

In the period between the Manhattan Project (early 1940s) and the 1960s, uranium mining and milling in the United States expanded rapidly. While initial expansion was spurred by the defense needs of the Cold War, development of nuclear power plants in the 1950s fueled further expansion. Production peaked in the early 1960s at a little over 15,000 metric tons a year of concentrated uranium. Concentrated uranium is obtained from uranium ore through a combination of crushing, grinding,

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and extraction. During the milling process, other uranium decay products, such as thorium-230 and radium-226, remain in the waste material and account for 85 percent of the ore's original radioactivity. This waste material, commonly referred to as tailings, was left behind in large piles when the mills shut down. Little or no effort was made to provide for the long-term disposal or stabilization of the tailings when many of the mills were abandoned.

In the late 1960s, direct gamma radiation and radon gas and its decay products at the abandoned mill sites were determined to be potential health hazards. As the concern about possible cancer links to radon grew, the U.S. Congress passed the *Uranium Mill Tailings Radiation Control Act* (UMTRCA) in 1978 to authorize the U.S. Department of Energy (DOE) to remediate and to permanently dispose of the radioactive mill tailings at 24 abandoned mill sites and associated vicinity properties (VP). The mill sites are located in 10 states: Arizona, Colorado, Idaho, New Mexico, North Dakota, Oregon, Pennsylvania, Texas, Utah, and Wyoming (Figure 1). By an amendment to the UMTRCA in 1983, the DOE was also given responsibility for cleaning up VPs near Edgemont, South Dakota.

To comply with the law, the DOE established the Uranium Mill Tailings Remedial Action (UMTRA) Project Office at the Albuquerque Operations Office in Albuquerque, New Mexico. Since 1983, the UMTRA Project Office has been performing remedial action on surface contamination, which includes uranium mill tailings and abandoned mill buildings. This effort, called the UMTRA Surface Project, is responsible for controlling the exposure and dispersion of uranium mill tailings by stabilizing this material in disposal cells.

Previous uranium processing activities resulted in the formation of contaminated ground water beneath and, in some cases, downgradient of most of the 24 inactive mill sites designated for remediation under UMTRCA (Figure 2). This contaminated ground water often contains elevated levels of hazardous constituents. The UMTRA Ground Water Project was initiated to evaluate the extent of this contamination and take whatever remedial action necessary to protect human health and the environment by meeting U.S. Environmental Protection Agency (EPA) standards.

The law authorizing DOE to clean up the inactive uranium mill tailings sites also directed the EPA to establish standards to be used during remedial action. The U.S. Nuclear Regulatory Commission (NRC) was directed to provide consultation and concurrence in the type of remedial action that would be performed. Before remedial action is started, the DOE must comply with the *National Environmental Policy Act* and perform detailed studies of the environmental impacts at each site.

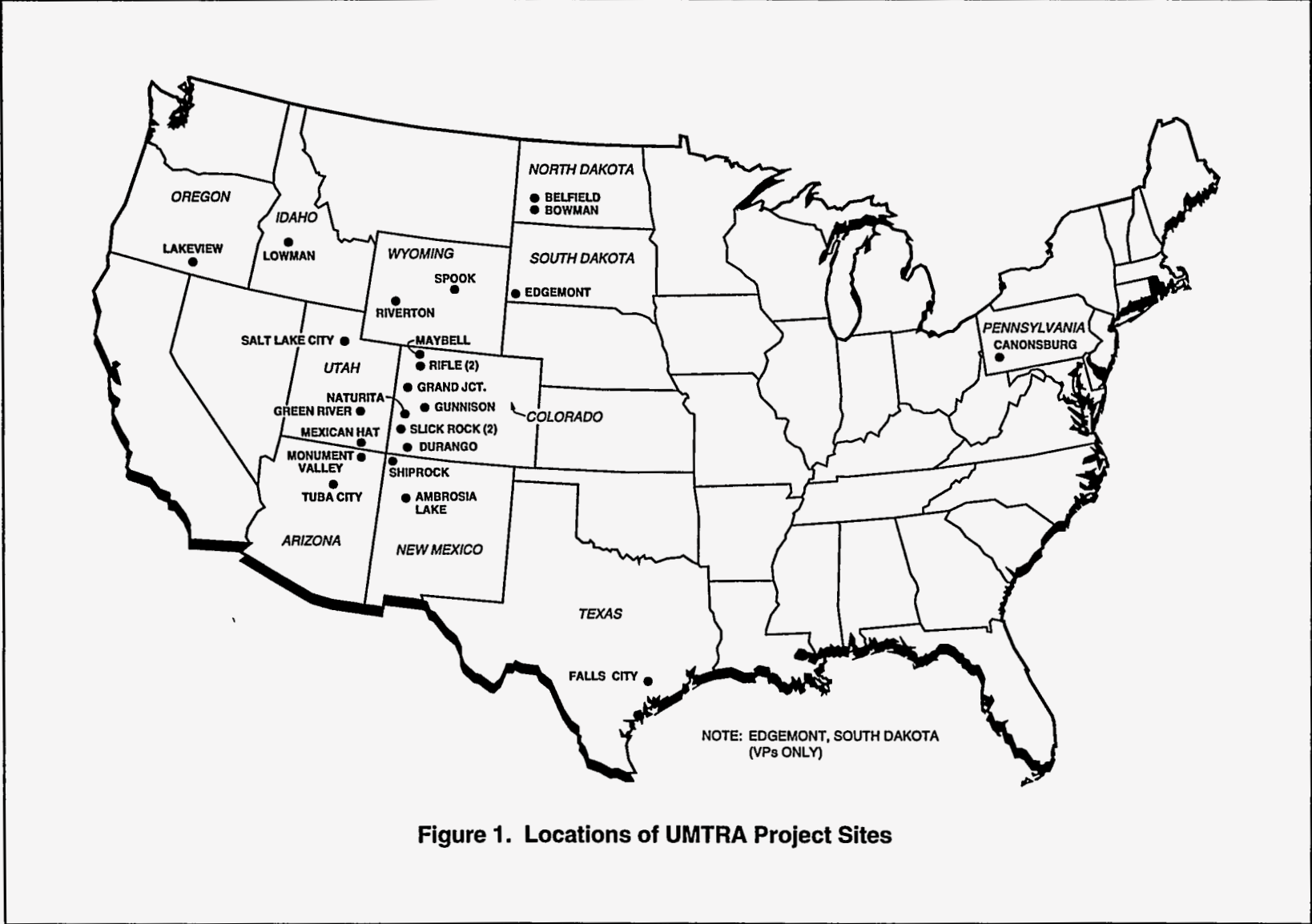


Figure 1. Locations of UMTRA Project Sites

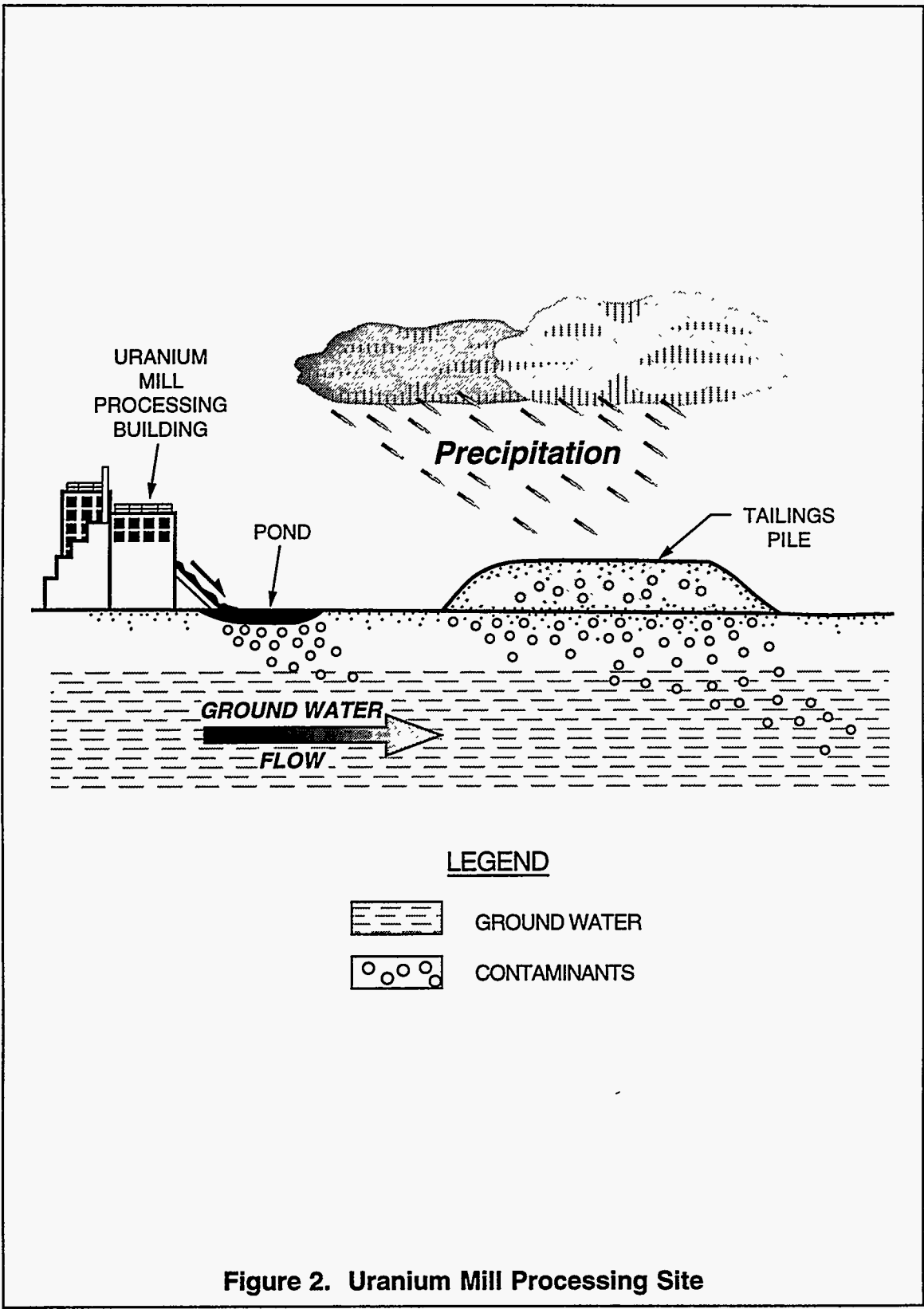


Figure 2. Uranium Mill Processing Site

MILLING PROCESSES AND PROCESS REAGENTS USED AT UMTRA PROJECT SITES

The 24 designated UMTRA Project processing sites were active for varying lengths of time from the 1940s into the 1970s. The available historical record of milling processes, quantities of chemicals used, and waste volume produced by previous operations at current UMTRA Project sites is incomplete. Ore types processed at the sites were typically vanadate sandstone deposits. The two sites in North Dakota processed uraniumiferous lignite ores. The Lakeview, Oregon, site processed arsenate ores predominantly. Information on the milling processes used was obtained from publications covering uranium milling at the time of plant operation. Information on milling processes and ore types used at the sites is summarized in Table 1.

The milling process used at each site depended on the type of ore being milled. The process frequently changed throughout the plant's history as one ore source was depleted and additional ore sources were discovered. The uranium extraction process was accomplished by either the acid or alkaline leach method. Commonly used reagents were sulfuric acid for the former technique and sodium carbonate for the latter. Acid leaching was the most effective technique and was used at most sites during their history. Alkaline leaching was used for ores with a high lime content and was used at the Ambrosia Lake, New Mexico; Durango, Colorado; Naturita, Colorado; Riverton, Wyoming; and Tuba City, Arizona, sites (Figure 1) during all or part of their history.

Solvent extraction and resin ion exchange were the two major techniques used for concentration and purification of the leach solutions. Solvent extraction involved the transfer of the dissolved uranium from the aqueous phase into an organic phase. Solvent extraction generally involved the use of alkyl phosphoric compounds or quaternary amines and long chain alcohols in an inert carrier such as kerosene or fuel oil. Typically, over 90 percent of the solvent consisted of this inert carrier (Merritt 1971). A second process was used to recover or strip the purified and concentrated uranium product into a second aqueous phase from which the uranium could be precipitated. Typical stripping agents included nitrates, chlorides, sulfates, carbonates, hydroxides, and acids. Solvent extraction was the primary concentration technique used at UMTRA Project sites.

Resin ion exchange used strong and intermediate base anionic (typically ammonium-bearing) synthetic resins. The acid or alkaline leach solutions were passed through these media, which selectively adsorbed the uranium. An eluting reagent was then used to release the uranium from the resins to produce a purified and concentrated uranium solution. Typical eluting agents included nitrate, chloride, and sulfuric acid solutions. Uranium could then be precipitated directly from this solution. Resin ion

Table 1. Ore Type and Milling Processes Used at UMTRA Project Sites

UMTRA Project Site	Ore Type	Milling Process
Ambrosia Lake, New Mexico	Limestones	Alkaline leach
Belfield, North Dakota	Lignites	Rotary kiln lignite ore-burning
Bowman, North Dakota	Lignites	Lignite ore-burning
Canonsburg, Pennsylvania	Vanadates and other ores, concentrates, and residues	N/A
Durango, Colorado	N/A	Acid leach, alkaline leach, and solvent extraction
Falls City, Texas	Sandstones	Acid leach and solvent extraction
Grand Junction, Colorado	Vanadates and oxides	Acid leach and solvent extraction
Green River, Utah	Vanadates	Upgrader unit and acid leach
Gunnison, Colorado	Oxides, silicates, and phosphates	Acid leach and solvent extraction
Lakeview, Oregon	Arsenate and phosphates	Acid leach and solvent extraction
Lowman, Idaho	Monazites	N/A
Maybell, Colorado	N/A	Upgrader unit, acid leach, and ion exchange
Mexican Hat, Utah	Sulfides	Acid leach and solvent extraction
Monument Valley, Arizona	Vanadates	Upgrader unit and acid leach
Naturita, Colorado	Vanadates	Alkaline leach and acid leach
New Rifle, Colorado	Vanadates	Acid leach and solvent extraction
Old Rifle, Colorado	Vanadates and lignite ash from the Belfield site	Water leach and acid leach
Riverton, Wyoming	N/A	Acid leach, solvent extraction, and Alkaline leach
Salt Lake City, Utah	N/A	Acid leach and solvent extraction
Shiprock, New Mexico	Vanadates	Acid leach, solvent extraction, and ion exchange
Slick Rock, Colorado ^a	Vanadates	Acid leach
Spook, Wyoming	Vanadates	Acid leach and ion exchange
Tuba City, Arizona	N/A	Acid leach, ion exchange, and alkaline leach

^aThe Slick Rock, Colorado, UMTRA Project site actually consists of two separate sites. They have been combined for this table.

N/A — not available.

exchange was used at the Maybell, Colorado; Shiprock, New Mexico; Spook, Wyoming; and Tuba City, Arizona, sites (Figure 1) during part of their history.

Precipitation of uranium from acid leach solutions to produce the final uranium product, yellowcake, was accomplished by two principal methods: direct neutralization with a base, such as lime, caustic soda, magnesia, or ammonia, and direct precipitation from acid solution with hydrogen peroxide. Precipitation of uranium from alkaline leach solutions was accomplished by one of the following methods: the addition of a strong base, acidification followed by neutralization with a base, or hydrogen reduction.

From the above discussion of milling processes used, it is obvious that a wide range of chemical wastes could be expected at UMTRA Project sites. The list of chemicals includes sulfuric acid, sulfates, carbonates, chlorides, nitrates, ammonia, lime, magnesium, manganese, and organic compounds, such as alkylphosphates, amines, alcohols, kerosene, and fuel oil. In addition, elements associated with uranium ores, such as uranium, selenium, vanadium, molybdenum, and arsenic may be dissolved in the waste stream. Approximately 0.91 to 4.55 tonnes of process waste water must be disposed of for every tonne of ore processed (Merritt 1971). Many of the milling sites processed between 273 to 910 tonnes of ore per day. Uranium milling required large amounts of water, ranging from 834 to 4172 liters of water per tonne of ore processed (Merritt 1971). A typical site processing 455 tonnes of ore per day required 2087 liters of water per tonne of ore processed; approximately 347 million liters (280 acre-feet) of water would be required at that site per year. Most of this water would end up as process waste water (raffinate) that, in most cases, would seep into the ground. This ground water pollution is a critical problem for UMTRA Project remediation.

TAILINGS SOLUTION CHEMISTRY

Generally, raffinate was the source of the solutions in the tailings because the tailings and raffinate were commonly slurried together to the tailings piles (though in some instances, excess raffinate was disposed of through evaporation ponds). Little is known about the specific types and amounts of process chemicals used at each site, and even less information is available for the composition of the raffinates. One analysis using available sources (Table 2) indicates that these solutions were generally acidic, with a pH of about 1, with total dissolved solids exceeding 120,000 milligrams per liter, and with large amounts of dissolved metals.

Because there are few records available for the composition of waste solutions produced at the UMTRA Project sites, tailings solutions have been collected (using suction lysimeters) to characterize the source of ground water contamination. The

Table 2. Analysis of Raffinate Produced at the Durango, Colorado, Mill Site^a

Parameter	Raffinate Concentration^b
Arsenic	16
Beryllium	30
Chloride	6500
Copper	23
Fluoride	12
Iron	370
Manganese	200
pH	0.8
Ra-226 (picocuries per liter)	148
Selenium	<0.01
Sodium	16,000
Sulfate	66,000
Total dissolved solids	120,000
Uranium	4 ^c
Vanadium	250
Zinc	300 ^c

^aData are in milligrams per liter except for radium-226 (picocuries per liter) and pH (standard units).

^bRaffinate analysis from Tsivoglou et al. 1960, except as noted in footnote c.

^cRaffinate uranium and zinc analyses from Tame et al. 1961.

results of analyses of tailings solutions collected are summarized in Table 3. In this table, constituents are listed in decreasing order of concentration within three broad categories: constituents having maximum concentration limits (MCL) for ground waters as defined by the EPA standards, other metals and metalloids, and other nonmetals.

Analysis to detect all major elements and hazardous inorganic constituents (Table 3) confirms their presence in these tailings fluids. The sources of several constituents are reagents used to process ores, including sulfate, ammonium, nitrate, chloride, and sodium. Other constituents are derived from dissolution of the ore-bearing rocks during the uranium extraction (acid or alkaline leach) stage of processing. In general, concentrations and frequency of occurrence of these constituents simply reflect the normal abundance (Krauskopf 1967) of such elements in common rocks. Thus, calcium, magnesium, potassium, aluminum, iron, manganese, nickel, and zinc, which are abundant in rocks, occur at high concentrations in the tailings solutions. By contrast, elements that occur at low levels in normal rocks, such as antimony, silver and mercury, occur less frequently and at low levels in tailings solutions.

Superimposed on the general trend of occurrence by normal abundance are elements concentrated in the sandstone-type uranium ores. These include uranium, molybdenum, selenium, vanadium, arsenic, and, to a lesser extent, cadmium and chromium. As a result of mineralization, these occur in the tailings solutions at levels greater than expected in average rocks. However, lead (an element associated with some uranium deposits) occurs at levels notably less than expected.

The tailings solutions are generally acid, having a median pH of 3.1. Study of tailings solutions (BEG 1992) indicates that the acidity is due to the presence of hydrolyzing salts (aluminum sulfate and ferric iron sulfate). Also, the tailings solutions are generally oxidizing. This combination of conditions keeps many trace metals in solution by preventing adsorption and precipitation.

Evaporation of raffinates and drying of the tailings result in precipitation of relatively soluble sulfate salts within evaporation ponds and tailings piles. Dissolving these salts reconstitutes an acid solution because of the presence of aluminum and iron sulfate. Thus, dried tailings provide a source of acid leachates via percolation of rainfall and snowmelt through the tailings piles long after the original raffinate has drained from the tailings piles. Also, the concentrations of leachates vary considerably within a tailings pile, depending on the amount of dilution by precipitation or concentration by evaporation. Thus, large ranges in concentrations are observed in sampled tailings solutions both from different sites (Table 3) and from the same site.

Table 3. Constituents in Tailings Solutions at Nine UMTRA Project Sites^a

Parameter	Frequency ^b	Median ^c	Range
pH	9/9	3.1	0.8 - 7.1
<i>Constituents having UMTRA Project MCLs</i>			
Nitrate	7/7	6.5	0.2 - 6100
Uranium	8/9	2.9	0.3 - 675
Arsenic	9/9	3.6	0.03 - 16
Selenium	5/9	2.05	<0.01 - 40
Molybdenum	9/9	0.52	0.14 - 23
Cadmium	7/7	0.32	0.1 - 0.7
Chromium	7/7	0.13	0.03 - 4
Lead	4/7	0.03	0.005 - 0.25
Silver	4/7	0.02	<0.01 - 0.08
Barium	0/6	<0.1	-
Mercury	6/6	0.0002	0.0001 - 0.0003
Radium - 226	3/3	148	12 - 280
Radium-228	2/3	4.2	<1 - 6
<i>Other metals</i>			
Aluminum	7/7	860	0.4 - 6300
Iron	9/9	370	0.1 - 2200
Manganese	8/8	61	1 - 360
Zinc	8/8	10	4.5 - 300
Vanadium	9/9	3	0.3 - 250
Cobalt	6/6	2.4	0.3 - 31
Nickel	7/7	1.8	0.2 - 25
Beryllium	5/5	0.16	0.01 - 30
Copper	6/6	0.13	0.03 - 4
Thallium	1/4	<0.05	<0.03 - 0.4
Antimony	5/6	0.02	<0.003 - 0.1
Tin	1/7	<0.01	<0.01 - 0.25
<i>Other constituents (nonmetals)</i>			
Sulfate	9/9	12700	1600 - 66000
Chloride	9/9	1100	27 - 6500
Sodium	9/9	570	94 - 16000
Calcium	9/9	560	420 - 670
Magnesium	7/7	390	180 - 2600
Silica	7/7	99	75 - 128
Potassium	7/7	57	6 - 220
Ammonium	6/6	58	4 - 1020
Fluoride	6/7	8.6	<0.1 - 58
Boron	7/7	1.3	0.2 - 3
Bromide	4/4	0.4	0.3 - 15
Phosphate	3/5	0.03	<0.1 - 27
^a Data are in milligrams per liter except for radium-226 and -228 (picocuries per liter) and pH (standard units). ^b Frequency indicates the number of sites where the constituent was detected followed by the number of sites where the constituent was analyzed. ^c Median and range figures comprise maximum values observed in ground water at each site.			

DISTRIBUTION AND ATTENUATION OF CONTAMINANTS IN GROUND WATER

Table 4 gives the occurrence of ground water contaminants (constituents in ground water which occur in excess of background levels or MCLs at 20 UMTRA Project sites. Three sites are not included in this analysis: Lowman, which has no process-related contamination; Ambrosia Lake, which is complicated by extensive contamination associated with adjacent properties; and Maybell, for which data summaries were not readily available. The table provides an estimate of the upper bounds for contaminant concentrations in ground water. Maximum concentrations measured at each site provide the range of values for each contaminant, given that the value was above background levels. Values at or below background levels are not reflected in the range and median. Constituents are in the same order as in Table 3.

Process-related constituents that occur at elevated concentrations in ground water include sulfate, chloride, and sodium at nearly all UMTRA Project sites. Nitrate and ammonium occur at about half these sites. These two reagents were apparently not used at all sites. Major elements derived from ores, such as calcium, magnesium, potassium, and strontium also occur at elevated concentrations at most sites. Constituents that commonly occur in ground water above background levels include ore-enriched metals and metalloids (uranium, molybdenum, selenium, vanadium, and arsenic) and commonly occurring metals (iron, manganese, zinc, and nickel). Metals that rarely occur in ground waters at these sites are lead, silver, barium, mercury, aluminum, cobalt, beryllium, and thallium.

In general, the contaminants most prevalent in ground water are those which occur in the tailings solutions at high concentrations. Significant differences between the occurrence of constituents in the tailings solutions and in the ground water provide insight into contaminant attenuation. These differences are summarized in Table 5. This table is based on a comparison of concentrations in tailings solutions to concentrations in ground water at the nine UMTRA Project sites for which tailings solution data are available. At each site, for each metal, the maximum observed concentration in ground water was expressed as a percentage of the maximum concentration in the tailings solution. The percentages of attenuation given for each constituent are the median and range for all sites. In all cases, there was a large range of values for each constituent, from less than 1 percent to more than 100 percent of the tailings solution concentrations. This range probably reflects changes in contaminant source concentrations with time due to leaching of the tailings by precipitation over several years. The median value for each constituent, however, provides a semiquantitative measure of the degree of attenuation of contaminants near the tailings-subsoil interface.

Table 4. Constituents in Ground Water Occurring Above Background Levels or MCLs at 20 UMTRA Project Sites^a

Constituent	Frequency ^b	Median ^c	Range	MCL
<i>Constituents having UMTRA Project MCLs</i>				
Nitrate	11/20	1110	45 - 5300	44
Uranium	20/20	1.2	0.014 - 70	0.044
Arsenic	11/20	0.08	0.018 - 1.3	0.05
Selenium	13/20	0.3	0.02 - 21	0.01
Molybdenum	17/20	0.4	0.004 - 1.0	0.1
Cadmium	9/20	0.06	0.1 - 0.7	0.01
Chromium	6/20	0.16	0.03 - 0.23	0.05
Lead	2/20	0.06	0.04 - 0.07	0.05
Silver	3/20	0.04	0.03 - 0.08	0.05
Barium	0/20	-	-	2
Mercury	1/20	-	0.009	0.0002
Radium - 226	7/20	17	1.3 - 654	5
Radium-228	0/20	-	-	5
<i>Other metals</i>				
Aluminum	1/20	-	796	N/A
Iron	15/20	6.5	0.1 - 460	N/A
Manganese	19/20	7.5	0.12 - 74	N/A
Zinc	15/20	0.4	0.03 - 7	N/A
Vanadium	13/20	0.4	0.03 - 14	N/A
Cobalt	3/20	0.7	0.4 - 1.2	N/A
Nickel	11/20	0.3	0.03 - 1.3	N/A
Beryllium	1/20	-	0.6	N/A
Copper	6/20	0.05	0.01 - 0.25	N/A
Thallium	2/20	0.2	0.06 - 0.4	N/A
Antimony	9/20	0.04	0.019 - 0.5	N/A
Tin	5/20	0.09	0.02 - 0.2	N/A
<i>Other constituents (nonmetals)</i>				
Sulfate	19/20	4800	210 - 15600	N/A
Chloride	17/20	970	40 - 6800	N/A
Sodium	18/20	1430	60 - 5800	N/A
Calcium	17/20	560	100 - 2100	N/A
Magnesium	16/20	450	26 - 2800	N/A
Silica	9/20	30	12 - 200	N/A
Potassium	16/20	50	4 - 240	N/A
Ammonium	11/20	270	12 - 1750	N/A
Fluoride	7/20	2.7	1 - 12	N/A
Boron	8/20	0.7	0.2 - 57	N/A
Bromide	5/20	0.5	0.3 - 19	N/A
Phosphate	1/20	0.9	-	N/A
^a Data in milligrams per liter except for radium-226 and -228 (picocuries per liter). ^b Frequency indicates the number of sites where the constituent was detected followed by the number of sites where the constituent was analyzed. ^c Median and range figures comprise maximum values observed in ground water at each site. N/A = not analyzed.				

Table 5. Attenuation of Constituents in Ground Water at Nine UMTRA Project Sites

Parameter	Number of Comparisons	Median in Percent ^a	Range in Percent ^a
<i>Constituents having UMTRA Maximum Concentration Levels</i>			
Nitrate	7	62	4 - > 100
Uranium	9	16	0.3 - > 100
Arsenic	9	12	0.1 - > 100
Selenium	5	15	1 - > 100
Molybdenum	8	98	10 - > 100
Cadmium	7	12	0.6 - > 100
Chromium	7	30	3 - > 100
Lead	4	40	20 - > 100
Silver	4	53	12 - > 100
Barium	0	-	-
Mercury	0	-	-
Radium - 226	2	5	<0.01 - 10
Radium-228	0	-	-
<i>Other metals</i>			
Aluminum	7	2	0.01 - > 100
Iron	9	30	0.1 - > 100
Manganese	8	74	0.3 - > 100
Zinc	8	1.6	0.01 - > 96
Vanadium	9	6.8	0.2 - > 100
Cobalt	6	10	0.3 - > 100
Nickel	7	16	0.4 - > 100
Beryllium	5	23	0.03 - > 100
Copper	8	0.4	0.05 - > 100
Thallium	0	-	-
Antimony	0	-	-
Tin	0	-	-
<i>Other constituents</i>			
Sulfate	9	57	12 - > 100
Chloride	9	88	17 - > 100
Sodium	9	280	22 - > 100
Calcium	7	120	98 - > 100
Magnesium	7	53	12 - > 270
Silica	7	43	17 - > 150
Potassium	7	86	29 - > 200
Ammonium	6	62	4 - > 100
Fluoride	6	21	13 - > 100
Boron	7	100	<0.01 - > 100
Bromide	4	100	38 - > 100
Phosphate	0	-	-
^a Median and range reflect the maximum level of constituents in ground water as a percentage of the maximum level of constituents in tailings solutions at each site			

One of the most important reactions resulting in attenuation of contaminant concentrations at the tailings-subsoil interface is the neutralization of the tailings seepage. The almost ubiquitous presence of carbonate minerals (generally calcite or calcium carbonate) in subsoil results in immediate neutralization of the tailings solutions beneath the tailings piles. Thus the pH, which averages 3 in tailings solutions, generally increases to an average value of 7 in ground water near the tailings piles.

The increase in pH promotes precipitation of aluminum and iron (as hydroxides and sulfate salts) and the adsorption of several of the trace metals. Constituents that tend to be strongly attenuated (to less than 10 percent of their concentration in the tailings) are radium-226, aluminum, vanadium, and zinc. Constituents that are moderately attenuated (to 10 to 50 percent of tailings solution concentrations) are uranium, arsenic, selenium, cadmium, chromium, lead, iron, cobalt, nickel, beryllium, magnesium, silica, and fluoride. The neutralization, precipitation, and adsorption of several these constituents in subsoils have been investigated at several sites (DOE 1994, DOE 1985, Markos and Bush 1982). These studies have found that, in general, constituents precipitated and sorbed from percolating tailings solutions are concentrated in subsoil within about 1 meter of the base of tailings. This 1-meter general depth of soil is commonly removed during surface remediation because of the elevated levels of radium-226 and thorium-230.

Constituents that are more mobile across the tailings-subsoil interface include nitrate, molybdenum, manganese, sulfate, chloride, potassium, ammonium, boron, and bromide. These tend to occur in the most contaminated ground waters at about the same concentration as in tailings solutions. Of these, chloride, nitrate, and sulfate are generally good indicators of ground water contamination downgradient from tailings piles due to their high concentrations relative to background and chemically conservative nature. Two components, sodium and calcium, actually increase across the tailings-subsoil interface because of reactions involving cation exchange and calcium carbonate dissolution.

CONTAMINANT ATTENUATION IN AQUIFERS

Further attenuation of contaminants occurs within the aquifers at each UMTRA Project site. In general, finer-grained aquifers containing significant amounts of sand, silt, and clay are most effective at attenuating contaminants. By contrast, gravel-bearing alluvial aquifers, which occur on floodplains at several of the sites, are least effective because of their low cation exchange and adsorption capacities resulting from lesser amounts of clay minerals and iron and manganese oxides. This effect is apparent in Table 6, where the sites are ranked according to the number of constituents that exceed MCLs. Typically, those sites which are immediately

Table 6. Relation Between Aquifer Type and Numbers of Constituents Exceeding MCLs

UMTRA Project Site	Aquifer Type	Velocity Category ^a	Number of Constituents Exceeding MCLs	Constituents Above MCLs ^b
<i>Sites with finer-grained aquifers</i>				
Monument Valley, Arizona	Sand over sandstone	High	1	NO ₃
Canonsburg, Pennsylvania	Sandy loam to silty clay loam, clay, fill material	High	1	U
Lakeview, Oregon	Silt, sand, sandy clay	Moderate	2	Mo, (As)
Salt Lake City, Utah	Sand, silt, clay	Low to moderate	2	U, Mo
Tuba City, Arizona	Sandstone	Low	3	U, Se, NO ₃
<i>Sites with alluvial gravel aquifers</i>				
Gunnison, Colorado	Sand and gravel	High	1	U
Riverton, Wyoming	Sand and gravel	High	2	U, Mo
Green River, Utah	Sand and gravel	High	3	U, (Se), NO ₃
Shiprock, New Mexico	Sand and gravel	High	3	U, Se, NO ₃
Durango, Colorado	Sand and gravel over sandstone	Moderate to high	4	U, Se, Mo, Cd
Grand Junction, Colorado	Sand and gravel	Moderate to high	4	U, Mo, As, Cd
Naturita, Colorado	Sand and gravel		4	(U), (Se), Mo, Ra-226
Lowman, Idaho	Sand and gravel and weathered granodiorite	Low		
Mexican Hat, Utah	Fractured siltstone	Moderate		
Rifle, Colorado (2 sites)	Sand and gravel	Low	5	U, Se, Mo, Cr, NO ₃
Slick Rock, Colorado (2 sites)	Sand and gravel	High	5	(U), Mo, As, Cd, NO ₃
		High	5	(U), Se, (Mo), Cd, NO ₃
<i>Sites with natural uranium mineralization</i>				
Belfield, North Dakota	Silty sandstone, claystone	Low	1	(U)
Bowman, North Dakota	Silty sandstone, claystone	Low	4	(U), (Se), (Mo), (NO ₃)
Spook, Wyoming	Sandstone	High	5	(U), (Se), (Cr), NO ₃ , Ra-226
Falls City, Texas	Sandstone	High	8	(U), (Se), Mo, As, Cd, Hg, NO ₃ , (Ra-226)
Maybell, Colorado	Sandstone	Low	7	(U), (Se), Mo, As, Cd, NO ₃
^a Ground water velocity categories: low <15 meters per year, moderate 15 to 30 meters per year, high >30 meters per year. ^b Parentheses surrounding a ground water constituent indicate ambient background contamination.				

underlain by gravel aquifers have the greatest number of constituents that exceed MCLs in ground water, while those sites which are underlain by finer-grained sediments have the fewest number.

There are exceptions to this rule. The Gunnison, Colorado, UMTRA Project site is unusual in that the aquifer is gravel-bearing but has only one contaminant (uranium) in excess of MCLs. This may be because of the unusually thick nature of the gravel aquifer (over 30 meters thick), which appears to have promoted dispersion of contaminants. At most other sites having gravel aquifers, the gravels are relatively thin (about 9 meters or less) and overlie less permeable bedrock. Some UMTRA Project sites having fine-grained aquifers (Bowman, North Dakota, and Spook, Wyoming) are in areas of natural uranium mineralization. At these sites, it is not clear if elevated concentrations in ground waters occur from process-related contamination or ambient, natural conditions. Another exception is the Mexican Hat, Utah, UMTRA Project site, where fine-grained siltstones are fractured. Ground water flow within the fractures, as in gravels, provides little opportunity for adsorption and precipitation. A final exception is the Falls City, Texas, UMTRA Project site, where the fine-grained aquifer is virtually devoid of calcium carbonate. This is an unusual condition because less than 0.1 weight percent of calcium carbonate in the subsoil is sufficient to neutralize a pore volume of tailings solution. In the absence of neutralization, acid plumes at Falls City have migrated up to 600 meters from the tailings piles. The acid conditions inhibit adsorption and precipitation of most constituents. In addition, the site is located in a uranium-mineralized area.

Two specific examples serve to demonstrate the effect of neutralization on attenuation and the contrast between the attenuation capacity of finer-grained sediments and alluvial gravels. These examples are taken from ground water quality data collected at the Falls City, Texas, and Durango, Colorado, mill tailings sites.

The low acid neutralization potential of the Deweesville Sandstone at Falls City, Texas, has allowed acidic ground water to persist downgradient of the former tailings piles. A comparison of the water quality downgradient of the tailings with the tailings solutions (Table 7) demonstrates that certain constituents can be attenuated under acid conditions. These are arsenic, chromium, molybdenum, and vanadium—elements that occur as anionic species under the relatively oxidizing conditions found at the site. (Compare the first and second columns in Table 7.) All of these species are more strongly sorbed under acid conditions relative to neutral or alkaline conditions (Rai and Zachara 1984). By contrast, metals that occur as cationic species (cobalt, copper, nickel, and zinc) and that are more strongly adsorbed under neutral to alkaline conditions tend to be mobile in the acid ground waters. With neutralization (within the same sandstone unit; see the right-hand column in Table 7), the concentrations of the cationic metals also decrease to below detection.

Table 7. Attenuation of Chemical Constituents in Acid and Neutralized Ground Water, Falls City, Texas, Uranium Processing Site^{a,b}

Constituent	Average Tailings Solution	Acidic Down-gradient Ground Water	Neutralized Down-gradient Ground Water	Primary Cause of Attenuation
pH	3.3	3.3	6.3	Neutralization by carbonate minerals
Chloride	1401	7000	1950	None
Sulfate	9114	1820	1660	Precipitation of sulfates
Alkalinity	0	0	1061	Buffered by carbonate minerals
Calcium	493	1750	1370	Buffered by carbonate minerals
Magnesium	275	270	120	Cation exchange
Potassium	188	76	19	Cation exchange
Sodium	1060	2070	730	Cation exchange
Iron	598	0.24	0.9	Precipitation sulfates and oxides
Aluminum	715	109	0.4	Precipitation of sulfates and oxides
Arsenic	2.2	<0.01	0.05	Adsorption
Chromium	0.06	<0.01	<0.05	Adsorption
Cobalt	0.8	0.6	<0.03	Adsorption
Copper	0.08	0.03	<0.01	Adsorption
Lead	<0.01	<0.005	<0.01	Adsorption
Molybdenum	1.25	<0.01	<0.01	Adsorption
Nickel	0.9	0.5	<0.04	Adsorption
Selenium	<0.02	0.1	<0.005	Adsorption
Uranium	8	6	25	Adsorption
Vanadium	1.7	0.1	<0.1	Adsorption
Zinc	4.3	1.9	<0.005	Adsorption
Radium-226	304	41	15	Adsorption/coprecipitation
^a Data in milligrams per liter except for radium-226 (picocuries per liter). ^b The symbol < indicates that concentration is below the detection limit (the number given).				

Chloride in the ground water, a conservative ion, tends to remain at or above levels found in the tailings solutions. The changes observed in the water quality are due to chemical reactions and not dilution. In fact, chloride concentrations suggest that shallow ground water at the Falls City UMTRA Project site is affected by evapotranspiration rather than dilution. Also, uranium concentrations appear to increase downgradient of the tailings. This may be from natural uranium associated with the widespread uranium mineralization at this site.

The contrast between the attenuation capacity of finer-grained sediments and alluvial gravels can be observed at Durango, Colorado UMTRA Project site. Here, ground water contaminated by raffinates has percolated down into alluvial gravels and into the underlying sandstone and shale bedrock. A comparison of the raffinate composition (identified in historic records) with contaminated ground water in basal alluvial gravel and the underlying bedrock is given in Table 8. Acid raffinates (with a pH of 0.8) seeping into the gravels were neutralized, so that the pH increased to about 7, and diluted, as indicated by decreasing chloride concentrations. Nearly all constituents were attenuated by these processes; however, the concentrations of sulfate and hazardous metals remained at high levels within the gravels. By contrast, contaminants in the bedrock (between 6 and 15 meters below the gravel) are at much lower levels. Hazardous metals are generally below detection (with the exceptions of uranium, selenium, and zinc). Sodium and sulfate concentrations are also reduced, while chloride remains about the same. The decrease in sodium and sulfate is probably from cation exchange of sodium for calcium in clays, accompanied by the precipitation of calcium sulfate (gypsum). This illustrates the natural capacity of finer-grained sediments to attenuate hazardous constituents to near-background levels over a short distance. This is due to the greater presence, in fine-grained sediments, of materials having great capacity for cation exchange and adsorption, including clay minerals, iron oxides, and manganese oxides.

BACKGROUND GROUND WATER QUALITY

Background water quality is defined as the quality of water that would exist at the site if processing had not taken place. Table 9 summarizes the levels of constituents found in background ground waters at 17 UMTRA Project sites. Background water quality varies considerably from site to site and, in several cases, background ground water contains elevated concentrations of constituents that are also associated with the source term of contamination. Sulfate is a common example. At nearly half of the sites, sulfate exceeds the EPA secondary standard for sulfate in drinking water (250 milligrams per liter). However, in most cases, the level of sulfate is still within the range acceptable for livestock (3000 milligrams per liter; NAS 1972). Eight UMTRA Project sites are in areas on or adjacent to naturally occurring uranium deposits (Maybell, Falls City, Naturita, Rifle, Slick Rock, Belfield, Bowman, and

Table 8. Attenuation of Chemical Constituents in Ground Water Beneath Raffinate Ponds at the Durango, Colorado, Uranium Processing Site^{a,b}

Constituent	Source Term (raffinate)	Shallow Alluvial Wells (n=3) ^c	Deeper Bedrock Wells (n=2) ^c	Background Bedrock Well (n=1) ^c	Primary Cause of Attenuation
pH	0.8	7.2	7.5	8.0	Neutralization by carbonate minerals
Chloride	6500	940	1100	70	Dilution and dispersion
Sulfate	66,000	12,000	2000	700	Precipitation of gypsum
Alkalinity	0	950	600	820	Buffered by carbonate minerals
Calcium	N/A	410	400	115	Buffered by carbonate minerals
Magnesium	N/A	450	290	160	Buffered by carbonate minerals
Potassium	N/A	170	25	7	Cation exchange
Sodium	16,000	4900	900	250	Cation exchange
Iron	370	0.1	0.15	0.02	Precipitation of iron hydroxides
Arsenic	16	0.02	<0.01	<0.005	Adsorption
Chromium	N/A	0.19	<0.01	<0.005	Adsorption
Cobalt	N/A	0.10	<0.01	<0.005	Adsorption
Copper	23	0.92	<0.01	<0.005	Adsorption
Lead	N/A	0.02	<0.01	<0.005	Adsorption
Molybdenum	N/A	<0.05	<0.05	<0.01	Adsorption
Nickel	N/A	0.06	<0.05	<0.01	Adsorption
Selenium	N/A	2.7	0.01	<0.03	Adsorption
Uranium	4.0	1.0	0.02	<0.005	Adsorption
Vanadium	250	0.15	<0.05	<0.005	Adsorption
Zinc	230	1.3	0.08	0.03	Adsorption
Radium-226	148	<2.0	<2.0	0.6	Adsorption/coprecipitation

^aData in milligrams per liter except for radium-226 (picocuries per liter).
^bThe symbol < indicates that concentration is below the detection limit (the number given).
^cData listed are the average of constituent levels measured at wells sampled on the same date. The number of wells included in the average is indicated by n.

N/A — not analyzed.

Table 9. Constituents in Background Ground Waters That Have Exceeded, at Least Once, Ground Water Standards^a

Constituent	Frequency ^b	Median ^c	Range ^c	Standard
<i>Constituents that have exceeded UMTRA Project MCLs^d</i>				
Selenium	7/17	0.05	0.012 - 1.1	0.01
Uranium	5/17	0.06	0.05 - 2.9	0.044
Chromium	4/17	0.08	0.06 - 0.09	0.05
Molybdenum	2/17	0.20	0.2 - 0.2	0.1
Cadmium	2/17	0.02	0.02 - 0.03	0.01
Arsenic	2/17	0.09	0.06 - 0.11	0.05
Lead	1/17	0.14	-	0.05
<i>Constituents that have exceeded EPA primary drinking water standards^e</i>				
Antimony	8/17	0.012	0.007 - 0.09	0.006
<i>Constituents that have exceeded EPA secondary drinking water standards^f</i>				
Manganese	12/17	0.8	0.15 - 4.2	0.05
Sulfate	11/17	1040	390 - 3300	250
Iron	9/17	2.4	0.08 - 12	0.05
Chloride	2/17	680	260 - 1090	250
Fluoride	1/17	4.7	-	4
<p>^aAll data are in milligrams per liter.</p> <p>^bFrequency indicates the number of sites where background ground water exceeded standards at least once followed by the number of sites where the constituent was analyzed.</p> <p>^cMedian and range apply only to sites where constituent levels in background ground water exceeded standards. Median and range are for maximum values observed at each site. Occurrences of lead and fluoride were singular.</p> <p>^dConstituent levels in background ground waters have not exceeded UMTRA Project MCLs for barium, mercury, nitrate, silver, and combined radium-226 and radium-228.</p> <p>^eConstituent levels in background ground waters have not exceeded EPA primary drinking water standards for cyanide and nickel. Beryllium and thallium could not be evaluated because of insufficient data.</p> <p>^fConstituent levels in background ground waters have not exceeded EPA secondary drinking water standards for copper or zinc.</p>				

Spook). Ground water at these sites typically contains uranium, selenium, chromium, and molybdenum. Natural ground water upgradient of the Lakeview, Oregon, UMTRA Project site is geothermal and contains naturally high levels of arsenic and molybdenum.

UMTRA PROJECT GROUND WATER REMEDIAL ACTION SELECTION

The stabilization of the surface contamination at the 24 UMTRA Project sites is almost evenly divided between stabilization on the site and disposal off the site. Most sites that have tailings relocated away from the site are either in urban settings or in river floodplains. Tailing stabilization was done to protect human health and the environment by reducing radon emanations and isolating the tailings from the environment. Stabilization of the mill tailings effectively removes the primary source of ground water contamination and is an important first step in ground water remediation.

The purpose of the UMTRA Ground Water Project is to protect human health and the environment by meeting EPA ground water standards in areas where ground water has been contaminated with constituents from former processing sites. The EPA standards establish numerical concentration limits based on MCLs or background concentrations for those contaminants expected to be associated with the uranium milling processes. Alternate concentration limits (ACL) may be established for contaminants without an established MCL or where a contaminant at a concentration above MCLs or background demonstrates no significant risk to human health or the environment. Supplemental standards may be established if such standards come as close to meeting the otherwise applicable standards as is reasonably achievable under the circumstances and protect human health and the environment. Supplemental standards are warranted under one of the following conditions: ground water at the site is of limited use in the absence of contamination from residual radioactive materials; complete restoration would cause more environmental harm than it would prevent; or complete restoration is technically impracticable from an engineering perspective.

Each UMTRA Project site's compliance strategy is directed toward satisfying the applicable EPA standards. The principal compliance strategies are: no further action through a demonstration of regulatory compliance, passive or natural flushing (allowing the natural flow of ground water to cleanse the aquifer), and active (engineered) remedial actions.

The strategy of no further action applies to UMTRA Project sites having no contamination and those sites where conditions warrant applying supplemental standards or ACLs. The strategy of passive compliance is targeted at sites where it

can be demonstrated that natural flushing of the aquifer will reduce contaminant concentrations to MCLs, ACLs, or background concentrations within 100 years, provided that human health and the environment can be protected during this period.

Active compliance strategies include engineered methods, such as gradient manipulation, contaminant isolation, ground water extraction, ground water treatment, *in situ* ground water treatment, and innovative technologies. Active compliance strategies would be used where passive compliance would not meet EPA standards, not adequately protect human health or the environment, or not be accepted by the public.

Table 10 provides the targeted ground water remedial action strategy proposed for each UMTRA Project site. No further action was selected as the targeted remedial action at 12 of the 24 UMTRA Project sites. In general, sites with high attenuation, usually with fine-grained aquifers, are good candidates for regulatory compliance. UMTRA Project sites such as Canonsburg, Lakeview, and Salt Lake City are good examples. In general, sites with natural uranium mineralization are also good candidates for regulatory compliance because of preexisting high levels of natural mineralization. For example, UMTRA Project sites such as Falls City, Spook, Maybell, Ambrosia Lake, Belfield, and Bowman are located in areas of poor background ground water quality. One UMTRA Project site (Lowman, Idaho) was selected for a strategy of no further action because no ground water contamination related to mill processing activities was identified.

Natural flushing was selected as the targeted remedial action at 10 of the UMTRA Project sites. In general, sites where natural flushing is targeted have some attenuation potential but have moderate to high ground water velocities, which have the capacity to decrease contaminant concentrations by the process of dispersion.

Active remediation was selected at two UMTRA Project sites (Monument Valley and Tuba City). Both sites are characterized as having finer-grained aquifers and greater attenuation potential, which have contributed to a low number of constituents exceeding MCLs. However, high levels of nitrate require an active approach to remedial action.

SUMMARY

Former uranium processing activities at most of the designated 24 UMTRA Project inactive mill sites resulted in the contamination of ground water beneath and, in some cases, downgradient of the sites. This paper has described the radiological and nonradiological constituents found in ground water at a typical U.S. uranium milling facility. Geochemical characterization at these sites is important in defining ground

Table 10. Targeted Ground Water Remedial Strategies at the UMTRA Project Sites

UMTRA Project Site	Aquifer Type	Velocity Category ^a	Remedial Strategy		Number of Constituents Exceeding MCLs	Constituents Above MCLs ^b
			Approach	Standards		
Falls City, Texas	Sandstone	High	AC	SS	8	(U), (Se), Mo, As, Cd, Hg, NO ₃ , (Ra-226)
Green River, Utah	Sand and gravel	High	AC	SS	3	U, (Se), NO ₃
Lakeview, Oregon	Silt, sand, and sandy clay	Moderate	AC	SS	2	Mo, (As)
Maybell, Colorado	Sandstone	Low	AC	SS	7	(U), (Se), Mo, As, Cd, NO ₃ , net gross alpha
Salt Lake City, Utah	Sand, silt, and clay	Low to moderate	AC	SS	2	U, Mo
Ambrosia Lake, New Mexico	Sandstone and shale	Low	CD	SS	7	(U), (Se), (Mo), (Cd), (NO ₃), (net gross alpha), (Ra-226)
Belfield, North Dakota	Silty sandstone, sandy claystone, and silty claystone	Low	CD	MCL/BG	1	(U)
Bowman, North Dakota	Silty sandstone, sandy claystone, and silty claystone	Low	CD	SS	4	(U), (Se), (Mo), (NO ₃)
Canonsburg, Pennsylvania	Sandy loam to silty clay loam, clay, and fill material	High	CD	ACL	1	U
Mexican Hat, Utah	Fractured siltstone	Low	CD	SS	5	U, Se, Mo, Cr, NO ₃
Spook, Wyoming	Sandstone	High	CD	SS	5	(U), (Se), (Cr), NO ₃ , Ra-226
Lowman, Idaho	Sand and gravel over weathered granodiorite	Moderate	No action	MCL/BG	0	None
Durango, Colorado	Sand and gravel over sandstone	Moderate to high	NF	MCL/BG	4	U, Se, Mo, Cd

Table 10. Proposed Ground Water Remedial Strategies at the UMTRA Sites (Concluded)

UMTRA Project Site	Aquifer Type	Velocity Category ^a	Remedial Strategy		Number of Constituents Exceeding MCLs	Constituents Above MCLs ^b
			Approach	Standards		
Grand Junction, Colorado	Sand and gravel	Moderate to high	NF	MCL/BG	4	U, Mo, As, Cd
Gunnison, Colorado	Sand and gravel	High	NF	MCL/BG	1	U
Naturita, Colorado	Sand and gravel	Low	NF	MCL/BG	4	(U), (Se), Mo, Ra-226
Rifle, Colorado (2 sites)	Sand and gravel	High	NF	MCL/BG	5	(U), Mo, As, Cd, NO ₃
Riverton, Wyoming	Sand and gravel	High	NF	MCL/BG	2	U, Mo
Shiprock, New Mexico	Sand and gravel	High	NF	MCL/BG	3	U, Se, NO ₃
Slick Rock, Colorado (2 sites)	Sand and gravel	High	NF	MCL/BG	5	(U), Se, (Mo), Cd, NO ₃
Monument Valley, Arizona	Sand over sandstone	High	active	MCL/BG	1	NO ₃
Tuba City, Arizona	Sandstone	Low	active	MCL/BG	3	U, Se, NO ₃

^aGround water velocity categories: low <15 meters per year, moderate 15 to 30 meters per year, high >30 meters per year.
^bParentheses surrounding a ground water constituent indicate ambient background contamination.

AC — additional characterization.
 NF — natural flushing.
 CD — compliance demonstration.
 SS — supplemental standards.
 MCL/BG — maximum concentration limits/background.
 ACL — alternate concentration limits.

water contaminants related to uranium processing activities and in determining contaminant interactions with the aquifer matrix. Geochemical characterization efforts are essential to developing ground water compliance strategies to meet long-term management requirements identified in the *Uranium Mill Tailings Radiation Control Act*.

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