

Chemical Constituents in Coal Combustion Product Leachate: Beryllium

Technical Report

Chemical Constituents in Coal Combustion Product Leachate: Beryllium

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PRODUCT DESCRIPTION

This report profiles the environmental behavior of the element beryllium. It includes discussions of beryllium's occurrence in soils and water, concentrations in coal and coal combustion products (CCPs), geochemistry, mobility in groundwater, toxicology, and treatment options for removal from water.

Results & Findings

Beryllium occurs naturally in soils and rocks but is rarely detected in water. The largest sources of beryllium to the environment are natural; however, it is also released by anthropogenic sources, including metals refining, stone/clay/glass manufacturing, petroleum production, and electric power generation. Typical concentrations of beryllium in U.S. soils, rocks, and coal are less than 5 mg/kg. Coal fly ash generally contains from 2 to 11 times the concentration of the feed coal. Beryllium concentrations in CCPs are usually lower than 25 mg/kg and typically range from 3 to 10 mg/kg, with the highest concentrations occurring in fly ash from subbituminous coal.

There have been few studies on the effects of beryllium on plants. Beryllium in water-soluble forms tends to be toxic to plants, but soil tends to fix beryllium in forms that makes it unavailable to plants. Beryllium is considered a potential human carcinogen, although there is no evidence that suggests negative human health effects for any pathway other than inhalation. Oral ingestion of beryllium has not been associated with any human clinical disease. While lung cancer may be associated with increased beryllium exposure from inhalation, the best data suggest that the high exposures that occurred at beryllium processing facilities in the past were responsible and that current practices greatly reduce the potential for exposure at cancer-causing levels. Beryllium has a relatively low maximum contaminant level (MCL) in drinking water of 4 μ g/L. The MCL is based on animal testing and on suggestive, although not conclusive, human epidemiology which showed that cancer was related to beryllium exposure, although not necessarily oral exposure.

Beryllium is typically not detected in CCP leachate. Leachate concentrations greater than $4 \mu g/L$ were only noted when pH was less than 4.5. Above pH 7.5, most beryllium occurs either in or on solid suspended matter. As a result, very little beryllium is found in groundwater or surface water; and transport is limited. Distribution coefficients (K_d) for beryllium under neutral pH conditions range from 100 to greater than 10,000 L/kg.

Information regarding treatment and remediation of beryllium is limited by the fact that beryllium is rarely of environmental concern. Treatment of beryllium is readily accomplished by pH control. Most beryllium solid phases are insoluble at near-neutral to alkaline pH. Beryllium is removed from solution due to its high affinity for adsorption and its strong tendency to precipitate or coprecipitate in insoluble phases. Other successful remediation techniques include cementation/vitrification and cement-based grouting.

Challenges & Objectives

Beryllium is rarely present in CCP leachate, but it is present in coal and solid coal combustion products and has been observed in acidic coal pile runoff. Because beryllium has a very low MCL, it is a potential concern from a regulatory compliance perspective; but there has been little research performed on the environmental fate and transport of beryllium. The objective of this report was to assemble and synthesize available information and present a comprehensive overview of environmental behavior data for beryllium.

Applications, Values & Use

The data presented in this report will help utilities to evaluate the risk associated with a potential release of beryllium from coal storage facilities and CCP sites. The report summarizes a wide range of information on the occurrence, health effects, groundwater transport, and treatment of beryllium. Data presented can be used to estimate leachate concentrations, to predict migration in soil and groundwater using transport models, to assess affects on potential receptors, and to narrow the field of potential treatment options.

EPRI Perspective

This is the second in a series of reports addressing key constituents at utility CCP management sites and coal storage areas. The first report was on boron (EPRI report 1005258); reports on arsenic and thallium are in process. EPRI has developed a wealth of data over the last 25 years on the leaching and groundwater transport characteristics for a wide range of inorganic constituents. This series of reports will synthesize these data along with other pertinent research results to provide a unique compendium for assessing their behavior in the environment. The reports will be updated as new information is developed by EPRI or becomes available in the published literature.

Approach

The project team conducted a literature search to identify relevant sources of information on beryllium. These sources included EPRI reports and databases, as well as other resources. The team then organized and summarized this information in one report so that key data and references can be quickly and easily accessed.

Keywords

Coal Ash Beryllium Leachate Attenuation

ABSTRACT

Beryllium occurs naturally at low concentrations in environmental media. Ambient surface and groundwater concentrations are typically lower than the maximum contaminant level (MCL) of 4 μ g/L, but may exceed the MCL under very acidic conditions. Low concentrations in water are attributable to its occurrence in solid media at low levels, and to its limited mobility. If released from solid media, beryllium's affinity for adsorption and (co)precipitation limits its mobility under most environmental conditions.

Average concentrations of beryllium in coal are similar to those in igneous rocks, shales, and United States soils (<5 mg/kg). Coal ash will generally contain from 2 to 11 times the concentration of the feed coal, and concentrations in both fly ash and flue gas desulfurization sludge are mostly less than 20 mg/kg. There is limited available data concerning the concentrations of beryllium in coal ash leachate, but existing data suggests that beryllium does not leach from coal ash under normal field conditions. Exceedingly high or low pH conditions may result in beryllium mobility, and field data have shown that beryllium is an indicator of groundwater impacts from coal storage piles that generate acidic runoff.

Beryllium resides in sparingly soluble phases in both coal and coal ash. Potential inorganic phases include chrysoberyl (BeAl₂O₄), phenakite (Be₂SiO₄), or beryllium hydroxide (Be(OH)₂). Concentrations in coal pile run-off and coal ash leachate appear to reflect the limited solubility of chrysoberyl, although a direct link has not been established. When present in solution, the most likely aqueous forms are Be²⁺, BeSO₄, or BeF⁺. Beryllium has a strong affinity for sorption, as reflected in its distribution coefficients. Beryllium sorption is pH-dependent, and reported K_d^{Be} values range from 23 L/kg at pH 1.36 to 100,000 L/kg at pH 8. Beryllium has an affinity for solids at pH 6 that is 10,000 times its affinity at pH 2.

Beryllium is considered a potential human carcinogen, although there is no evidence that suggests negative human health effects for any pathway other than inhalation. Oral ingestion of beryllium has not been associated with any human clinical disease. While lung cancer is probably associated with increased beryllium exposure from inhalation, the best data seem to indicate that the high exposures that occurred at beryllium processing facilities in the past were responsible and suggest that current practices greatly reduce the potential for exposure at cancer-causing levels.

Remediation of beryllium is readily accomplished by pH control. Most beryllium solid phases are insoluble at near-neutral to alkaline pH. Beryllium can be removed from solution by its high affinity for adsorption and its strong tendency to precipitate or coprecipitate in insoluble phases. Other successful remediation techniques include cementation/vitrification and cement-based grouting.

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1 INTRODUCTION

Coal combustion products (CCPs) include those byproducts produced from the burning of coal to generate electricity, including fly ash, bottom ash, and flue gas desulfurization gypsum. CCP production in the United States increased by 0.6% from 2003 (121.7 million tons produced) to 2004 (122.5 million tons produced). Also, the CCP utilization rate increased from 38.1% in 2003 to 40.1% in 2004 (ACAA, 2005). CCPs that are not utilized are typically stored in landfills.

The degree of utilization and storage of CCPs is directly influenced by regulatory issues. The regulatory status of metals and trace constituents in CCPs is continually shifting as new data on health and ecological effects are developed. Many electric utility companies currently monitor groundwater at CCP disposal facilities under increasingly stringent state regulations, and beneficial reuse is often limited by industrial solid waste regulations. Evolving Federal regulations under Subtitle D of the Resource Conservation and Recovery Act (RCRA) will require that most companies monitor groundwater in the 2007 to 2010 time frame.

This report describes the current understanding of beryllium occurrence and behavior in natural systems and at CCP disposal facilities. Beryllium concentrations in coal and CCPs are summarized, as well as the potential for beryllium to enter the environment through CCP storage and disposal. Additionally, environmental fate and transport and the known human health and ecological effects of beryllium are discussed.

2 OCCURRENCE, USE, AND SOURCES OF BERYLLIUM

Occurrence of Beryllium

Beryllium concentrations in various environmental media are summarized in Table 2-1. Beryllium ranks 47th in crustal abundance and averages approximately 3.1 mg/kg in the upper continental crust (Wedepohl, 1995). Concentrations in granitic rocks, shales, sandstones, and limestones typically range from <0.2 to 5 mg/kg, and clay sediments generally contain more beryllium than sands. Most beryllium occurs in silicate minerals, such as feldspar, biotite, and muscovite (Grew, 2002). Approximately 85 to 98% of crustal beryllium may be bound in feldspar minerals, and the highest concentrations are associated with cordierites and pegmatite deposits (Beus, 1966; Evenson and London, 2000; Navratil et al., 2000). United States coals contain between 0.03 and 18.00 mg/kg, averaging 2.13 mg/kg (Finkelman et al., 1994).

Environmental media		Concentrations	Reference	
Atmosphere		<0.03 ng/m ³	ASTDR (2002)	
United Sta	tes soil	0.63 mg/kg	Shacklette and Boerngen (1984)	
Water	Precipitation	0.05 to 0.08 μg/L	Meehan and Smythe (1967)	
	Groundwater	13.0 μg/L	USEPA (2000)	
	Rivers	1.9 μg/L	Eckel and Jacob (1988)	
	Estuaries	0.001 μg/L	Grew (2002)	
	Ocean	0.00005 µg/L to 0.0003 µg/L	Grew (2002)	
Rocks and minerals	Igneous	3.6 mg/kg		
	Shale	2.1 mg/kg	Horn and Adams (1966)	
	Sandstone	0.26 mg/kg	nom and Adams (1900)	
	Carbonate	0.18 mg/kg		
United States coals		0.03 to 18.0 mg/kg Average: 2.13 mg/kg St. Dev.: 1.73 mg/kg Median: 1.80 mg/kg	Finkelman et al. (1994)	

 Table 2-1

 Typical beryllium concentrations in environmental media

Occurrence, Use, and Sources of Beryllium

Average soil beryllium concentrations of 2.8 to 5.0 mg/kg have been estimated (ASTDR, 1993), an estimate that is skewed by areas with exceptionally high concentrations. Soils can be locally enriched in beryllium due to the presence of beryllium-rich rocks and minerals. For example, concentrations as high as 300 mg/kg have been reported for Alaskan soils (Sainsbury et al. 1986), and 30.5 mg/kg in Southeastern U.S. Coastal Plain and Piedmont soils (Anderson et al., 1990). Shacklette and Boerngen (1984) report a geometric mean concentration of 0.63 mg/kg for the coterminous U.S.

The average atmospheric concentration of beryllium in the U.S. is 0.03 ng/m^3 , although it is slightly higher in cities (ASTDR, 2002). Ambient concentrations have been observed to be very similar for a variety of land use categories (ASTDR, 2002).

Aqueous concentrations of beryllium decrease in the order: soil water > groundwater > low-order stream water > river water > estuary water > ocean water (Grew, 2002). Eckel and Jacob (1988) reported that beryllium was detected in 2,760 of 50,000 United States Geological Survey surface water sampling locations, with an average detected concentration of 1.9 μ g/L. Beryllium is typically detected in less than 10% of groundwaters, with average detected concentrations of approximately 13.0 μ g/L (USEPA, 2000). Estuaries generally contain 0.001 μ g/L, and ocean water concentrations can range from 0.00005 to 0.0003 μ g/L (Grew, 2002).

Uses of Beryllium

Beryllium is commonly used as beryllium metal, beryllium oxide, or as an alloy (Table 2-2). The greatest use of beryllium is in making metal alloys for nuclear reactors and the aerospace industry. As of 1998, electronic and electrical components, aerospace, and defense applications accounted for more than 80% of beryllium use. The telecommunications and computer sector was the major market for beryllium in 2003 (Cunningham, 2003).

Sources of Beryllium Release to the Environment

Beryllium enters water through many sources, mostly from weathering of rocks and soil. The United States EPA has found that the levels of beryllium in drinking water in different parts of the United States are extremely low in most cases, and that water containing beryllium at these commonly observed levels is safe to drink (ASTDR, 2002).

The atmospheric flux of beryllium is primarily derived from industrial emissions, wind-blown dust, and metallurgical processes in the space and weapons industry. Data from the USEPA Toxic Release Inventory for 2002 (available on-line at http://www.epa.gov/tri/) show that the electric utility industry accounted for approximately 84% of point source air emissions of beryllium, and 83% of surface water discharges (Table 2-3). Air emissions are estimated to account for 1.3% of the total beryllium emissions from U.S. power plants (Table 2-4). Emission factors for coal combustion range from 1 to 5.8% of total beryllium, and are lowest for coals with high ash concentrations (Bezacinsky et al. 1984; Dubansky et al. 1990).

Table	2-	2	
Uses	of	beryllium	ì

Form	Uses	Useful properties
Metal	Aircraft disc brakes, x-ray transmission windows, space vehicle optics and instruments, aircraft/satellite structures, missile guidance systems, nuclear reactor neutron reflectors, nuclear warhead triggering devices, fuel containers, precision instruments, rocket propellants, navigational systems, heat shields, mirrors, high speed computer and audio components	Strong, refractory, transparent to X-rays, resistant to corrosion, reflector of neutrons
Oxide	High technology ceramics, electronic heat sinks, electrical insulators, microwave ovens, gyroscopes, military vehicle armor, rocket nozzle crucibles, thermocouple tubing, laser structural components, substrates for high-density electrical circuits, automotive ignition systems, and radar electronic countermeasure systems	Excellent heat conductors, with a high level of hardness and strength
Copper- alloy	Springs, electrical connectors and relays, precision instruments, bushings and bearings in aircraft and heavy machinery, nonsparking tools, submarine cable housing and pivots, wheels and pinions, automobile switches, molds for injection molded plastics, radar, telecommunications, factory automation, computers, home appliances, instrumentation and control systems, tubing in oil and drilling equipment, connectors for fiber optics, integrated circuits	Electrical and thermal conductivity, high strength and hardness, good corrosion and fatigue resistance, and nonmagnetic properties

Table 2-3 Industrial beryllium releases for the U.S. in 2002

Industry	Fugitive air emissions	Point source air emissions	Surface water discharges
Petroleum	106	63	250
Stone/Clay/Glass	0	534	0
Primary metals	507	385	16
Metal mining	50	30	0
Electric utilities	116	5,351	1,303
Waste and solvent recovery (RCRA)	75	1	0
Total	855	6,366	1,574

Data from TRI (2002); available online at http://www.epa.gov/tri/ All values in pounds of Be compounds

Occurrence, Use, and Sources of Beryllium

Approximately 94% of the beryllium contained in U.S. surface impoundments is attributed to the electric utilities. Overall, electric utilities accounted for 78% of total off-site disposal and other releases, and 41% of total on- and off-site disposal and other releases. Most beryllium released by the electric utility industry is contained in landfills or other surface impoundments (Table 2-4). Non-utility releases of beryllium involve accidental overflows, leaks, spills, infiltration and seepage of copper mining products, and waste streams (Table 2-5).

Table 2-42002 Beryllium releases in the electric utility industry

On-Site Releases	Amount	% of total
Landfill (non-hazardous waste)	152,585	38.3
Emitted to air (point and fugitive emissions)	5,467	1.3
Released to surface water (point source and runoff)	1,303	0.3
Injected to groundwater (Class I well)	0	0.0
Land treatment (incorporation into soil)	785	0.2
Other surface impoundments (liquid wastes, settling ponds)	220,140	55.3
Other land disposal (waste piles, spills or leaks)	18,093	4.5
Total on-site disposal and release	398,373	100

Data from TRI (2002); available online at http://www.epa.gov/tri/ All values in pounds of Be compounds

Table 2-5

Beryllium-contaminated soil, surface water (SW), groundwater (GW), and air related to mining activities in the United States (from Taylor et al., 2003)

Site	Operation	Media	Maximum concentration	Release type
Phelps Dodge, Inc., Morenci, AZ	Copper mine	GW	16.6 µg/L	Infiltration of unlined surface impoundment
ASARCO Ray Complex, Pinal Co., AZ	Copper mine	GW SW	104 μg/L (GW) 12 μg/L (SW)	Spills, leaks, and infiltration from leachate collection ponds and electrowinning plant
DuPont Titanium Plant, New Johnsonville, TN	Titanium processing plant	GW	14 µg/L	Leakage from on-site landfill
Franklin Slag Pile, Philadelphia, PA	Copper smelting	Soil, air, and potentially SW and GW	129 mg/kg in slag, 113 mg/kg in surrounding soil	Dust from slag piles blown from site to surrounding areas

3 HEALTH AND ECOLOGICAL EFFECTS

Concentration and Absorption in Plants and Animals

Trees and shrubs usually contain small amounts of beryllium (<0.1 mg/kg). In the United States, beryllium has been detected in orchard leaves and in various trees and shrubs at concentrations of 0.026 and <0.001 mg/kg (ASTDR, 2002). In selected species of macrohydrophytes in Poland, beryllium content ranged from 0.7 to 136.6 mg/kg and was dependent upon the particular plant species and beryllium concentrations in the water (Sarosiek and Kosiba, 1993).

There seems to be little evidence for the accumulation of beryllium in fish or shellfish. In the tissue of bottom fish from Commencement Bay, Tacoma, Washington, the beryllium concentration was 6 mg/kg (Nicola et al., 1987). In oyster and clams from Lake Pontchartrain, Louisiana, beryllium ranged from 0.083 to 0.38 mg/kg of dry weight (Byrne and DeLeon, 1986). Oysters and clams from U.S. coastal waters used for shellfish production contained beryllium concentrations that were not detected up to 0.002 mg/kg wet weight (Capar and Yess, 1996). Mussels harvested from the Neosho River in Kansas contained up to 0.04 mg/kg dry weight beryllium (Allen et al., 2001).

Experimental studies with animals have demonstrated beryllium absorption through the skin, the gastrointestinal tract, and the lungs (Reeves, 1991a). The intact skin presents a relatively good barrier to beryllium. However, there is recent concern that skin exposure might cause beryllium sensitization in humans (Day et al., 2005), and skin exposures in a mouse model appeared to be associated with sensitization to beryllium (Tinkle et al., 2003). Insoluble compounds of beryllium have been shown to penetrate the skin only after trauma. Soluble beryllium compounds can cause local irritation but systemic absorption is thought to be minimal. Similarly, absorption through the gastrointestinal tract is minimal and a U.S. Public Health Service study (Hyslop et al., 1943) established that 0.006% of ingested beryllium was absorbed. The absorption of beryllium within the gastrointestinal tract occurs mainly in the acid environment of the stomach. Once the beryllium passes into the alkaline intestine, the beryllium becomes precipitated as a phosphate and is excreted in the feces (Vorwald and Reeves, 1959).

The major site for the absorption of beryllium in animals and humans is the lung (Reeves and Vorwald, 1967; Reeves, 1968). Inhaled beryllium must reach the alveolar compartment of the lung in order to be absorbed. Particles larger than 5.0 μ m in diameter will be filtered out in the upper airways and either expectorated or swallowed and excreted in the feces. Dusts of beryllium with a particle size between 0.5 and 5.0 μ m will reach the alveolar compartment and be ingested by alveolar macrophages and retained. Mists of soluble beryllium will impact on the alveolar walls and precipitate as beryllium hydroxide and beryllium phosphate and then be

Health and Ecological Effects

ingested and retained. In rats, retention in the lungs and the tracheobronchial lymph nodes can be demonstrated after inhalation of beryllium (Reeves and Vorwald, 1967; Reeves, 1968). The beryllium was cleared in two phases. Forty percent was cleared within two weeks. The remaining beryllium was cleared very slowly with a half-life of about 36 weeks. Using radioactive beryllium in rats and guinea pigs, it can be shown that inhaled beryllium will be retained not only in the lungs but also the skeleton (Zorn et al., 1977). Excretion occurs not only in the feces but also in the urine.

Effects on Plants, Animals, and Humans

Effects on Plants

There have been few studies on the effects of beryllium on plants. Beryllium in water-soluble forms tends to be toxic to plants (Romney and Childress, 1965). However, soil tends to fix beryllium in forms that make it unavailable to plants.

Oral Uptake by Animals and Humans

Oral ingestion has not been shown to cause abnormalities in humans. Rats fed beryllium can develop rickets due to the precipitation of phosphate in the gastrointestinal tract and subsequent phosphate deficiency (reviewed in Reeves (1991b)). However, this effect was not seen if the dose level was kept below 0.1% in feed. The threshold dose of beryllium for rickets in rats was estimated to be about 25 mg/kg/day. In dogs, gastrointestinal lesions and bone marrow hypoplasia were seen after three years of being fed 1-17 mg/kg/day beryllium sulfate (reviewed in USEPA (1998)).

Inhalation by Animals and Humans

In animals, inhalation can cause an acute toxic reaction (Reeves, 1991b), while in humans both an acute toxic reaction (Van Ordstrand et al., 1943), and a chronic granulomatous reaction have been described (Hardy and Tabershaw, 1946). The major toxic effect of beryllium in humans is thought to be due to the strongly acidic solutions that are the result of dissolving beryllium salts in water. These solutions will be toxic to tissues and are thought to be the cause of the acute chemical pneumonitis that results from the inhalation of high concentrations of beryllium (Van Ordstrand et al., 1943). In animal experiments, a mild acute chemical pneumonitis occurred when airborne concentrations of beryllium were above 13 μ g Be/L for 1 hr (Sendelback et al., 1986). Any cases of acute chemical tracheobronchitis or pneumonitis that have occurred since the late 1950's were likely associated with accidents (i.e., explosions) at beryllium processing plants (Eisenbud and Lisson, 1983). In contrast, the chronic granulomatous reaction to beryllium continues to occur in industry and is the major beryllium health hazard today. The granulomatous reaction to beryllium is due to a hypersensitivity reaction to beryllium (see below).

Skin Exposure of Animals and Humans

Skin exposures may result in local immune reactions (see below) or toxic or irritant reactions. In humans, five types of skin reactions to beryllium have been described (Epstein, 1991). Prolonged or unusual contact with soluble salts of beryllium can cause a chemical irritation or contact dermatitis. This is most common among workers at beryllium processing plants exposed to BeF₂. An allergic contact dermatitis may occur in up to 25% of workers exposed to BeF₂. Once this occurs, any re-exposure will result in recurrence. A chemical ulcer can be the result of lacerations or abrasions of a soluble beryllium compound. These ulcers may last several months until the crystals have been removed. An ulcerating granuloma occurs when there is a subdermal implant of a beryllium shard or crystal. Total excision is curative. Finally, delayed onset skin granulomas can occur as part of chronic beryllium disease. These reactions are immunologically based and treatment is with corticosteroids.

Mutagenic and Carcinogenic Effects

Beryllium is mutagenic in various cell systems (reviewed in Reeves (1991c)) and has been shown to cause bone tumors and lung tumors in laboratory animals (reviewed in Reeves (1991b)). In humans, a greater excess mortality from lung cancer was noted for individuals who had acute beryllium disease than those who had chronic beryllium disease [standardized mortality ratio (SMR) = 2.32 vs. 1.57, respectively] in a review of cases reported to the U.S. beryllium case registry (Steenland and Ward, 1991). A review of the studies supporting an association between beryllium exposure and lung cancer and the critique of these studies has been published (USEPA, 1998). A major criticism of these studies was their failure to control for cigarette smoking. In a recent study in which an attempt was made to correct for the effect of smoking, an excess incidence of lung cancer was still observed for workers in a beryllium processing plant (Sanderson et al., 2001). An increased risk of lung cancer was mainly observed when higher exposures were lagged 10 or 20 years. Thus, there does appear to be an increased risk of lung cancer from beryllium exposure. However, the risk appears to be low with a SMR <2.0 and associated with exposures that no longer occur in industry.

Immunologic Effects

Beryllium appears to have a unique place in immunological reactions in that it is the smallest element known that can be involved in an immune response. Curtis (1951) demonstrated that beryllium could be sensitizing to humans. Within 48 hr of application of an aqueous solution containing 2% $BeSO_4$ or BeF_2 to the skin (i.e., patch or skin test), a localized reaction of erythema, swelling, and induration occurred (Curtis, 1951). This reaction was noted to occur in beryllium extraction workers with a history of dermatitis and in beryllium workers with chronic pulmonary beryllium disease. While this reaction was negative in controls, in a later study, Curtis was able to sensitize 8 of 16 controls (Curtis, 1959). Additionally, the safety of skin testing has been questioned because there have been reports of an acceleration of pulmonary disease after skin testing (Sneddon, 1955).

Health and Ecological Effects

After in-vitro tests were developed for cell-mediated immune responses, Hanifin was the first to apply these tests to beryllium (Hanifin et al., 1970). When the peripheral blood lymphocytes from patients with chronic beryllium disease were cultured in the presence of beryllium oxide or beryllium sulfate, the lymphocytes would undergo a blastogenic transformation (i.e., they would grow and proliferate). The importance of macrophages was noted because if macrophages were first incubated with beryllium, washed, and then added to lymphocytes, the lymphocytes still underwent proliferation. Another way to measure the ability of lymphocytes to respond immunologically to beryllium is to measure the production of lymphokines (i.e., soluble factors released by lymphocytes after contact with an antigen). One such factor is macrophage migration inhibition factor (MIF). The production of MIF was only observed from the blood lymphocytes of patients with chronic beryllium disease and not from controls (Henderson et al., 1972). Thus, one in-vivo (patch testing) and two in-vitro tests demonstrated that a specific immune response could occur to beryllium.

In the 1980's, further studies expanded our knowledge of the immune response elicited by beryllium. Beryllium appeared to evoke a cell-mediated immune response and not a humoral or antibody-mediated immune response. Only a single report has documented the presence of IgG antibodies to beryllium (Clarke, 1991). However, it is not certain as to whether beryllium can cause the production of these antibodies or whether these antibodies were generated by contact with some other antigen and are really only cross-reacting to beryllium. There appears to be no relation of the presence of these antibodies and any clinical syndrome.

There is no question that a cell-mediated immune response to beryllium is an important immune response to beryllium and is the major pathogenic mechanism for chronic beryllium disease. The evidence for this was gathered from observations on cells collected from the lungs of normal volunteers and from patients with chronic beryllium disease. When beryllium lymphocyte proliferation tests were applied to lung lymphocytes from patients with chronic beryllium disease, all patients had cells that responded to beryllium (Rossman et al., 1988). Thus, an immune response to beryllium could be demonstrated in every patient with chronic beryllium disease. In addition, a greater number of cells that were reactive to beryllium could be obtained from the lung than from the peripheral blood. This demonstrated not only that patients had an immune response to beryllium but also that the cells were accumulating at the site of disease activity. Lung cells that are reactive to beryllium have not been observed from normal volunteers or from patients with a similar disease but of unknown etiology (i.e., sarcoidosis). Finally, lymphocytes that are reactive to beryllium have never been shown to be reactive with any other metal (Marx and Burrell, 1973; Saltini et al., 1989).

Further studies on the cell-mediated immune response have determined that mainly a subset of lymphocytes (i.e., CD4+ T cells) respond to beryllium by a proliferative response (Saltini et al., 1989; Milovanova et al., 2004). This response can be blocked with antibodies to HLA Class II molecules. This is an important observation because in a classic immune response, CD4+ T cells must be presented antigenic peptides by HLA Class II molecules by a cell known as an antigenpresenting cell. HLA Class II molecules are proteins that bind immunogenic peptides that are degraded from antigens digested in the endosomal/lysosomal compartment of the cells. The HLA Class II molecules with bound peptides migrate to the surface of the cells and present the peptide for recognition by CD4+ lymphocytes. Genetic epidemiological studies associated

beryllium disease with specific HLA Class II molecules, i.e., those containing glu-69 of HLA DPB1 (Richeldi et al., 1993; Wang et al., 1999; Rossman et al., 2002; Maier et al., 2003). Invitro studies have confirmed the necessity of glu-69 containing HLA DPB1 molecules for the successful stimulation of beryllium sensitive T cell lines and clones (Lombardi et al., 2001; Fontenot et al., 2000). In addition, antibodies to HLA DP could block the in-vitro beryllium response (Lombardi et al., 2001; Fontenot et al., 2000; Amicosante et al., 2001).

In contrast to acute beryllium disease, chronic beryllium disease still occurs but is due to an immune response to beryllium that occurs in a minority of exposed workers (Eisenbud and Lisson, 1983). Therefore, in this condition the determination of an immune response to beryllium has more important clinical utility than the determination of beryllium levels (DOE, 1999).

Hazard Evaluation and Limiting Concentrations

In 1949, the Atomic Energy Commission adopted limits for beryllium exposure in the workplace of 25 μ g/m³ in air as a peak value and 2 μ g/m³ averaged over an 8-hour day. These standards have essentially eliminated acute beryllium disease and the last reported case in the U.S. occurred in the 1960's (Eisenbud and Lisson, 1983). However, chronic beryllium disease has continued to occur and the Department of Energy has adopted 0.2 μ g/m³ as an action level (DOE, 1999). Whether this new lower standard will eliminate this disease is not yet known. Unfortunately, because of the long latency from initial exposure until the development of clinical disease (i.e., greater than 10 years), it will take at least 10 years and the evaluation of only newly hired workers to determine whether or not this new standard will eliminate the disease. Because the beryllium lymphocyte proliferation test can detect beryllium sensitization before the onset of clinical symptoms, use of this test may be helpful in evaluating the safety of the new standard. For all workers and former workers who had exposures above 0.2 μ g/m³, a small but significant lifetime risk exists (~ 3%) for the development of chronic beryllium disease.

Risks for the development of chronic beryllium disease have also existed for family members of workers. This is thought to be from beryllium brought home on work clothes. In addition, individuals living in communities surrounding a beryllium processing plant may be exposed due to excessive plant emissions. While current plant emissions in the United States are considered safe, cases of community acquired chronic beryllium disease continue to occur from exposures that have occurred in the past (Milton Rossman, personal communication).

Skin exposure to beryllium has been associated with disease in the past (Epstein, 1991). Whether skin exposure today is a cause of beryllium sensitization is uncertain (Tinkle et al., 2003; Henneberger et al., 2001). Nevertheless, prudence would dictate precautions to eliminate the potential for skin exposures to workers.

Oral ingestion of beryllium has not been associated with any human clinical disease. While lung cancer is probably associated with increased beryllium exposures, the best data seems to indicate that the high exposures that occurred in the past were responsible and suggest that current practices greatly reduce the potential for exposure at cancer-causing levels.

Integrated Risk Information System (IRIS) Data

The Integrated Risk Information System (IRIS) is a U.S. EPA database that contains summaries of potential adverse health effects that may result from chronic exposure to chemical substances found in the environment. Available information includes reference doses (RfDs) for noncancer human health effects resulting from oral exposure, reference concentrations (RfCs) for noncancer health effects resulting from inhalation exposure, cancer weight-of-evidence (WOE) designations, and cancer slope factors (CSFs) and inhalation unit risks (IURs) for the carcinogenic effects of chemicals resulting from ingestion and inhalation, respectively.

In 2000, EPA began a screening-level review of the scientific literature for chemicals in the IRIS database, including beryllium. The current status of beryllium-related IRIS information and the results of the recent screening-level review (USEPA, 2001) are discussed below.

Oral Reference Dose (RfD): 0.002 mg/kg-day

The IRIS RfD was derived based on a dietary study in dogs published in 1976. Since its derivation in 1998, no new literature has been identified that would be directly useful in revising the oral RfD (USEPA, 2001).

Inhalation Reference Concentration (RfC): 0.00002 mg/m³

The RfC was derived in 1998, based on an occupational study of humans and a community exposure study. The recent literature search has revealed no new evidence that would be directly useful for deriving an RfC (USEPA, 2001).

Oral Slope Factor (CSF): NA

No CSF value was assigned for beryllium and its compounds due to insufficient data as of 1998. At this time, there is still insufficient data to derive an accurate CSF value (USEPA, 2001).

Inhalation Unit Risk (IUR): $0.0024 (mg/m^3)^{-1}$

The current IUR was derived in 1987 based on a 1980 occupational study. A more recent publication has been produced by the National Institute of Occupational Safety and Health that may result in a change to the current IRIS IUR value. The change has not been reported to date (USEPA, 2001).

Cancer Weight-of-Evidence (WOE) Classification: B1

Beryllium is listed as a probable human carcinogen (B1). There has been no indication in the literature that a change to the current WOE classification is necessary (USEPA, 2001).

Environmental Regulations

Maximum Contaminant Level (MCL) for Beryllium

Beryllium has been assigned the second lowest MCL, 4 μ g/L, for inorganic contaminants, second only to mercury and thallium at 2 μ g/L. A potential link between oral exposure to beryllium and human cancer has been assumed based on the following (USEPA, 1980):

- High frequency of osteosarcomas in rabbits induced by intravenous beryllium (Cloudman et al., 1949) and of reticulum cell sarcomas in rats fed beryllium (Morgareidge et al., 1975);
- Positive results from mutagenic assays [beryllium chloride has been shown to increase the error frequency of nucleotide base incorporation into DNA in an *in vitro* assay designed to detect potential metal mutagens (Sirover and Loeb, 1976)]; and
- Suggestive, but not conclusive, human epidemiology which showed that cancer was related to beryllium exposure, but not necessarily oral exposure.

At the time of the ambient water quality determination, no studies had established an incontrovertible link between beryllium oral exposure and human cancer (USEPA, 1980). The study of Morgareidge et al. (1975) could not be used for a cancer-based criterion because: 1) the results were never officially published, and 2) the dose-response curve was atypical in that the lowest dose produced the greatest responses (USEPA, 1980). USEPA (1980) reported that the assumption that beryllium is carcinogenic after oral administration is questionable because the results of oral studies designed to test this assumption are either negative or uncertain.

However, due to the potential that beryllium could be a human carcinogen, USEPA (1980) recommended a criteria of 0 mg/L beryllium in water for maximum protection of human health. Due to the likelihood that 0 mg/L is unattainable, data from dietary studies conducted by Schroeder and Mitchener (1975 a,b) were used to estimate a criterion associated with a lifetime cancer risk of 10^{-5} (the probability of one additional case of cancer for every 100,000 people exposed). USEPA (1980) recognized that this particular study showed no significant differences in tumor frequencies between rats and mice that were exposed to 5 mg/L beryllium in water for a lifetime and those that were not exposed (Schroeder and Mitchener (1975 a,b). Regardless, this criterion was calculated to be 0.037 µg/L, and was the final recommended criterion of USEPA (1980). The current MCL concentration 4 µg/L is considered the lowest achievable beryllium concentration by use of modern waste water treatment technology.

Generic Soil Screening Levels (SSLs)

The generic soil screening level guidance (USEPA, 1996) was developed to help standardize and accelerate evaluation and cleanup of soils at sites on the National Priorities List with future residential land use. The SSLs were created as conservative screening tools to identify soils that may require further investigation, but not necessarily cleanup. They are risk-based concentrations derived from equations that combine exposure information assumptions with EPA toxicity data and address the following exposure pathways:

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- Direct ingestion
- Inhalation of volatiles or fugitive dusts
- Ingestion of contaminated groundwater caused by migration of chemicals through soil to an underlying potable aquifer
- Dermal absorption
- Ingestion of homegrown produce that has been contaminated via plant uptake, and
- Migration of volatiles into basements

Generic SSLs for beryllium have been calculated for residential and commercial/industrial scenarios (Table 3-1). The lowest SSLs for beryllium are associated with the migration to groundwater pathway. The SSL for this scenario represents the soil concentration above which soil leachate will exceed the maximum contaminant level in drinking water. The value for this pathway is largely dependent upon the dilution attenuation factor (DAF) that is assumed for the site. The DAF is defined as the ratio of soil leachate concentration to receptor point concentration. The lowest possible DAF is 1, which assumes that there is no dilution by clean groundwater or attenuation of the contaminant between the soil from which it is leached and the receptor point. The SSL derivation only addresses dilution as an attenuation process, and a default DAF of 20 is generally considered protective for contaminated soil sources up to, and potentially larger than, 0.5 acre in size (USEPA, 1996). However, it is suggested that DAF values be calculated for each site, based upon procedures described in USEPA (1996).

Scenario	Ingestion / Inhalation of		Inhalation of fugitive	Migration to groundwater	
	German	volatiles	particulates	DAF = 1	DAF = 20
Residential	160	NA	1,400	3	63
Commercial/ industrial outdoor worker	2,300	NA	2,600	3	63
Commercial/ industrial indoor worker	4,100	NA	NA	3	63

Table 3-1Generic beryllium SSLs for residential and commercial/industrial scenarios

Values in mg/kg. NA = SSL not available at this time Source: USEPA (1996)

Ecological Soil Screening Levels

Ecological Soil Screening Levels (Eco-SSLs) are concentrations of contaminants in soil that are protective of ecological receptors that commonly come into contact with soil or ingest biota that live in or on soil. They are derived separately for plants, soil invertebrates, birds, and mammals. They are conservative estimates that are designed to be used at the screening level stage of an ecological risk assessment, and they are not intended to be applied as cleanup levels. However, they are presumed to provide adequate protection of terrestrial ecosystems (USEPA, 2005a).

So far, there is insufficient data to derive Eco-SSLs for plants and avian wildlife. However, Eco-SSLs have been derived for soil invertebrates and mammals (Table 3-2). The Eco-SSL value for mammalian wildlife has been updated since its derivation in 2003. A recently published update shows that the mammalian wildlife Eco-SSL has been lowered from 36 mg/kg to 21 mg/kg. The 2005 update was based on revised exposure factors and bioaccumulation models used in Eco-SSL calculations. The changes relate to the food ingestion rates, soil intake values, and equations for estimating the uptake of metals from soil into plants, soil invertebrates, and mammals. The changes are described in detail in USEPA (2005b).

Table 3-2Ecological soil screening levels for beryllium

Date	Plants	Avian wildlife	Soil invertebrates	Mammalian wildlife
2003	NA	40 mg/kg	NA	36 mg/kg
2005	NA	40 mg/kg	NA	21 mg/kg

NA = Not Available Source: USEPA (2005a)

4 GEOCHEMISTRY, FATE, AND TRANSPORT

Basic Physical and Chemical Properties

Beryllium is an alkaline earth metal with two oxidation states (2+ and 0). It has the highest charge-to-radius ratio of all elements, and its chemical behavior is more similar to aluminum than to other Group II A members. It exists mostly as a divalent cation with eight isotopes, including one stable isotope (⁹Be) and three radiogenic isotopes (⁷Be, ⁸Be, and ¹⁰Be) (Table 4-1).

Physical Properties		Isotopes	
Property	Value	Isotope	Half Life
Atomic No.	4	⁶ Ве	5.0 x 10 ⁻²¹ seconds
Atomic Wt.	9.012182(3)	⁷ Be	53.28 days
Configuration	[He]2s ²	⁸ Ве	~7 x 10 ⁻¹⁷ seconds
Density, 20°C	1.848 g/cm ³	°Ве	stable
Melting Point	1287°C	¹⁰ Be	1.52 x 10 ⁶ years
Boiling Point	2471°C	¹¹ Be	13.8 seconds
Valence	2	¹² Be	24 milliseconds
Ionic Radius	0.31 Å	¹⁴ Be	4.3 milliseconds

Table 4-1Beryllium isotopes and selected physical properties

Source: CRC Handbook of Chemistry and Physics, 79th Ed. (1998-1999); table modified after Grew (2002).

Beryllium's small size and high charge-to-radius ratio result in compound formation with smaller anions such as fluoride and oxide which can be covalent in character (Krejci and Scheel, 1966). Many beryllium compounds will react to form beryllium hydroxide in water at neutral pH (Table 4-2). Others, such as beryllium fluoride compounds, can remain soluble at neutral pH. However, it forms mostly insoluble hydroxides and complexes between pH 5 and 8, making

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most beryllium compounds relatively insoluble from pH 4 to 9, the pH range of most natural waters (Langmuir, 1997).

Compound	Formula	Reaction in a neutral pH environment	Comments
Beryllium fluoride	BeF₂	BeF ₂ + 2H ₂ O → [BeF ₂ (H ₂ O) ₂ aq] and other complexes	Remains soluble in neutral environments
Beryllium hydroxide	Be(OH) ₂	$Be(OH)_2 \rightarrow no reaction$	Insoluble in neutral environments
Beryllium oxide	BeO	BeO + H₂O → Be(OH)₂	Forms insoluble Be(OH) ₂ in neutral environment
Beryllium nitrate (trihydrate)	Be(NO ₃) ₂ 3H ₂ O	$\begin{array}{c} Be(NO_3)_2 3H_2O + 2MOH \rightarrow \\ Be(OH)_2 + 2[M]aq + 2NO_3 aq + \\ 3H_2O \end{array}$	Forms insoluble Be(OH) ₂ in neutral environment
Beryllium sulfate (tetrahydrate)	BeSO₄4H₂O	$\begin{array}{c} Be(SO_4) \cdot 4H_2O + 2MOH \rightarrow \\ Be(OH)_2 + 2[M]aq + SO_4^{-2} \cdot aq + \\ 4H_2O \end{array}$	Forms insoluble Be(OH) ₂ in neutral environment
Ammonium tetrafluoroberyllate (ammonium beryllium fluoride)	(NH ₄) ₂ BeF ₄	(NH₄)2BeF₄ → 2[NH₄]⁺aq + [BeF₄]²aq	Remains soluble in a neutral environment
Beryllium basic acetate	$Be_4O(C_2H_3O_2)_6$	Be ₄ O(C ₂ H ₃ O ₂) ₆ + 6MOH + H ₂ O → 4Be(OH) ₂ + 2[M] ⁺ aq + 6[C ₂ H ₃ O ₂] ⁻ aq	Forms insoluble Be(OH)₂ in neutral environment

Table 4-2 Beryllium compounds and hydrolysis reactions (after ASTDR, 2002)

Solid Phases

Beryllium is an essential element in approximately 110 mineral species. The most abundant beryllium-containing minerals are silicates, followed by phosphates, oxides and hydroxides, and borates (Grew, 2002). Among solid phases of beryllium with reported thermodynamic data, the most stable are phenakite (BeSiO₄), chrysoberyl (BeAl₂O₄), and beryllium hydroxide [β -Be(OH)₂] (Table 4-3) (EPRI, 1984). Currently, there is little available data regarding what solid phases may control beryllium mobility in the environment, but it is speculated that phenakite, chrysoberyl, and the hydroxide are responsible for generally low aqueous environmental concentrations. EPRI (1984) noted that concentrations in surface waters are similar to concentrations predicted by chrysoberyl or beryllium hydroxide.

The hydroxide form of beryllium exhibits amphoteric behavior, forming positive ions in acidic solutions and negatively charged ions in basic solutions (Grew, 2002). Its solubility in pure water resembles that of amorphous aluminum hydroxide (Figure 4-1). The equilibrium solubilities of chrysoberyl, phenakite, and amorphous beryllium hydroxide were calculated using PHREEQC-2 (Figure 4-2), with supplemental thermodynamic data from Parker et al. (1971). Chrysoberyl solubility was calculated at equilibrium saturation with Al(OH)₃(am) to control aluminum concentrations, and phenakite with quartz saturation to control dissolved silica concentrations. Each phase exhibits minimum solubility in the neutral to circum-neutral pH range. Chrysoberyl exhibits the lowest calculated solubility of the three phases.

Reaction	Log K°	Reference
$Be(OH)_2$ (a) = $Be^{2+} + 2OH^2$	-21.0	Smith and Martell (1976)
$\alpha - \operatorname{Be}(\operatorname{OH})_2 = \operatorname{Be}^{2+} + 2\operatorname{OH}^{-}$	-21.14	Parker et al. (1971)
β - Be(OH) ₂ = Be ²⁺ + 2OH ⁻	-21.59	Parker et al. (1971)
$Be_{3}(PO_{4})_{2} = 3Be^{2+} + 2PO_{4}^{3-}$	-37.7	Sillen and Martell (1964)
$BeNH_4PO_4 = Be^{2+} + NH_4^+ + PO_4^{3-}$	-19.7	Sillen and Martell (1964)
$BeAl_2O_4 + 8H^+ = Be^{2+} + 2Al^{3+} + 4H_2O$	22.44	Parker et al. (1971)
$BeO + 2H^+ = Be^{2+} + H_2O$	6.41	Parker et al. (1971)
α -BeSO ₄ = Be ²⁺ + SO ₄ ²⁻	5.34	Parker et al. (1971)
$BeSO_4 4H_2O = Be^{2+} + SO_4^{2-} + 4H_2O$	0.08	Parker et al. (1971)
$BeSO_4^2H_2O = Be^{2+} + SO_4^{2-} + 2H_2O$	-1.33	Parker et al. (1971)
$Be_2SiO_4 + 4H^+ = 2Be^{2+} + H_4SiO_4^{0}$	6.05	Parker et al. (1971)
$BeO'3Al_2O_3 + 20H^+ = Be^{2+} + 6Al^{3+} + 10H_2O$	64.11	Parker et al. (1971)
$\alpha - BeF_2 = Be^{2+} + 2F^{-}$	-6.36	Parker et al. (1971)
$\alpha - \operatorname{BeCl}_{2} = \operatorname{Be}^{2+} + 2\operatorname{Cl}^{2}$	34.45	Parker et al. (1971)
$\beta - \operatorname{BeCl}_{2} = \operatorname{Be}^{2+} + 2\operatorname{Cl}^{2}$	33.87	Parker et al. (1971)

Table 4-3 Equilibrium stability constants for various beryllium reactions at 25°C and 1 atm pressure

Source: EPRI (1984)



Figure 4-1 Solubility of amorphous beryllium hydroxide in pure water at 25°C and 1 atm pressure (calculated with PHREEQC)



Figure 4-2

Equilibrium solubilities of amorphous beryllium hydroxide, phenakite, and chrysoberyl at 25°C and 1 atm pressure [calculated with PHREEQC with thermodynamic data from EPRI (1984)]
Aqueous Speciation

Inorganic Complexes

The most commonly observed beryllium complexes occur with fluoride, sulfate, chloride, and bicarbonate. The most important species of beryllium in waters with low total dissolved solids (soft waters) are Be-F and Be-OH complexes. Be-F complex formation is favored between pH 4.2 and 5.0, and may dominate up to pH 6.0 in waters with high concentrations of F^- . At pH less than 4, beryllium is present mostly as uncomplexed Be²⁺.

High aluminum concentrations can suppress the formation of Be-F complexes. Navratil et al. (2002) found that the most important factors affecting beryllium speciation in the Lesni Potok catchment (Czech Republic) surface waters were concentrations of Al^{3+} , F⁻, and pH. However, sulfate complexation can also affect beryllium mobility. At the Lysina catchment in the Czech Republic, Kram et al. (1998) found that inflow water with high sulfate concentrations enhanced beryllium mobility from the mineral soil. Beryllium concentrations in the Lysina catchment stream water were positively correlated with both fluoride and sulfate.

Organic Complexes

Organic complexes may also affect beryllium mobility. Binding to fulvic acids can occur above pH 6. Esteves de Silva et al. (1996) reported stability constants of beryllium with fulvic acids at pH 6 to 7 of log K = 5.9 to 6.5, respectively. Stability constants decrease with decreasing pH below pH 6 and decrease slightly from pH 6 to pH 7. With decreasing pH, the relative abundance of fulvic acid complexes to all dissolved beryllium complexes decreases. Humic material has been shown to increase beryllium solubility in solutions in contact with kaolinite and amorphous silica (Takahashi et al., 1999).

Predicting Inorganic Aqueous Beryllium Speciation

Equilibrium speciation can be calculated with geochemical equilibrium speciation programs. The most comprehensive thermodynamic database containing beryllium data is the updated MINTEQA2 database (USEPA, 1999). This database has been incorporated into the newest versions of MINTEQA2, MINEQL+, and PHREEQC-2. As an example, the equilibrium speciation of beryllium in a typical ash porewater solution can be calculated at various pHs using porewater data and assuming a beryllium concentration of $0.3 \,\mu$ g/L, the detection limit of EPA method 6010 B (Table 4-4 and Figure 4-3).

Table 4-4
Median coal combustion ash porewater composition

Solution	SO ₄ ²⁻ (mg/L)	Al³⁺ (mg/L)	PO ₄ ³⁻ (mg/L)	F ⁻ (mg/L)	Cl ⁻ (mg/L)	NO ₃ ⁻ (mg/L)
Ash porewater	791	0.07	0.25	1.0	11.01	0.4

Source: EPRI (1998)

From these calculations, it appears that beryllium speciation in an average ash porewater is similar to that of a pure water solution, with the exception of contributions from $BeSO_4^{0}(aq)$ at low pH and BeF^+ at neutral pH. Complexation with chloride, phosphate, and nitrate are not quantitatively important (<1% of total species).



Figure 4-3

Dissolved beryllium speciation in average ash porewater solution at 25°C and 1 atm pressure (calculated with PHREEQC)

Adsorption and Desorption

Mechanisms

Adsorption and desorption processes exert a major impact on beryllium mobility in soil and leaching to groundwater and surface water. In most environmental porous media, the most important sorbent solids are clay minerals, organic matter, and oxyhydroxides of iron and manganese. For a given weight of sorbent solid, metal adsorption will be proportional to surface area and surface site density. The greatest surface site densities and cation exchange capacities are exhibited by organic matter and the oxyhydroxides (USEPA, 2004b).

Adsorption onto oxyhydroxides can be modeled by exchange with surface functional groups, a process called surface complexation. The fundamental surface functional group, OH, can be protonated or deprotonated with decreasing or increasing pH. These sorbents have surface charges that are largely dependent on pH, attributable to the successive dominance of surface SOH_2^+ , SOH, and SO⁻ species (Langmuir, 1997). The general sorption reaction of divalent cations can be written:

 $SOH + M^{2+} = SOM^+ + H^+$

Where: S = solid surface such as hydrous ferric oxide (HFO) and $M^{+2} = a$ divalent metal cation.

Complexation of beryllium onto HFO is represented by the following equations and equilibrium constants:

 $SOH + M^{2+} = SOM^+ + H^+$

Where: S = solid surface such as hydrous ferric oxide (HFO) and $M^{2+} = a$ divalent metal cation.

Complexation of beryllium onto HFO is represented by the following equations and equilibrium constants:

$$HFO_sOH + Be^{2+} = HFO_sOBe^{+} + H^{+} \qquad \log K_1 = 5.7$$

$$HFO_wOH + Be^{2+} = HFO_wOBe^+ + H \qquad \log K_2 = 3.3$$

Where: HFO_sOH = HFO strong sorbing site and HFO_wOH = HFO weak sorbing site.

Based on data published by Dzombak and Morel (1990), HFO will preferentially adsorb metal cations in the order:

$$Hg^{2+}\!\!>\!\!Be^{2+}\!\!>\!\!Ba^{2+}\!\!>\!\!Cr^{3+}\!\!>\!\!Pb^{2+}\!\!>\!\!Cu^{2+}\!\!>\!\!Cd^{2+}\!\!\sim\!\!Zn^{2+}\!\!>\!\!Ni^{2+}\!.$$

Geochemistry, Fate, and Transport

This suggests that only mercury has a stronger affinity for HFO than beryllium with respect to cationic metals.

Influence of pH

The most important factor influencing the adsorption of beryllium and other divalent cations on hydrous oxides is pH. Figure 4-4 shows the pH-dependent sorption of cations onto HFO, and Figure 4-5 illustrates the same modeled adsorption behavior for beryllium.



Figure 4-4 pH-dependent sorption of cations on hydrous ferric oxide with metal concentrations set at 10⁻⁷ M and reactive sorption sites at 10⁻⁴ M (from USEPA, 2004b; after Stumm, 1992)

Removal of all cations from solution approaches 100% as pH increases, and beryllium removal is complete near pH 4.5. Adsorption affinity increases sharply between pH 3 and 4.5. In general, increasing soil pH leads to a rapid increase in the net negative surface charge of sorbing surfaces (Naidu et al., 1998). This results in an enhanced affinity for metal ions such as beryllium. Beryllium has an increased affinity for solids at pH 6 that is 10,000 times its affinity at pH 2 (Grew, 2002).





Influence of Other lons

Complexation of beryllium with other ligands such as fluoride or sulfate can impact its affinity for sorption. The strong bonds formed between beryllium and fluoride can inhibit the ability of potential sorbents to remove beryllium from solution. Metal complexes with fluoride, sulfate, and carbonate are generally poorly sorbed (USEPA, 2004b).

The role of organic ligands has been reported with contrasting implications. Davis and Leckie (1978) reported that the presence of certain adsorbed organic ligands on hydrous oxide surfaces may enhance uptake of beryllium. Takahashi et al. (1999) reported that humic acid increased the solubility of beryllium significantly. They compared the adsorption of beryllium on kaolinite and amorphous silica in the presence and absence of humic materials. In the absence of humic materials, beryllium adsorbed strongly to kaolinite and silica. No soluble beryllium was observed above pH 5.2 for silica and 5.5 for kaolinite. However, in the presence of humic materials, adsorption decreased by 70% in the kaolinite system and 30% in the silica system. The role of organic matter in beryllium mobility deserves more focused attention.

Environmental Fate and Transport

Disposal of coal ash, incinerator ash, and industrial wastes may increase the amount of beryllium in soil. It has been estimated that, because beryllium tends to exist in an insoluble form, beryllium compounds may remain in the soil for long periods of time (thousands of years) without leaching into the groundwater. Under most environmental conditions, beryllium compounds are highly insoluble and mobilization in soil is limited (ASTDR, 2002). Additionally, beryllium is very strongly adsorbed by finely dispersed sedimentary materials, such as clays, iron hydroxides, and organic material (Izmerov, 1985). Above pH 7.5, most beryllium occurs either in or on solid suspended matter. Because of these factors, very little beryllium is found in groundwater or surface water (USEPA, 2003), and transport is limited.

Transport Mechanisms

The primary processes governing environmental fate and transport of beryllium in the subsurface are advection, dispersion, matrix diffusion, and retardation. Advection and dispersion are dependent upon the system in question rather than the properties of beryllium. Matrix diffusion is dependent on the properties of beryllium, but is mostly omitted from transport modeling studies. Retardation is determined by a number of factors, including sorption, ion exchange, precipitation, colloid formation, biofixation, and organic matter interactions. Other important processes include changes in pH, oxidation-reduction potential, salinity, concentrations of competing ions, the nature of sorbent phases and their surface areas, and surface site densities (USEPA, 2004b).

One approach toward quantifying the various processes that contribute to the retardation of mobility of a particular contaminant relative to water is referred to as the retardation factor, $R_{\rm f}$.

$$R_f = \frac{V_p}{V_c}$$

where R_f = retardation factor, V_p = velocity of water through a control volume, and V_c = velocity of contaminant through a control volume (USEPA, 2004b).

The partitioning of beryllium between solid and water phases is characterized by its distribution coefficient, K_d^{Be} . $K_d^{Be}=C_S/C_L$, where C_S is the concentration of beryllium on solids (measured in mg/kg) and C_L is the concentration of beryllium in water (measured in mg/L). The value of K_d is a function of pH, charge density of the solid surface, mineralogy, equilibration time, concentration of the solid in the mixture, and composition and ionic strength of the solution (Grew, 2002).

The retardation factor is related to the distribution coefficient by the equation (USEPA, 2004b):

$$R_f = 1 + (\rho_b / \eta_e) K_d$$

where ρ_b = porous media bulk density and n_e = effective porosity at saturation.

For 1-dimensional advection-dispersion flow with chemical retardation, the transport equation incorporates the retardation factor as follows:

$$\frac{\partial C_i}{\partial t} = \frac{\left[D_x \frac{\partial^2 C_i}{\partial x^2_i} - v_x \frac{\partial C_i}{\partial x}\right]}{R_f(i)}$$

Distribution Coefficient (K_a)

The value of K_d^{Be} generally increases by a factor of 10⁴ from pH 2 to 6, and increases loglinearly with increasing pH above approximately pH 6 (Figure 4-6). This means that beryllium has an affinity for solids at pH 6 that is 10,000 times its affinity at pH 2. Above pH 6, beryllium mobility has been shown to be at least partially a function of Be(OH)₂ solubility (Aldahan et al., 1999).

Reported values of K_d^{Be} are listed in Table 4-5. Hawley et al. (1986) found that increasing particle density resulted in decreasing K_d^{Be} values. You et al. (1989) reported that K_d^{Be} for river mud increased with increasing temperature (log $K_d^{Be} = 6.26$ at 60°C and 5.49 at 25°C). Olsen at al. (1986) concluded that the amount of iron on particles correlates with the K_d^{Be} value.



Figure 4-6 K_d^{Be} values calculated at various pHs (USEPA, 1996)

Log K ^{Be} d (L/kg)	Reference	Comments
4.2 to 5.3	Olsen et al. (1986)	River sediments
4.08 to 5.97	Vesely et al. (2001)	River sediments
5.3 to 5.93	Vogler et al. (1996)	Lake sediments
1.36 (pH = 4.9)		
2.89 (pH = 6.8)	U.S. EPA (1996)	Calculated with MINTEQA2
5 (pH = 8.0)		
4.64 to 6.08	You et al. (1989)	Suspended solids concentration of 200 mg/L
4.48 to 6.3	Hawley et al. (1986)	Suspended solid concentrations of 0.6 to 2 mg/L
2.02	Hawley et al. (1986)	Lake Michigan sediments
5.5 to 5.9	Hawley et al. (1986)	Seawater

Table 4-5 Log K_d^{Be} values reported in the literature

5 BERYLLIUM LEACHING AT CCP AND COAL STORAGE SITES

This section summarizes data compiled by EPRI and others on beryllium occurrence in coal, CCP solids, and CCP leachate. Beryllium rarely leaches from coal or coal ash except under highly acidic conditions. Evaluation of leachate data from coal storage piles and CCP facilities reveals that beryllium leaching is controlled by the solution pH in contact with the solid material, and that the pH at which beryllium leaches from coal ash is rarely encountered in field settings.

Beryllium Concentrations in Coal and CCPs

Total solid composition data are presented in box-and-whisker plots. Figure 5-1 provides an example box-whisker plot with explanation for the symbols.



Figure 5-1 Example box-and-whisker plot

Coal

The average concentration of beryllium in U.S. coal is approximately 2.13 mg/kg, although there is some variation with coal grade (Figure 5-2). Bituminous coals tend to have higher beryllium contents than other coal grades.



Figure 5-2 United States coal beryllium concentrations (Finkelman et al., 1994)

With the exception of Bulgaria, coal beryllium concentrations are relatively uniform throughout the world, with most reported average concentrations not exceeding 7.0 mg/kg (Table 5-1). Bulgarian coal has reported average beryllium concentrations as high as 32.6 mg/kg (Kortenski and Sortirov, 2003). Kortenski and Sortirov (2003) proposed that high beryllium concentrations in Bulgarian coal are most likely caused by beryllium-enrichment in rocks surrounding depositional basins.

Source Area	Concentration (mg/kg)	Reference	Notes	
	0.03 to 18.0	Bragg et al. (1998)		
United States	Average: 2.13	Database	N = 7,107	
Western Venezuela	0.01 to 3.24	Hackley et al. (2005)	-	
Argentina	2.5 and 3.6	Brooks and Willett (2004)	Rio Turbio and Pico Quemado mines, respectively	
Canada	0.59 to 1.20	Goodarzi (2002)	Found associated with fine silicate minerals embedded in organic fraction	
Turkov	0.15 to 6.3	Palmor at al. (2004)	Liquito	
тикеу	Average.: 1.3 ± 0.97	Paimer et al. (2004)	Lignite	
China	1.3 to 3.1	Xu et al. (2003)	-	
Britain	Arithmetic mean: 1.5	Xu et al. (2003)	N=23	
Australia	Arithmetic mean: 1.5	Xu et al. (2003)	N=452; bituminous	
Dulaaria	0.7 to 6.9	Eskenazy and Valceva (2003)	Lignite; predominant inorganic association	
Duigana	2.0 to 32.6	Kortenski and Sotirov (2003)	High coal beryllium results from local basin lithology	

 Table 5-1

 Beryllium concentrations in coals from around the world

Ash

The concentration of beryllium is commonly either enriched in fly ash and depleted in bottom ash, or it is equally distributed between bottom and fly ash (Ratafia-Brown, 1994). Data from the CBEAS database (EPRI, 2003) suggest that the highest concentrations in CCPs are found in bituminous fly ash (Figure 5-3).

Data from Affolter et al. (1998) and Brownfield et al. (1999) also suggest that beryllium is most concentrated in fly ash relative to bottom ash (Table 5-2). In their studies, beryllium concentrations in fly ash represented an approximate 10-fold enrichment relative to feed coal concentrations.

Beryllium Leaching at CCP and Coal Storage Sites



Figure 5-3 Beryllium concentrations in coal ash

Fable 5-2	
Beryllium concentrations in coal and coal ash from the Illinois and Appalachian basin	S

Unit	Whole coal (mg/kg)	Fly ash (mg/kg)	Bottom ash (mg/kg)
High sulfur (~3 wt. %)	1.5 (± 0.55)	19 (± 6.7)	14 (± 5)
Low sulfur (<1 wt. %)	2.4 (± 0.35)	Coarse fraction: 22 (\pm 4.1) Fine fraction: 27 (\pm 4.3)	16 (± 2.5)

Data from Affolter (1998) and Brownfield et al. (1999)

Observations of Kortenski and Sotirov (2003) for Bulgarian coal and coal ash indicate that coal ash beryllium concentrations depend on feed coal concentration (Table 5-3). Beryllium was enriched by a factor of 2 to 11 in coal ash relative to whole coal. Using enrichment factors similar to those reported by Kortenski and Sortirov (2003) as a guide, average United States coal ash would contain between 4.26 mg/kg and 23.43 mg/kg. Fly ash data from EPRI's CBEAS database supports this estimate (Table 5-4).

			1	
Basin	Coal (mg/kg)	Coal ash (mg/kg)	Enrichment	Number of samples
Maritza-West	2.0	11.1	5.55	38
Beli Breg	0.4	2.0	5.00	38
Stanyantzi	1.3	7.7	5.92	39
Sofia	4.3	15.0	3.48	59
Karlovo	11.0	54.7	4.97	23
Samokov	2.9	8.4	2.89	31
Gabrovitza	32.6	86.6	2.65	40
Kyustendil	11.5	37.9	3.29	39
Oranovo	0.3	3.3	11.00	61
Katrishte	1.5	5.1	3.4	22
Pernik	1.8	7.6	4.22	35
Suhostrel	1.2	4.1	3.41	17
Balkan	1.6	5.8	6.23	58
Svoge	2.9	7.0	2.41	90

Table 5-3	
Average beryllium concentrations in coal and coal ash from various Bulgarian basi	ins

Source: Kortenski and Sortirov (2003)

Table 5-4 Beryllium concentrations in fly ash and bottom ash (in EPRI CBEAS database)

Туре	Average	Minimum	Maximum	Number analyzed
Fly ash	4.62	0.33	23.64	41
Bottom ash	2.98	0.89	5.20	26

Beryllium Leaching at CCP and Coal Storage Sites

Flue Gas Desulfurization Sludge

Beryllium concentrations in FGD sludge are shown in Figure 5-4, as reported in the CBEAS database. Concentrations range from 0.6 to 20.7 mg/kg, and average 6.14 mg/kg. Reported concentrations of beryllium in FGD sludge are similar to those in coal ash (Figure 5-5).







Figure 5-5 Solid composition data from ash and FGD solids (from EPRI CBEAS database)

Chemical Forms of Beryllium in Coal and CCPs

The chemical form of beryllium in coal and CCPs will have a large influence on its tendency to dissolve and be subsequently transported in solution. Many studies have investigated the chemical form of beryllium in coal, and most have concluded that beryllium is associated with the organic portion of coal. Some, however, suggest an inorganic affinity of beryllium in coal, while others have proposed an intermediate affinity (Xu et al., 2003).

The mineral forms of some trace elements in coal and coal ash were reported by EPRI (1999a). Beryllium was found to be associated with organics, oxides, silicates, and sulfides (Table 5-5). The authors reported that forms of beryllium in the silicate fraction may include beryllium aluminum silicate, beryllium aluminate, and beryllium oxides. However, they also noted that this fraction (determined by leaching with HF) could possibly represent beryllium that is associated with organic matter.

Beryllium Leaching at CCP and Coal Storage Sites

Mada of Occurrence	Coal Fly Ash		Sh	sh Bottom Ash		
Mode of Occurrence	Bit	Sub	Bit	Sub	Bit	Sub
Organic	0	0	0	0	0	0
Oxides	50	20	0	0	0	0
Silicates	25	70	100	100	100	100
Sulfides	25	10	0	0	0	0

Table 5-5Mode of occurrence of beryllium in coal and coal ash

Bit = Bituminous, Sub = Subbituminous Data reported as % of total Be content in sample Source: EPRI (1999a)

Palmer et al. (2002) performed sequential chemical extractions on coal samples from eastern Kentucky. They found 50% of beryllium to be extractable by hydrofluoric acid (HF soluble), indicating an inorganic association with silicate minerals. The unleached fraction indicated an organic association. In another study, data from Palmer et al. (1998) suggested that 20-65% of beryllium in a subbituminous coal was organically-associated. On the other hand, Querol and Huerta (1998) found nearly all of beryllium in the same coal analyzed by Palmer et al. (1998) to be organically-associated based upon flotation experiments. Palmer et al. (2002) concluded that the reported association of beryllium in coal samples varies with selective extraction technique, and that the exact mode of occurrence of beryllium in coal and coal ash is unresolved.

Beryllium Leaching from Coal and CCPs

Ash Leachate Experiments: Laboratory and Field Conditions

Existing data suggests that beryllium does not leach appreciably from CCP solids. Three large-scale leaching studies were reviewed in which beryllium was included as a constituent:

- EPRI (1995) Twenty-nine results are reported for TCLP and ASTM leaching tests performed on FGD solids. Only 3 of 29 results were detectable concentrations (detection level not reported), each resulting from ASTM leaching.
- EPRI (1998) Fifty-four results are reported for TCLP and SPLP leaching tests performed on a variety of CCP samples. Seventeen of 27 TCLP results were above the detection limit, and only 3 of 27 SPLP results contained detectable beryllium. Detection levels were not reported, but results indicated that TCLP leaching is more likely to leach beryllium from coal ash than SPLP.
- EPRI (2006) By far, the most representative cross-section of field data available comes from a recently completed EPRI study in which field leachate samples were collected from well-documented and well-described CCP management sites. Eighty-one field leachate

samples were collected from 29 sites, and only 5 samples contained detectable beryllium concentrations (Table 5-6). The concentrations of those five samples ranged from 0.8 to $8.6 \mu g/L$. Of the five detectable concentrations, four came from two fly ash impoundments where pH ranged from 4.3 to 6.0; these four samples were the only samples in the study with a pH lower than 6.5. The other sample was from a FGD impoundment with a pH of 6.8, and where water is recirculated (zero-discharge plant) such that total dissolved solids were very high, greater than 40,000 mg/L. The results of this investigation are consistent with the solubility and sorption data presented in Section 4, which suggest that beryllium has low solubility at neutral pH and is likely to be sorbed to the solid phase under neutral and alkaline pH.

ССР Туре	Management Method	Source Coal	Be Conc. (μg/L)	pH (SU)	Notes
Ash	IMP	Bit	8.6	4.26	
Ash	IMP	Bit	5.2	4.35	
Ash	IMP	Bit	1.6	5.79	
Ash	IMP	Bit	0.8	5.97	
FGD	IMP	Lig	1.5	6.8	Recirculated water

Table 5-6Detected beryllium concentrations in field leachate samples

84 field leachate samples were collected: the five samples listed here were the only detects, the remaining 79 samples had concentrations below detection limits (<0.8 μ g/L)

Source: EPRI (2006)

Case Study: West Virginia Power Plant Groundwater Quality

EPRI (1999b) investigated groundwater quality at West Virginia power plants, and found that beryllium was an indicator parameter (along with nickel and cadmium) of groundwater impacts near coal storage piles. Concentrations in groundwater and runoff were highest near coal pile storage areas, ranging from less than detection to 80 μ g/L. Out of 453 groundwater samples taken during the study, 42 (9%) contained beryllium at concentrations higher than the MCL of 4 μ g/L. Of these 42, 32 occurred in groundwater downgradient of coal pile storage areas and 10 occurred in groundwater downgradient of wastewater ponds.

Figure 5-6 shows beryllium concentrations from coal pile runoff (31 samples), ash disposal leachate (36 samples), and wastewater treatment ponds (51 samples). Only three of 36 ash disposal site leachate samples had beryllium concentrations above the MCL, and these were limited to samples with very low pH. Concentrations exceeding the MCL were only observed at pH less than approximately 4.



Figure 5-6

Beryllium concentrations as a function of pH found in EPRI (1999b) West Virginia power plant study

Potential Leaching Mechanisms

Analyses of the chemical form of beryllium in coal and coal ash suggest that beryllium is probably retained in silicate, aluminate, oxide, or organic phases that are sparingly soluble. There is limited thermodynamic data available for beryllium solid phases, but available data suggests that phenakite (Be₂SiO₄), chrysoberyl (BeAl₂O₄), and beryllium hydroxide [β -Be(OH)₂] are the most stable beryllium-containing solid phases (EPRI, 1984).

Field data collected from the Field Leaching and West Virginia power plant studies provide additional insight as to what mechanisms may limit beryllium release from CCP solids. The most important factor that seems to limit beryllium solubility at CCP facilities is the pH range encountered in the majority of CCP environments (Figure 5-6). Figure 5-7 shows field data from

the West Virginia power plants plotted with calculated solubility of phenakite, chrysoberyl, and beryllium hydroxide. Data suggest that the amount of beryllium released to solution from coal and coal ash storage areas at the West Virginia power plants could be controlled by chrysoberyl dissolution, or by a more stable phase for which thermodynamic data are not available.



Figure 5-7 Field data from EPRI (1999b) West Virginia power plant study plotted with selected beryllium mineral solubilities

Another explanation for the pH-dependency of beryllium concentrations is that beryllium release may be limited by adsorption. The increase in beryllium concentrations observed in the EPRI (1999b) study is in the pH range of the calculated adsorption edge for beryllium (pH 3-5). To test the hypothesis that adsorption controls beryllium concentrations, a modeling simulation was performed with PHREEQC using adsorption data contained in the MINTEQ thermodynamic database. The previous chapter illustrated beryllium adsorption onto HFO at a total beryllium concentration of 10⁻⁷ M, and showed that 100% removal from solution was predicted to occur at approximately pH 5.0 to 5.5. Beryllium was completely in solution (0% bound) at pH 2.

Beryllium Leaching at CCP and Coal Storage Sites

The maximum beryllium concentration observed in the EPRI (1999b) study was 0.024 mg/L at pH 2.5. For modeling purposes, this concentration was designated as the maximum total concentration because it was assumed that all beryllium exists in solution at pH 2.5. Figure 5-8 shows the predicted dissolved concentration by adsorption plotted with field data at two concentrations of reactive sorption sites, 10^{-3} M and 10^{-5} M.



Figure 5-8

Field data from EPRI (1999b) West Virginia power plant study compared to concentrations predicted by adsorption onto hydrous ferric oxide (calculated with PHREEQC)

The simulation suggests that adsorption likely plays a key role in limiting aqueous beryllium concentrations in CCP environments; perhaps more so than mineral solubility. By this mechanism, the amount of beryllium in solution is affected by the amount of reactive adsorption sites in contact with the solution (e.g., the mass of hydrous ferric oxide or other adsorption surfaces). These results also suggest that there are more available sorption sites for cations in coal piles than in ash, based on the closer fit of coal pile runoff samples to the higher reactive site concentration. This may result from the relative abundance or organic matter in coal as compared to ash.

6 ANALYSIS, TREATMENT, AND REMEDIATION

Analytical Techniques

Laboratory analysis methods for environmental samples containing beryllium are given in Table 6-1. For solid sample analysis, various digestion methods are used for sample preparation, depending upon whether complete solid digestion or partial digestion is desired. The EPA publication SW-846 (*Test Methods for Evaluating Solid Waste, Physical/Chemical Methods*), describes analytical and sampling methods that have been evaluated and approved for use in complying with the RCRA regulations. The publication functions primarily as a guidance document for acceptable methods for the regulated and regulatory communities to use in responding to RCRA-related sampling and analysis requirements (USEPA, 2004a).

Sample matrix	Procedure*	EPA method	Detection limit
	FLAA	7090	5 µg/L
Aqueous samples, extracts, wastes	ICP	6010	0.18 μg/L
	GFAA	7091	0.2 μg/L
Oils, greases, waxes	FLAA	7090	5 µg/L
(organic extract)	ICP	6010	0.18 μg/L
	FLAA	7090	5 µg/L
Sediment, sludges, soils	ICP	6010	0.18 μg/L
	GFAA	7091	0.2 μg/L

 Table 6-1

 Environmental sample analysis methods for beryllium (after Taylor et al., 2003)

* FLAA: flame atomic absorption spectrometry; GFAA: graphite furnace atomic absorption spectrometry; ICP: inductively coupled argon plasma emission spectrometry

Treatment and Remediation

Information regarding treatment and remediation of beryllium is limited by the fact that beryllium is rarely of environmental concern, especially at CCP disposal facilities. Recognized

Analysis, Treatment, and Remediation

and potential treatment technologies for water and solid materials containing beryllium are described below and summarized in Table 6-2. The most effective treatment for beryllium-containing environmental media is pH control to circum-neutral to alkaline pH, a natural characteristic of most coal fly ash leachate. Treatability studies need to be performed on candidate remedial technologies to determine effectiveness and cost on a site-specific basis.

Water Treatment

- Activated alumina relies on adsorption of beryllium to an extremely porous media composed mainly of alumina ore. The ore is activated by passing oxidizing gases through the material at high temperatures, producing pores that result in very high adsorption capacity. Water is passed through a cartridge or canister of activated alumina, whereby the beryllium is adsorbed to the media. The media can become ineffective due to its finite capacity of adsorption sites; therefore, flushing or removal and disposal of the media will be required (USDOI, 2001).
- *Coagulation and filtration* involves chemical addition, rapid mixing, coagulation with dry alum, flocculation, and dual media filtration. Chemical coagulation and flocculation utilizes a coagulant such as Al₂(SO₄)₃ (proven most effective for insoluble Be removal) combined with mechanical flocculation to promote settling of fine material and flocculation of dissolved components. The final step requires filtration of both flocs and suspended solids (USDOI, 2001). EPRI (1989) found that iron adsorption/coprecipitation could remove beryllium from a variety of wastewater streams at pH greater than 6.
- *Cation ion exchange (IX)* is a process by which cations in a solution are exchanged for cations that exist on an insoluble, permanent, solid resin bed. Because water solutions tend toward a balance of electrical charge, only ions of similar charge are exchanged. A polymer resin bed, composed of millions of medium sand grain size spherical beads, is placed in contact with the beryllium-containing solution. As water passes through the bed, beryllium ions are allowed to exchange with cations loaded on the bed (USDOI, 2001).
- *Lime softening* includes adding Ca(OH)₂ in sufficient amounts to raise the solution pH (optimum pH is approximately 10) while keeping low alkalinity levels to precipitate carbonate hardness. Beryllium will precipitate as Be(OH)₂ during this process. The precipitation of Be(OH)₂ is followed by coagulation and flocculation of suspended materials and subsequent clarification. The solution is clarified in an upflow solids contact clarifier where water flows up and over weirs, while settled particles are removed by pumping or other mechanisms (USDOI, 2001).
- *Reverse osmosis (RO)* is the process of applying pressure on contaminated feed water to direct it through a semipermeable membrane, such as asymmetric cellulose acetate or a polyamide thin film composite. Natural osmotic pressure will cause diluted water to diffuse into a concentrated solution to balance solution concentrations. This process is overcome by applying pressure to the concentrated solution. RO membranes have the ability to reject ions based on size and electrical charge. Rejected material, called concentrate, can be removed from the process and disposed of (USDOI, 2001).

Solid Treatment

- *Fixation and vitrification*: Bhat and Pillai (1997) studied the immobilization of beryllium in solid waste by fixation and vitrification. They found that incorporating beryllium into cement blocks and vitrification of red mud (bauxite ore processing waste) are both effective means of reducing the leaching potential of beryllium from solid waste.
- *Cement-based grouting*: Conley and Gilliam (1998) successfully immobilized beryllium in a cement-based grout. The waste consisted of lab grade beryllium, beryllium oxide powders, beryllium powder mixed with other metals, beryllium detector windows, and beryllium metal, foil, shavings, and shot. They reported that RCRA Universal Treatment standards were achieved with 40% (by weight) waste loadings.
- *Chemical fixation*: One of the most useful properties of beryllium with respect to treatment is its strong tendency toward hydrolysis and the insolubility of its hydroxide form in nearneutral solutions. As a result, almost any precipitate which is formed in solution at nearneutral pH will co-precipitate beryllium to some extent. This becomes especially useful with respect to precipitation of Fe- and Al- hydroxides. Therefore, treatments that utilize Fe or Al hydroxide precipitation at near-neutral pH are expected to be highly effective in immobilizing beryllium in a solid matrix.

Technology	Estimated costs (2006 \$)	
	Capital (total; 1 MGD)	Annual operation and maintenance (\$/yr; 1 MGD)
Activated alumina adsorption ^{1*}	\$639,000	\$284,000
Coagulation and filtration ^{1*}	\$3,000,000	\$161,000
Cation exchange ^{1*}	\$562,000	\$99,000
Lime softening ¹	\$65,000	\$33,000
Reverse osmosis ¹	\$730,000	\$324,000
In situ vitrification ²	~\$300-\$400/yd ³	
In situ chemical fixation with Portland cement ³	\$95-\$200/yd ³	

Table 6-2 Potential beryllium treatment technologies and relative costs

¹Reference: "Technologies and Costs for Removal of Arsenic from Drinking Water," U.S. EPA., Office of Water, EPA Report 815-R-00-028, December 2000. (www.epa.gov/safewater). Based on 1 million gallons per day (MGD) treated.

²USEPA (1997a); available online at http://www.clu-in.org/products/costperf/VITRIFIC/Parsons2.htm#TSCOST ³EPRI (1996)

*Includes waste disposal costs.

7 CONCLUSIONS AND RECOMMENDATIONS

Conclusions

- Average concentrations of beryllium in coal are similar to those of igneous rocks, shales, and United States soils (<5 mg/kg). Alaskan soils are reported to contain up to 300 mg/kg, and soils of the Southeastern U.S. Coastal Plain up to 30.5 mg/kg. Bulgarian coals contain the highest beryllium concentrations found in the literature during this study (up to 32.6 mg/kg). Coal fly ash will generally contain from 2 to 11 times the concentration of the feed coal, and concentrations in both fly ash and flue gas desulfurization solids are mostly less than 20 mg/kg. Beryllium is usually concentrated in fly ash relative to bottom ash.
- Beryllium's mode of occurrence in coal and coal ash is unresolved. Some studies report a primary organic affinity of beryllium in coal, while others report an inorganic association. One proposition is that beryllium may be contained in very fine silicate phases embedded within the organic matrix. Beryllium in coal ash likely has an inorganic association.
- Beryllium does not leach appreciably from coal ash, which typically is in the neutral to alkaline pH range, but can leach from coal piles under acidic conditions. Available field data suggest that beryllium leaching from coal and coal ash may result from either mineral dissolution (chrysoberyl) or desorption from solid phases. The most important factor that seems to limit beryllium mobility at CCP facilities is pH. Concentrations generally do not exceed the MCL of 4 μ g/L above pH 4.
- Beryllium mobility in soil and groundwater is limited by its strong tendency to adsorb to solid phases or to precipitate as beryllium hydroxide. Complete removal of beryllium from solution by adsorption can occur at and above pH of approximately 5.5. For this reason, surface water and drinking water levels of beryllium are mostly very low. Aqueous concentrations decrease in the order: soil water > groundwater > stream water > river water > estuary water > ocean water.
- When in solution, beryllium will usually occur as Be-F, Be²⁺, Be-OH, or BeSO₄⁰ species. Complexing of metals with fluoride, sulfate, and carbonate usually tends to enhance metal solubility. Binding to fulvic acids can occur above pH 6. Humic material has been shown to increase beryllium solubility in solutions in contact with kaolinite and amorphous silica.
- There have been few studies on the effects of beryllium on plants. Beryllium in watersoluble forms tends to be toxic to plants, but soil tends to fix beryllium in forms that makes it unavailable to plants.
- Oral ingestion of beryllium has not been associated with any human clinical diseases. While lung cancer is probably associated with increased beryllium exposures from inhalation, the best data seem to indicate that the high exposures that occurred at beryllium processing

Conclusions and Recommendations

facilities in the past were responsible, and suggest that current practices greatly reduce the potential for exposure at cancer-causing levels.

- Using a suggested dilution attenuation factor of 20, the generic soil screening level calculated for beryllium is 63 mg/kg. Ecological soil screening levels are similarly high: 40 mg/kg for avian wildlife and 21 mg/kg for mammalian wildlife. Background soil concentrations are mostly much lower, averaging 0.63 mg/kg across the coterminous United States. Coal and coal ash concentrations (maximum = 18 and 27 mg/kg, respectively) are almost always lower than these screening levels, and are usually below 10 mg/kg.
- Beryllium in the aqueous phase is rarely of environmental concern, and information regarding beryllium treatment is limited. Available data suggest that pH control is a dominant factor for removing beryllium from solution. The following technologies have potential for treating beryllium in both liquid and solid forms:
 - o Activated alumina adsorption
 - Coagulation and filtration
 - Cation exchange
 - Lime softening
 - Reverse osmosis filtration
 - Physical fixation and vitrification
 - Cement-based grouting
 - Chemical fixation

Recommendations

- Additional studies involving beryllium leachability from solid materials should place focused attention on final pH of the solution. Initial pH of the extractant solution provides little insight as to what controls beryllium leaching. The final pH represents an equilibrium condition reached between the extractant solution and the solid phase. Beryllium leaching should also be studied in the context of aluminum and silica leaching.
- More studies involving beryllium adsorption onto natural soil materials should be conducted. Environmental fate and transport of beryllium may be readily predicted with better understanding of beryllium partitioning between the aqueous phase and natural soils.
- The mode of occurrence of beryllium in coal and other solid material may be better determined by more advanced sequential chemical extraction procedures. Often, methods consisting of few extraction steps create ambiguous results. Therefore, more detailed, seven-step procedures may help to better define the relationship of beryllium with specific mineral fractions.
- Beryllium concentrations in coal and CCP material will most often exceed background soil concentrations, but will likely not exceed generic and ecological soil screening levels. The limiting effect of low regulatory standards for beryllium with respect to the beneficial use of CCP material deserves focused attention.

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