

6. POTENTIAL FOR HUMAN EXPOSURE

6.1 OVERVIEW

Aluminum has been identified in at least 596 of the 1,699 hazardous waste sites that have been proposed for inclusion on the EPA National Priorities List (NPL) (HazDat 2008). However, the number of sites evaluated for aluminum is not known. The frequency of these sites can be seen in Figure 6-1. Of these sites, 590 are located within the United States, 2 are located in Guam, 3 are located in the Commonwealth of Puerto Rico, and 1 is located in the Virgin Islands (not shown).

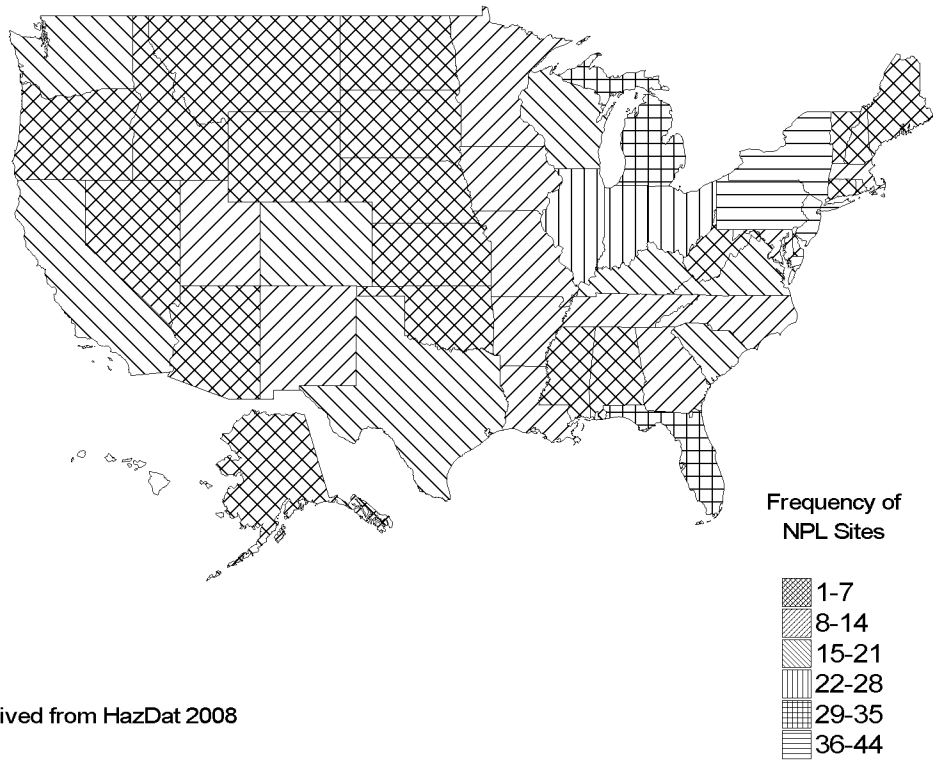
Aluminum is the most abundant metal and the third most abundant element in the earth's crust, comprising about 8.8% by weight (88 g/kg). It is never found free in nature and is found in most rocks, particularly igneous rocks as aluminosilicate minerals (Lide 2005; Staley and Haupin 1992). Aluminum is also present in air, water, and many foods. Aluminum enters environmental media naturally through the weathering of rocks and minerals. Anthropogenic releases are in the form of air emissions, waste water effluents, and solid waste primarily associated with industrial processes, such as aluminum production. Because of its prominence as a major constituent of the earth's crust, natural weathering processes far exceed the contribution of releases to air, water, and land associated with human activities (Lantzy and MacKenzie 1979).

The behavior of aluminum in the environment depends upon its coordination chemistry and the characteristics of the local environment, especially pH. The major features of the biogeochemical cycle of aluminum include leaching of aluminum from geochemical formations and soil particulates to aqueous environments, adsorption onto soil or sediment particulates, and wet and dry deposition from the air to land and surface water.

Generally, aluminum is not bioaccumulated to a significant extent. However, certain plants can accumulate high concentrations of aluminum. For example, tea leaves may contain very high concentrations of aluminum, >5,000 mg/kg in old leaves (Dong et al. 1999). Other plants that may contain high levels of aluminum include *Lycopodium* (Lycopodiaceae), a few ferns, *Symplocos* (Symplocaceae), and *Orites* (Proteaceae) (Jansen et al. 2002). Aluminum does not appear to accumulate to any significant degree in cow's milk or beef tissue and is, therefore, not expected to undergo biomagnification in terrestrial food chains (DOE 1984). Similarly, because of its toxicity to many aquatic organisms, including fish, aluminum does not bioconcentrate in aquatic organisms to any significant degree (Rosseland et al. 1990).

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Figure 6-1. Frequency of NPL Sites with Aluminum Contamination



Derived from HazDat 2008

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Background concentrations of aluminum in rural air typically range from 0.005 to 0.18 $\mu\text{g}/\text{m}^3$ (Hoffman et al. 1969; Pötzl 1970; Sorenson et al. 1974), whereas concentrations in urban and industrial areas can be considerably higher, ranging from 0.4 to 8.0 $\mu\text{g}/\text{m}^3$ (Cooper et al. 1979; Dzubay 1980; Kowalczyk et al. 1982; Lewis and Macias 1980; Moyers et al. 1977; Ondov et al. 1982; Pillay and Thomas 1971; Sorenson et al. 1974; Stevens et al. 1978). Concentrations of aluminum are highly variable in drinking water, ranging from <0.001 to 1.029 mg/L (Schenk et al. 1989). The use of alum (aluminum sulfate) as a flocculent in water treatment facilities typically leads to high aluminum concentrations in finished waters (DOI 1970; Letterman and Driscoll 1988; Miller et al. 1984a). In a survey of 186 community water systems, the median aluminum concentration in finished water receiving coagulation treatment using alum was 0.112 mg/L, compared to 0.043 mg/L in finished water that received no coagulation treatment (Miller et al. 1984a). Dissolved aluminum concentrations in surface and groundwater vary with pH and the humic acid content of the water. High aluminum concentrations in natural water occur only when the pH is <5; therefore, concentrations in most surface water are very low.

Since aluminum is ubiquitous in the environment, the general population will be exposed to aluminum by the inhalation of ambient air and the ingestion of food and water. The consumption of foods containing aluminum-containing food additives are a major sources of aluminum in the diet (Saiyed and Yokel 2005; Soni et al. 2001). The use of other consumer items such as antiperspirants, cosmetics, internal analgesics (buffered aspirins), anti-ulcerative medications, antidiarrheals, and antacids that also contain aluminum compounds will result in exposure to aluminum. The intake of aluminum from food and drinking water is low, especially compared with that consumed by people taking aluminum-containing medicinal preparations. Daily intakes of aluminum from food range from 3.4 to 9 mg/day (Biego et al. 1998; MAFF 1999; Pennington and Schoen 1995), whereas aluminum-containing medications contain much higher levels of aluminum, for example 104–208 mg of aluminum per tablet/capsule/5 mL dose for many antacids (Zhou and Yokel 2005). While aluminum is naturally present in food and water, the greatest contribution to aluminum in food and water by far is the aluminum-containing additives used in water treatment and processing certain types of food such as grain-based products and processed cheese. Aluminum has no known physiological role in the human body (Nayak 2002).

The aluminum content of human breast milk generally ranged from 9.2 to 49 $\mu\text{g}/\text{L}$ (Fernandez-Lorenzo et al. 1999; Hawkins et al. 1994; Koo et al. 1988; Simmer et al. 1990; Weintraub et al. 1986). Soy-based infant formulas contain higher concentrations of aluminum, as compared to milk-based infant formulas or breast milk. Recent reports provide average aluminum concentrations of 460–930 $\mu\text{g}/\text{L}$ for soy-based

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infant formulas and 58–150 µg/L for milk-based formulas (Fernandez-Lorenzo et al. 1999; Ikem et al. 2002; Navarro-Blasco and Alvarez-Galindo 2003).

Occupational exposures to aluminum occur during the mining and processing of aluminum ore into metal, recovery of scrap metal, production and use of aluminum compounds and products containing these compounds, and in aluminum welding. Individuals living in the vicinity of industrial emission sources and hazardous waste sites; individuals with chronic kidney failure requiring long-term dialysis or treatment with phosphate binders; patients requiring intravenous fluids; infants, especially premature infants fed soy-based formula containing high levels of aluminum; and individuals consuming large quantities of antacids, anti-ulcerative medications, antidiarrheal medications may also be exposed to high levels of aluminum.

According to the Toxic Chemical Release Inventory, in 2005, total releases of aluminum (fume or dust) to the environment (including air, water, and soil) from 329 large processing facilities were 45.6 million pounds ($\sim 2.07 \times 10^4$ metric tons) (TRI05 2007). In addition, in 2005, total releases of aluminum oxide (fibrous forms) to the environment (including air, water, and soil) from 59 large processing facilities were 2.59 million pounds (~ 1180 metric tons) (TRI05 2007). Tables 6-1 and 6-2 list amounts released from these facilities grouped by state. The TRI data should be used with caution because only certain types of facilities are required to report (EPA 2005). This is not an exhaustive list.

6.2 RELEASES TO THE ENVIRONMENT

The Toxics Release Inventory (TRI) data should be used with caution because only certain types of facilities are required to report (EPA 2005). This is not an exhaustive list. Manufacturing and processing facilities are required to report information to the TRI only if they employ 10 or more full-time employees; if their facility is included in Standard Industrial Classification (SIC) Codes 10 (except 1011, 1081, and 1094), 12 (except 1241), 20–39, 4911 (limited to facilities that combust coal and/or oil for the purpose of generating electricity for distribution in commerce), 4931 (limited to facilities that combust coal and/or oil for the purpose of generating electricity for distribution in commerce), 4939 (limited to facilities that combust coal and/or oil for the purpose of generating electricity for distribution in commerce), 4953 (limited to facilities regulated under RCRA Subtitle C, 42 U.S.C. section 6921 et seq.), 5169, 5171, and 7389 (limited S.C. section 6921 et seq.), 5169, 5171, and 7389 (limited to facilities primarily engaged in solvents recovery services on a contract or fee basis); and if their facility produces,

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Table 6-1. Releases to the Environment from Facilities that Produce, Process, or Use Aluminum (Fume or Dust)^a

State ^c	RF ^d	Reported amounts released in pounds per year ^b								
		Air ^e	Water ^f	UI ^g	Land ^h	Other ⁱ	Total release			On- and off-site
							On-site ^j	Off-site ^k		
AK	1	0	0	0	0	1	0	0	0	
AL	4	2,453	0	0	45,887	48,344	2,453	45,887	48,340	
AR	3	0	0	0	0	3	0	750	750	
AZ	4	7,167	0	0	230,729	237,900	7,217	230,679	237,896	
CA	16	182,017	0	0	1,662,654	1,844,688	1,802,363	42,364	1,844,727	
CO	1	1,500	0	0	53,058	54,559	1,500	53,058	54,558	
CT	2	0	0	0	0	2	0	0	0	
FL	5	1,624	0	0	23	1,652	1,624	23	1,647	
GA	10	37,680	0	0	108,219	145,909	37,680	108,871	146,551	
IA	8	11,570	0	0	43,052	54,630	11,570	43,052	54,622	
ID	2	2,864	0	0	653,345	656,211	518,203	138,006	656,209	
IL	16	62,008	0	0	520,607	582,631	62,008	525,882	587,890	
IN	29	149,220	0	0	10,023,429	10,172,678	149,225	10,024,174	10,173,399	
KS	3	0	0	0	0	3	0	0	0	
KY	13	254,892	0	0	2,799,380	3,054,285	2,419,281	634,991	3,054,272	
LA	4	1,184	0	0	13	1,201	1,197	0	1,197	
MA	2	No data	No data	No data	No data	No data	No data	0	0	
MD	2	0	0	0	0	2	0	0	0	
MI	16	17,862	0	0	1,215,365	1,233,243	17,862	1,224,508	1,242,370	
MN	6	58,268	0	0	157,233	215,507	58,268	157,277	215,545	
MO	8	29,495	0	0	1,941,390	1,970,893	1,828,685	7,037,274	8,865,959	
MS	1	0	0	0	550	551	0	550	550	
NC	8	62,432	0	0	55,340	117,780	63,182	54,590	117,772	
NE	2	0	0	0	31,105	31,107	21,263	9,842	31,105	
NJ	6	4,413	0	0	5,222	9,641	4,413	5,222	9,635	
NM	1	No data	No data	No data	No data	No data	No data	0	0	
NV	4	259	0	0	330,084	330,347	330,343	0	330,343	
NY	7	58,438	312	0	198,222	256,979	60,436	213,027	273,463	
OH	33	50,159	2	0	2,170,512	2,220,706	253,161	2,237,892	2,491,053	
OK	9	9,654	0	0	447,920	457,583	10,416	447,158	457,574	
OR	4	646	0	0	1,295	1,945	646	1,295	1,941	
PA	23	8,594	0	0	294,833	303,450	8,866	317,627	326,493	
RI	1	No data	No data	No data	No data	No data	No data	0	0	
SC	4	7,841	0	0	750	8,595	7,841	1,500	9,341	
TN	21	93,021	0	0	887,911	980,953	185,281	795,651	980,932	
TX	19	89,879	0	0	7,670,584	7,760,481	2,590,719	5,171,544	7,762,262	

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Table 6-1. Releases to the Environment from Facilities that Produce, Process, or Use Aluminum (Fume or Dust)^a

State ^c	RF ^d	Reported amounts released in pounds per year ^b							
		Air ^e	Water ^f	UI ^g	Land ^h	Other ⁱ	Total release		On- and off-site
							On-site ^j	Off-site ^k	
UT	6	187	0	0	372,634	372,827	147,182	225,639	372,821
VA	2	137	0	0	1,000	1,139	137	1,000	1,137
WA	4	846	0	0	128,819	129,669	846	129,421	130,267
WI	14	75,933	0	0	289,933	365,880	75,933	290,099	366,032
WV	5	3,112	0	0	4,732,012	4,735,129	3,112	4,732,012	4,735,124
Total	329	1,285,354	314	0	37,073,110	38,359,108	10,682,912	34,900,865	45,583,777

^aThe TRI data should be used with caution since only certain types of facilities are required to report. This is not an exhaustive list. Data are rounded to nearest whole number.

^bData in TRI are maximum amounts released by each facility.

^cPost office state abbreviations are used.

^dNumber of reporting facilities.

^eThe sum of fugitive and point source releases are included in releases to air by a given facility.

^fSurface water discharges, waste water treatment-(metals only), and publicly owned treatment works (POTWs) (metal and metal compounds).

^gClass I wells, Class II-V wells, and underground injection.

^hResource Conservation and Recovery Act (RCRA) subtitle C landfills; other on-site landfills, land treatment, surface impoundments, other land disposal, other landfills.

ⁱStorage only, solidification/stabilization (metals only), other off-site management, transfers to waste broker for disposal, unknown

^jThe sum of all releases of the chemical to air, land, water, and underground injection wells.

^kTotal amount of chemical transferred off-site, including to POTWs.

RF = reporting facilities; UI = underground injection

Source: TRI05 2007 (Data are from 2005)

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Table 6-2. Releases to the Environment from Facilities that Produce, Process, or Use Aluminum Oxide (Fibrous Forms)^a

State ^c	RF ^d	Reported amounts released in pounds per year ^b								
		Air ^e	Water ^f	UI ^g	Land ^h	Other ⁱ	Total release			
							On-site ^j	Off-site ^k	On- and off-site	
AL	2	0	0	0	0	0	0	0	0	0
AR	1	0	0	0	0	0	0	0	0	0
CA	1	0	No data	0	0	0	0	0	0	0
CO	1	0	5	0	480	2,749	485	2,749	3,234	
CT	1	0	0	0	0	0	0	0	0	0
GA	2	16	175	0	2,957	0	191	2,957	3,148	
IA	2	0	0	0	40,320	0	0	40,320	40,320	
IL	5	76	0	0	122,002	22,660	76	144,662	144,738	
IN	3	901	250	0	5	10	1,156	10	1,166	
KY	3	243	0	0	26,631	0	243	26,631	26,874	
LA	2	0	0	0	0	0	0	0	0	
MI	2	0	0	0	375,000	0	0	375,000	375,000	
MO	1	250	0	0	750	0	1,000	0	1,000	
NC	4	56	10	0	60,797	4,342	61	65,144	65,205	
NE	1	2	0	0	20	0	22	0	22	
NM	1	0	0	0	0	0	0	0	0	
NY	2	250	0	0	0	29,808	250	29,808	30,058	
OH	2	980	0	0	110,958	0	980	110,958	111,938	
OK	1	0	No data	0	0	0	0	0	0	
PA	6	247	0	0	178,893	6,781	247	185,674	185,920	
SC	2	14	0	7	23,556	424	14	23,987	24,001	
TN	4	3	0	0	0	0	3	0	3	
TX	4	11	0	0	431,166	0	11	431,166	431,177	
VA	2	500	0	0	37,159	0	37,409	250	37,659	
WI	3	260	0	0	1,059,128	0	1,059,138	250	1,059,388	

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Table 6-2. Releases to the Environment from Facilities that Produce, Process, or Use Aluminum Oxide (Fibrous Forms)^a

State ^c	RF ^d	Air ^e	Water ^f	UI ^g	Land ^h	Other ⁱ	Reported amounts released in pounds per year ^b		
							Total release		On- and off-site
							On-site ^j	Off-site ^k	
WV	1	0	0	0	48,000	0	48,000	0	48,000
Total	59	3,810	440	7	2,517,822	66,774	1,149,287	1,439,565	2,588,852

^aThe TRI data should be used with caution since only certain types of facilities are required to report. This is not an exhaustive list. Data are rounded to nearest whole number.

^bData in TRI are maximum amounts released by each facility.

^cPost office state abbreviations are used.

^dNumber of reporting facilities.

^eThe sum of fugitive and point source releases are included in releases to air by a given facility.

^fSurface water discharges, waste water treatment-(metals only), and publicly owned treatment works (POTWs) (metal and metal compounds).

^gClass I wells, Class II-V wells, and underground injection.

^hResource Conservation and Recovery Act (RCRA) subtitle C landfills; other on-site landfills, land treatment, surface impoundments, other land disposal, other landfills.

ⁱStorage only, solidification/stabilization (metals only), other off-site management, transfers to waste broker for disposal, unknown

^jThe sum of all releases of the chemical to air, land, water, and underground injection wells.

^kTotal amount of chemical transferred off-site, including to POTWs.

RF = reporting facilities; UI = underground injection

Source: TRI05 2007 (Data are from 2005)

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imports, or processes $\geq 25,000$ pounds of any TRI chemical or otherwise uses $>10,000$ pounds of a TRI chemical in a calendar year (EPA 2005).

6.2.1 Air

Estimated releases of 1.29 million pounds (~586 metric tons) of aluminum (fume or dust) to the atmosphere from 329 domestic manufacturing and processing facilities in 2005, accounted for about 2.8% of the estimated total environmental releases from facilities required to report to the TRI (TRI05 2007).

Estimated releases of 3,810 pounds (~1.73 metric tons) of aluminum oxide (fibrous forms) to the atmosphere from 59 domestic manufacturing and processing facilities in 2005, accounted for about 1.5% of the estimated total environmental releases from facilities required to report to the TRI (TRI05 2007).

These releases are summarized in Tables 6-1 and 6-2.

Aluminum is released to the environment by both natural processes and anthropogenic sources. Because of its prominence as a major constituent of the earth's crust, natural processes far exceed the contribution of anthropogenic releases to the environmental distribution of aluminum (Lantzy and MacKenzie 1979). Anthropogenic releases are primarily to the atmosphere. The largest source of airborne aluminum-containing particulates is the flux of dust from soil and the weathering of rocks (Lee and Von Lehmden 1973; Sorenson et al. 1974). In addition, aluminum-containing dust is generated by volcanic activity (Varrica et al. 2000). Human activities, such as mining and agriculture, contribute to this wind-blown dust (Eisenreich 1980; Filipek et al. 1987). About 13% of atmospheric aluminum is attributed to anthropogenic emissions (Lantzy and MacKenzie 1979). The major anthropogenic sources of aluminum-containing particulate matter include coal combustion, aluminum production, and other industrial activities, such as smelting, that process crustal minerals (Lee and Von Lehmden 1973). Aluminum concentrations in air particulate emissions from iron and steel foundries and brass and bronze refineries range from about 100 to 1,000 ppm (Lee and Von Lehmden 1973). Que Hee et al. (1982) also found that aluminum was one of the most abundant elements quantified in coal stack emissions from power plants located in both the eastern and western United States. In addition, in U.S. cities, motor vehicle emissions contribute an estimated 0.9–9% of the observed elemental concentration of aluminum in these atmospheres (Ondov et al. 1982).

Aluminum has been identified in air samples collected at 14 of the 596 NPL hazardous waste sites where it has been detected in some environmental media (HazDat 2008).

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6.2.2 Water

Estimated releases of 314 pounds (~0.14 metric tons) of aluminum (fume or dust) to surface water from 329 domestic manufacturing and processing facilities in 2005, accounted for about 0.0007% of the estimated total environmental releases from facilities required to report to the TRI (TRI05 2007).

Estimated releases of 440 pounds (~0.20 metric tons) of aluminum oxide (fibrous forms) to surface water from 59 domestic manufacturing and processing facilities in 2005, accounted for about 0.017% of the estimated total environmental releases from facilities required to report to the TRI (TRI05 2007). These releases are summarized in Tables 6-1 and 6-2.

Aluminum occurs ubiquitously in natural waters as a result of the weathering of aluminum-containing rocks and minerals. Of the known geochemical responses to environmental acidification, the best documented is the mobilization of aluminum from terrestrial to aquatic environments (Campbell et al. 1992). This mobilization of aluminum is often episodic in nature and is associated with pH depressions (acidification) occurring during the spring snowmelt or associated with erosion from specific storm events (Campbell et al. 1992; Nelson and Campbell 1991; Rosseland et al. 1990).

Aluminum concentrations in surface waters can be increased directly or indirectly by human activity through industrial and municipal discharges, surface run-off, tributary inflow, groundwater seepage, and wet and dry atmospheric deposition (Eisenreich 1980). For example, aluminum is released to surface waters in the effluent from bauxite processing and aluminum manufacturing facilities at concentrations that can be toxic to aquatic life (His et al. 1996; Trieff et al. 1995). However, the effluents of these facilities typically contain not only aluminum, but also a complex mixture of heavy metals such as iron, chromium, and mercury, as well as minerals, silica, and other compounds, and synergistic effects of these metals and compounds cannot be ruled out. The use of aluminum sulfate and other aluminum compounds as coagulating agents in the treatment of raw drinking water supplies can significantly increase the total aluminum content in finished water (Cech and Montera 2000; Henshaw et al. 1993; Miller et al. 1984a; Qureshi and Malmberg 1985; USGS 1984b). Weathering of sulfide ores exposed to the atmosphere in inactive mines and tailings dumps releases large quantities of sulfuric acid and metals such as aluminum (Filipek et al. 1987). Increasingly, acid environments caused by such acid mine drainage or by acid rain will subsequently cause an increase in the dissolved aluminum content of the surrounding waters (Brusewitz 1984; Filipek et al. 1987). In addition, atmospheric deposition is a source of aluminum input to surface water. The atmospheric loading of aluminum to Lake Michigan was estimated to be 5 million

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kg/year, of which 74% was to the southern basin where the influence of agricultural and industrial activity (e.g., steel manufacturing and cement production) was greatest (Eisenreich 1980).

Aluminum has been identified in surface water and groundwater samples collected at 251 and 391 of the 596 NPL hazardous waste sites, respectively, where it has been detected in some environmental media (HazDat 2008).

6.2.3 Soil

Estimated releases of 37.1 million pounds ($\sim 1.68 \times 10^4$ metric tons) of aluminum (fume or dust) to soils from 329 domestic manufacturing and processing facilities in 2005, accounted for about 81% of the estimated total environmental releases from facilities required to report to the TRI (TRI05 2007).

Estimated releases of 2.52 million pounds ($\sim 1,140$ metric tons) of aluminum oxide (fibrous forms) to soils from 59 domestic manufacturing and processing facilities in 2005, accounted for about 97% of the estimated total environmental releases from facilities required to report to the TRI (TRI05 2007). An additional 7 pounds (3 kilograms) of aluminum oxide (fibrous forms) were released via underground injection (TRI05 2007). These releases are summarized in Tables 6-1 and 6-2.

Aluminum is the most abundant metal and the third most abundant element in the earth's crust, comprising about 8.8% by weight (88 g/kg) (Staley and Haupin 1992). Aluminum can be released naturally by the weathering of aluminum-containing rocks. Aluminum is also released to soil as a major constituent of many mining wastes and is also contained in solid wastes from coal combustion and aluminum reduction and other metal processing operations (DOI 1983, 1984). Wilson et al. (2002) estimated that several hundred thousand pounds of aluminum containing chaff have been released to the Chesapeake Bay during research and training operations by the Naval Research Laboratory-Chesapeake Bay Detachment over the past 25 years.

Aluminum has been identified in soil and sediment samples collected at 253 and 190 of the 596 NPL hazardous waste sites, respectively, where it has been detected in some environmental media (HazDat 2008).

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6.3 ENVIRONMENTAL FATE**6.3.1 Transport and Partitioning**

Aluminum is the most abundant metal in the earth's crust, but is never found in its elemental state in nature. In compounds, aluminum occurs in its only oxidation state (+3) (Lide 2005). Aluminum occurs widely in nature with silicates, such as mica and feldspar, as the hydroxo oxide (bauxite), and as cryolite (Na_3AlF_6) (Cotton et al. 1999). Aluminum's behavior in the environment is strongly influenced by its coordination chemistry. Aluminum partitions between solid and liquid phases by reacting and complexing with water molecules and anions such as chloride, fluoride, sulfate, nitrate, phosphate, and negatively charged functional groups on humic materials and clay.

The transport and partitioning of aluminum in the environment is determined by its chemical properties, as well as the characteristics of the environmental matrix that affect its solubility. At a pH >5.5, naturally occurring aluminum compounds exist predominantly in an undissolved form such as gibbsite, $\text{Al}(\text{OH})_3$, or as aluminosilicates except in the presence of high amounts of dissolved organic material or fulvic acid, which binds with aluminum and can cause increased dissolved aluminum concentrations in streams and lakes (Brusewitz 1984). Organic acids have been found to be important weathering agents for dissolving and transporting aluminum in an alpine soil environment (Litaor 1987). The ability of these organic acids to complex aluminum in sub-alpine soil solutions was found to increase as the pH rose from 3.8 to 5 (Dahlgren and Ugolini 1989). In this study, dissolved aluminum was found primarily as organic complexes when organic carbon/metal ratios were >50 (Dahlgren and Ugolini 1989).

In general, decreasing pH (acidification) results in an increase in mobility for monomeric forms of aluminum (Goenaga and Williams 1988), which is of concern with respect to the occurrence of acid rain and the release of acid mine drainage. Aluminum in soil solutions and surface waters in a mining region rich in metallic sulfides was in a labile form, as Al-SO_4 and Al^{3+} species. Acidic conditions are created by the microbial oxidation of sulfides in tailing piles, resulting in sulfuric acid. In contrast, in areas not affected by acidification, aluminum in solution was partitioned between labile and non-labile forms, the latter being predominantly bound to fluorine (Alvarez et al. 1993). In soils, the most soluble form of aluminum under acidic conditions is nonsiliceous, organically-bound aluminum (Mulder et al. 1989).

In groundwater or surface water systems, an equilibrium with a solid phase form is established that largely controls the extent of aluminum dissolution which can occur. In acid sulfate waters resulting from mine drainage, gibbsite and kaolinite are not stable, and the solubility of the minerals jurbanite

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($\text{Al}(\text{SO}_4)(\text{OH})\cdot\text{H}_2\text{O}$) or alunite ($\text{KAl}_3(\text{SO}_4)_2(\text{OH})_6$) may control aluminum levels (Filipek et al. 1987). In a Colorado alpine watershed soil, the chemical equilibria of aluminum in interstitial water at a pH range of 4.4–7.2 were controlled by amorphous aluminosilicate rather than gibbsite (Litaor 1987).

In addition to the effect of pH on mobility, the type of acid entering environmental systems may also be important. Nitric acid was found to leach more aluminum from soil columns representative of high-elevation forest floor soils than did sulfuric acid (James and Riha 1989). However, in mineral horizons below the forest floor, the study found that concentrations of aluminum leached by these acids did not differ from concentrations of aluminum leached by distilled, deionized water at a pH of 5.7. The authors concluded that soluble constituents from the forest floor affected the aluminum solubility in the underlying mineral horizons under the leaching conditions that they used. These constituents may have included natural buffering agents which resist changes in pH and, therefore, negate or mediate the effect of the acid.

The ability of mineralized soil to control the migration of aluminum was observed in another study. Acidic leachate from coal waste containing aluminum was percolated through soil containing varying amounts of calcium carbonate (Wangen and Jones 1984). Soluble aluminum was found to decrease dramatically as the pH of the percolating leachate increased and aluminum oxide precipitates formed; at pH 6, no dissolved aluminum was measured. The authors concluded that alkalized carbonaceous soils provide the best control material for acidic leachates from coal mineral wastes.

The adsorption of aluminum onto clay surfaces can be a significant factor in controlling aluminum mobility in the environment, and these adsorption reactions, measured in one study at pH 3.0–4.1, have been observed to be very rapid (Walker et al. 1988). However, clays may act either as a sink or a source for soluble aluminum depending on the degree of aluminum saturation on the clay surface (Walker et al. 1988).

The presence of high levels of suspended solids in stream surface water during storm episodes resulted in higher concentrations of adsorbed aluminum than in the absence of suspended solids (Goenaga and Williams 1988). The increased adsorption was not strictly linear, with higher concentrations of suspended solids due to variations in the particle size distribution and the nature of the particles.

Within the pH range of 5–6, aluminum complexes with phosphate and is removed from solution. Because phosphate is a necessary nutrient in ecological systems, this immobilization of both aluminum

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and phosphate may result in depleted nutrient states in surface water (Brusewitz 1984). Conversely, aluminum has been added to a nutrient-rich lake in Sweden with some success in an effort to arrest the "aging process" caused by an overabundance of phosphate (Jernelov 1971).

Aluminum salt coagulants are used in the treatment of potable drinking water, and unretained aluminum (approximately 11% of the added aluminum) was found to be transported through a water distribution system (Driscoll and Letterman 1988).

Aluminum, as a constituent of soil, weathered rock, and solid waste from industrial processes, is transported through the atmosphere as windblown particulate matter and is deposited onto land and water by wet and dry deposition. Atmospheric loading rates of aluminum to Lake Michigan were estimated at 5 million kg/year (Eisenreich 1980). In this study, most of the aluminum was generally associated with large particles that were deposited near their source. In a study, the wet and dry deposition of aluminum was measured biweekly for 1 year at two sites on Massachusetts Bay, Turro and Nahant. The average total deposition rate was 0.1 g/m²-year, of which 29% was in rain (wet deposition) (Golomb et al. 1997).

Plant species and cultivars of the same species differ considerably in their ability to take up and translocate aluminum to above-ground parts (Kabata-Pendias and Pendias 1984). Tea leaves may contain very high concentrations of aluminum, >5,000 mg/kg in old leaves (Dong et al. 1999). Other plants that may contain high levels of aluminum include Lycopodium (Lycopodiaceae), a few ferns, Symplocos (Symplocaceae), and Orites (Proteaceae) (Jansen et al. 2002). Aluminum is often taken up and concentrated in root tissue (Kabata-Pendias and Pendias 1984). In sub-alpine ecosystems, the large root biomass of the Douglas fir, *Abies amabilis*, takes up aluminum and immobilizes it, preventing large accumulation in above-ground tissue (Vogt et al. 1987). It is unclear to what extent aluminum is taken up into root food crops and leafy vegetables. An uptake factor (concentration of aluminum in the plant/concentration of aluminum in soil) of 0.004 for leafy vegetables and 0.00065 for fruits and tubers has been reported (DOE 1984), but the pH and plant species from which these uptake factors were derived are unclear. Based upon these values, however, it is clear that aluminum is not taken up in plants from soil, but is instead biodiluted.

Transfer coefficients of 0.0002 (kg/day)⁻¹ for uptake into milk and 0.0015 (kg/day)⁻¹ for uptake into beef tissue have been reported (DOE 1984). The transfer coefficients represent the fraction of daily aluminum intake in feed that is transferred to a kilogram of milk or beef muscle. Based upon the above values,

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aluminum is not transferred to beef muscle or milk from feed to any appreciable extent and therefore would not be expected to bioaccumulate in terrestrial food chains.

The potential for accumulation of aluminum has been studied in several aquatic species including fish (Buckler et al. 1995; Cleveland et al. 1991; Hamdy 1993; McDonald et al. 1991; Wilkinson and Campbell 1993), amphibians (Freda and McDonald 1990), crustaceans (Madigosky et al. 1991), snails (Brooks et al. 1992), aquatic insects (Frick and Herrmann 1990; Guerold et al. 1995; Krantzberg and Stokes 1990), and aquatic plants (Albers and Camardese 1993; Vuori et al. 1990). Bioconcentration of aluminum in fish is a function of the water quality (e.g., pH and total organic carbon) (Cleveland et al. 1989).

Brook trout have been shown to accumulate slightly more aluminum (measured as whole-body residues) at pH 5.6–5.7 than at pH 6.5–6.6 (Cleveland et al. 1989). Cleveland et al. (1991) reported that the estimated steady-state bioconcentration factors (BCF) for aluminum in brook trout were 215, 123, and 36 at pH 5.3, 6.1, and 7.2, respectively. When transferred to water of the same pH without added aluminum, brook trout eliminated aluminum from tissues more rapidly at pH 5.3 than at pH 6.1 and 7.2. In tissues of smallmouth bass, aluminum concentrations were higher and more variable in gill tissue than in other tissues (Brumbaugh and Kane 1985). Aluminum concentrations in rainbow trout from an alum-treated lake, an untreated lake, and a hatchery were highest in gill tissue and lowest in muscle (Buerger and Soltero 1983). Aluminum residue analyses in brook trout have shown that whole-body aluminum content decreases as the fish advance from larvae to juveniles (Cleveland et al. 1989). These results imply that the aging larvae begin to decrease their rate of aluminum uptake, to eliminate aluminum at a rate that exceeds uptake, or to maintain approximately the same amount of aluminum while the body mass increases. The decline in whole-body aluminum residues in juvenile brook trout may be related to growth and dilution by edible muscle tissue that accumulated less aluminum than did the other tissues (Cleveland et al. 1989). Wilkinson and Campbell (1993) studied aluminum uptake in Atlantic salmon at a pH of 4.5 under conditions simulating spring snowmelt. These authors reported that gill uptake was slow, approaching a steady state only after 3 days of exposure. The greatest fraction of the gill-associated aluminum was not sorbed to the gill tissue, but to the gill mucus. The authors believe that the mucus appears to retard aluminum transport from solution to the membrane surface, thus delaying the acute biological response of the fish. Buckler et al. (1995) reported concentrations of aluminum in whole-body tissue of the Atlantic salmon exposed to high concentrations of aluminum ranging from 3 µg/g (for fish exposed to 33 µg/L) to 96 µg/g (for fish exposed to 264 µg/L) at pH 5.5. After 60 days of exposure, BCFs ranged from 76 to 190 and were directly related to the aluminum exposure concentration. In acidic waters (pH 4.6–5.3) with low concentrations of calcium (0.5–1.5 mg Ca/L), labile aluminum between

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25 and 75 µg/L is toxic (Rosseland et al. 1990). Because aluminum is toxic to many aquatic species, it is not bioaccumulated to a significant degree (BCF <300) in most fish and shellfish; therefore, consumption of contaminated fish does not appear to be a significant source of aluminum exposure in humans.

Aluminum uptake for the leopard frog (*Rana pipiens*) was positively correlated to exposure time and pH; however, no BCF values were reported because the authors felt that the body aluminum accumulation was too variable for useful prediction of the exposure history or physiological status of the frogs (Freda and McDonald 1990).

Bioconcentration of aluminum has also been reported for several aquatic invertebrate species. BCF values ranging from 0.13 to 0.5 in the whole-body were reported for the snail, *Helix aspersa*, fed a single 24-hour meal containing aluminum in a barley-flour pellet (Brooks et al. 1992). Madigosky et al. (1991) reported high tissue residues of aluminum in the red swamp crayfish (*Procambarus clarkii*) collected from roadside drainage ditches in Louisiana. Mean aluminum concentrations as µg/g dry weight in crayfish from roadside ditches ranged from 1.75 to 6.39 in abdominal muscle, 3.1–22.74 in the hepatopancreas, 309.4–981.50 in the alimentary tract, 10.85–77.45 in the exoskeleton, and 30–140 in the blood. These values were significantly elevated above those of control crayfish where the concentrations (µg Al/g dry weight) were 1.22 in abdominal muscle, 1.42 in the hepatopancreas, 26.97 in the alimentary tract, 4.28 in the exoskeleton, and 37.9 in the blood.

Bioconcentration of aluminum has also been reported for aquatic insects. Frick and Herrmann (1990) reported aluminum accumulation in mayfly nymphs (*Heptagenia sulphurea*) at low pH (4.5). The nymphs were exposed at two concentrations (0.2 and 2 mg Al/L) and for two exposure times (2 and 4 weeks), the longer time period including a molting phase. When nymphs were exposed to the higher concentration of aluminum for two instar periods, with a molt in between, the aluminum content (2.34 mg Al/g dry weight) nearly doubled compared with that of a one-instar treatment (1.24 mg Al/g dry weight). The major part of the aluminum was deposited in the exuviae of the nymphs, as the aluminum determination in the nymphs showed a 70% decrease in aluminum content after molting. These authors speculate that internally accumulated aluminum in the nymphs may be transferred to terrestrial predators (e.g., birds). They also hypothesized that externally deposited aluminum may be transferred to terrestrial food chains by aquatic invertebrates that leave the water in their last instar to molt on shore. An important contribution to the idea of biomagnification of aluminum was made by Nyholm (1981). Using semi-quantitative multi-element microanalysis, he related impaired breeding of pied flycatchers (*Ficedula hypoleuca*) in Sweden to the occurrence of aluminum in the bone marrow of the birds. A diet of

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stoneflies was suspected of forming a link between the lake and the terrestrial predators. Although the matter is far from clear, Nyholm (1981) seems to imply that the insects (stoneflies) were adults and that these could contain significant amounts of aluminum even after having left the exuviae behind (Frick and Herrmann 1990).

Vuori et al. (1990) sampled tufts of the aquatic moss, *Fontinalis dalecarlica*, from the River Lestijoki in Western Finland. The concentrations of aluminum in the water were low (87–196 µg/L, pH 6.5–7.0) relative to the concentrations in the young terminal shoots of *F. dalecarlica* appeared to be quite high (303–1,852 µg/g dry weight). The authors concluded that there was an effective accumulation of aluminum in the moss tissue. Albers and Camardese (1993) compared concentrations of aluminum and other metals in aquatic species of three acidified (pH≈5) and three nonacidified (pH≈6.5) constructed wetlands. They found that the metal content of *Sparganium americanum* (bur-reed) was only slightly affected by acidification.

6.3.2 Transformation and Degradation

As an element, aluminum cannot be degraded in the environment, but may undergo various precipitation or ligand exchange reactions. Aluminum in compounds has only one oxidation state (+3), and would not undergo oxidation-reduction reactions under environmental conditions. Aluminum can be complexed by various ligands present in the environment (e.g., fulvic and humic acids). The solubility of aluminum in the environment will depend on the ligands present and the pH.

6.3.2.1 Air

Aluminum-containing particulate matter in the atmosphere is mainly derived from soil and industrial processes where crustal materials (e.g., minerals) are processed. Aluminum is found as silicates, oxides, and hydroxides in these particles (Eisenreich 1980). Aluminum compounds cannot be oxidized and atmospheric transformations would not be expected to occur during transport. If aluminum metal particulates were released to air during metal processing, they would be rapidly oxidized.

6.3.2.2 Water

The trivalent aluminum ion is surrounded by six water molecules in solution (Cotton et al. 1999). The hydrated aluminum ion, $[\text{Al}(\text{H}_2\text{O})_6]^{3+}$, undergoes hydrolysis, in which a stepwise deprotonation of the coordinated water ligands forms bound hydroxide ligands (e.g., $[\text{Al}(\text{H}_2\text{O})_5(\text{OH})]^{2+}$, $[\text{Al}(\text{H}_2\text{O})_4(\text{OH})_2]^+$)

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(Snoeyink and Jenkins 1980). The speciation of aluminum in water is pH dependent. The hydrated trivalent aluminum ion is the predominant form at pH levels below 4. Between pH 5 and 6, the predominant hydrolysis products are $\text{Al}(\text{OH})^{2+}$ and $\text{Al}(\text{OH})_2^+$, while the solid $\text{Al}(\text{OH})_3$ is most prevalent between pH 5.2 and 8.8. The soluble species $\text{Al}(\text{OH})_4^-$ is the predominant species above pH 9, and is the only species present above pH 10 (Martell and Motekaitis 1989). Polymeric aluminum hydroxides appear between pH 4.7 and 10.5, and increase in size until they are transformed into colloidal particles of amorphous $\text{Al}(\text{OH})_3$, which crystallize to gibbsite in acid waters (Brusewitz 1984). Polymerization is affected by the presence of dissolved silica; when enough silica is present, aluminum is precipitated as poorly crystallized clay mineral species (Bodek et al. 1988).

Hydroxyaluminum compounds are considered amphoteric (e.g., they can act as both acids and bases in solution) (Cotton et al. 1999). Because of this property, aluminum hydroxides can act as buffers and resist pH changes within the narrow pH range of 4–5 (Brusewitz 1984).

Monomeric aluminum compounds, typified by aluminum fluoride, chloride, and sulfate, are considered reactive or labile compounds, whereas polymeric aluminum species react much more slowly in the environment (USGS 1984a). Aluminum has a stronger attraction for fluoride in an acidic environment compared to other inorganic ligands (Brusewitz 1984). Fulvic acid is also an important ligand for aluminum under acidic conditions, and it has been observed that as the temperature is lowered, the rate of complexation of aluminum with fluoride is considerably slowed, while the rate of complexation between aluminum and fulvic acid is only slightly decreased in rate (Plankey and Patterson 1987). This suggests that during snow-melt conditions, when aluminum and hydrogen ion concentrations increase, complexation with fulvic acid could preferentially occur over complexation with fluoride.

6.3.2.3 Sediment and Soil

Aluminum is present in many primary minerals. The weathering of these primary minerals over time results in the deposition of sedimentary clay minerals, such as the aluminosilicates kaolinite and montmorillonite. The weathering of soil results in the more rapid release of silicon, and aluminum precipitates as hydrated aluminum oxides such as gibbsite and boehmite, which are constituents of bauxites and laterites (Bodek et al. 1988). Aluminum is found in the soil complexed with other anions, such as fluoride, sulfate, and phosphate.

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6.4 LEVELS MONITORED OR ESTIMATED IN THE ENVIRONMENT

Reliable evaluation of the potential for human exposure to aluminum depends in part on the reliability of supporting analytical data from environmental samples and biological specimens. Concentrations of aluminum in unpolluted atmospheres and in pristine surface waters are often so low as to be near the limits of current analytical methods. In reviewing data on aluminum levels monitored or estimated in the environment, it should also be noted that the amount of chemical identified analytically is not necessarily equivalent to the amount that is bioavailable. The analytical methods available for monitoring aluminum in a variety of environmental media are detailed in Chapter 7.

6.4.1 Air

There are varying levels of aluminum in the atmosphere, depending on the location of the sampling site, meteorologic conditions, and the level of industrial activity or traffic in the area. Aluminum levels are expected to be low in areas influenced by the ocean and high in areas with wind-blown soil. Background concentrations of aluminum in the atmosphere generally range from 0.005 to 0.18 $\mu\text{g}/\text{m}^3$ (Hoffman et al. 1969; Pötzl 1970; Sorenson et al. 1974). In rural areas of Hawaii, aluminum concentrations have been measured at a range of 0.005–0.032 $\mu\text{g}/\text{m}^3$ (Hoffman et al. 1969), whereas a concentration range of 0.27–0.39 $\mu\text{g}/\text{m}^3$ has been reported in Manitoba National Park in Canada (AEC 1971). Atmospheric aluminum concentrations in U.S. cities and industrial areas are considerably higher, ranging from about 0.4 to 8.0 $\mu\text{g}/\text{m}^3$ (Cooper et al. 1979; Dzubay 1980; Kowalczyk et al. 1982; Lewis and Macias 1980; Moyers et al. 1977; Ondov et al. 1982; Pillay and Thomas 1971; Sorenson et al. 1974; Stevens et al. 1978). The range of the concentration of aluminum in fine (<1–2.5 μm) and coarse (2.5–10 μm) particles from two industrial areas, Southeast Chicago, Illinois and East St. Louis, Illinois were 22–539 ng/m^3 (125 ng/m^3 mean) and 24–1,370 ng/m^3 (153 ng/m^3 mean), respectively, for fine particles and 8.2–1760 ng/m^3 (390 ng/m^3 mean) and 17–2,120 ng/m^3 (442 ng/m^3 mean), respectively, for coarse particles. At a rural site (Bondville, Illinois), the aluminum concentrations in fine and coarse particles ranged from 32 to 293 ng/m^3 (95 ng/m^3 mean) and from 32 to 3,120 ng/m^3 (338 ng/m^3 mean), respectively which was not much different than the aluminum concentration from the industrial sites (Sweet et al. 1993). A mean aluminum concentration of 474.6 ng/m^3 (range 38.4–2,619.6 ng/m^3) was reported in particulate matter collected in air from downtown Rio de Janeiro, Brazil; samples were collected during the period from September 2001 to August 2002 (Quiterio et al. 2004). Mean aluminum concentrations in winter and summer indoor air sampled in 1999 were 41 and 39 ng/m^3 in the homes of 46 high school students from West Central Harlem, New York City who participated in the Toxic Exposure Assessment a Columbia/Harvard (TEACH) study (Kinney et al. 2002). Aluminum concentrations can also vary with

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seasonal meteorological conditions. For example, in Mackinac Island, Michigan, summer concentrations averaged about $0.25 \mu\text{g}/\text{m}^3$, while winter concentrations were only about $0.18 \mu\text{g}/\text{m}^3$ (AEC 1971).

6.4.2 Water

The concentrations of dissolved aluminum in water vary with pH and the humic-derived acid content of the water (Brusewitz 1984). Aluminum is only sparingly soluble in water between pH 6 and 8. Because the pH of about 95% of naturally-occurring water is between 6 and 9 and since high aluminum concentrations occur in surface water bodies only when the pH is <5 , the aluminum concentration in most natural waters is extremely low (Filipek et al. 1987; Snoeyink and Jenkins 1980; Sorenson et al. 1974). In general, aluminum concentrations in surface waters at pH levels above 5.5 will be $<0.1 \text{ mg/L}$ (Brusewitz 1984; Miller et al. 1984a; Sorenson et al. 1974; Taylor and Symons 1984). However, even at neutral pH levels, higher aluminum concentrations have been found in lakes with a high humic acid content (Brusewitz 1984). Aluminum concentrations in marinewaters tend to be much lower (i.e., $<0.001 \text{ mg/L}$) than those found in freshwater lakes and streams (Brusewitz 1984), probably because of increased alkalinity in marinewater compared to fresh water.

At lower pH levels, the aluminum content significantly increases because of increased solubility of aluminum oxide and salts in acidic solutions. For example, aluminum has been found at concentrations of up to 90 mg/L in tributaries that drain mines containing massive sulfide deposits (Filipek et al. 1987). In heavily contaminated surface waters in a mining region rich in sulfides, the water was highly acidic (pH <3.5) and the levels of soluble aluminum were $>2 \text{ mmol/L}$ (50 mg/L) (Alvarez et al. 1993). Similarly, surface water samples contaminated with acidic mine drainage collected at seven different locations in the vicinity of abandoned coal mines in west-central Indiana had aluminum concentrations of $6.0\text{--}269 \text{ mg/L}$ (Allen et al. 1996). The pH ranged from 2.1 to 3.4 at these sites.

Aluminum was detected at dissolved aluminum concentrations ranging from 0.001 to 2.760 mg/L with a mean concentration of 0.074 mg/L in 456 of 1,577 raw surface water samples collected during a 5-year survey at various locations across the United States (DOI 1970). Dissolved aluminum concentrations were detected in about 48% of the 380 finished drinking waters sampled and ranged from 0.003 to 1.6 mg/L with a mean of 0.179 mg/L (DOI 1970). In another survey of 186 community water systems, median aluminum concentrations for all finished drinking water samples ranged from 0.026 to 0.161 mg/L (Miller et al. 1984a). These authors further reported that the median aluminum concentration in finished water that received no coagulation treatment was 0.043 mg/L (range, $0.016\text{--}1.167 \text{ mg/L}$)

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compared to the median of 0.112 mg/L (range, 0.014–2.670 mg/L) in finished water receiving alum (aluminum sulfate) coagulation treatment. In the supplies in which no coagulant was used during treatment, 29% of supplies using surface water as their source had aluminum concentrations exceeding 0.05 mg/L, whereas only 4% of supplies using groundwater sources exceeded this level. When aluminum coagulants were used, 69% of all supplies had residual aluminum concentrations >0.05 mg/L (Miller et al. 1984a). In another study, the aluminum content in treated water at facilities using alum coagulation treatment of raw waters ranged from about 0.01 to 1.3 mg/L with a mean of about 0.157 mg/L (Letterman and Driscoll 1988). Tap water samples were collected in 1998 in the service area of East Houston, Texas water purification plant; 44% of these samples had aluminum concentrations >0.2 mg/L. Aluminum concentrations as high as 0.53 mg/L were observed in samples collected near the treatment plant that used an alum coagulant. An average decrease of 7 µg/L/km was observed along the distribution system (Cech and Montera 2000).

Schenk et al. (1989) measured aluminum concentrations in drinking water collected primarily in the western and central parts of the United States from outlets from which water was consumed rather than from the original water treatment plant. Aluminum concentrations in drinking water in various regions of the United States are listed in Table 6-3. Although aluminum concentrations in drinking water may range from undetectable to 1.029 mg/L, aluminum concentrations in most drinking water in the United States were generally <0.1 mg/L (Schenk et al. 1989). While several water sources in the west coast states (California, Oregon, and Washington) were found to contain undetectable concentrations of aluminum (<0.001 mg/L), several cities in other geographic areas of the United States had high aluminum concentrations (>0.4 mg/L). These included Peoria, Illinois (0.467 mg/L); Coos Bay, Oregon (0.483 mg/L); Watertown, South Dakota (0.502 mg/L); Waco, Texas (0.520 mg/L); Yellowstone National Park, Wyoming (0.608 mg/L); Philadelphia, Pennsylvania (0.688 mg/L); and Charleston, South Carolina (1.029 mg/L).

Henshaw et al. (1993) studied concentrations of various components, including aluminum, in drinking water derived from the Great Lakes in six communities in the United States and Ontario, Canada. Alum was used as a coagulant in all six communities. It was found that aluminum concentrations were generally higher in treated waters as compared to raw water. Between 1986 and 1990, mean aluminum concentrations in raw water were 0.020–0.053, 0.058–0.070, 0.012–0.023, 0.020–0.037, and 0.058–0.476 mg/L in Milwaukee, Wisconsin; Rochester, New York; Thunder Bay, Ontario; Toronto, Ontario; and Windsor, Ontario, respectively. Between 1986 and 1990, mean aluminum concentrations in treated water were 0.085–0.200, 0.070–0.115, 0.027–0.032, 0.080–0.139, and 0.113–0.727 mg/L in Gary,

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Table 6-3. Aluminum Concentrations Detected in Drinking Water in Various Regions of the United States

U.S. States	Aluminum concentration ($\mu\text{g/L}$) ^a
California	0–274
Colorado	42–166
Hawaii	12–124
Idaho	28–63
Illinois	3–467
Indiana	1–137
Kansas	12–245
Kentucky	9–400
Louisiana	12–210
Michigan	6–123
Minnesota	24–93
Missouri	2–368
Montana	11–98
New York ^b	254–299
Nevada	5–126
Ohio	2–245
Oregon	0–483
Pennsylvania ^c	688
South Carolina	2–1,029
South Dakota	2–502
Tennessee ^d	45
Texas	1–520
Utah	19–51
Washington	0–118
Wisconsin	12–118
Wyoming	16–608

^aRange in values reported for each state

^bWater sampled in New York City only

^cWater sampled in Philadelphia only (one sample)

^dWater sampled in Memphis only (one sample)

Source: Schenk et al. 1989

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Indiana; Rochester, New York; Thunder Bay, Ontario; Toronto, Ontario; and Windsor, Ontario, respectively. Data for raw water in Gary, Indiana and treated water in Milwaukee, Wisconsin were not provided (Henshaw et al. 1993). Aluminum concentrations in 172 samples of bottled water sold in Canada ranged from <0.010 to 0.568 $\mu\text{g/g}$ (<0.010–0.567 mg/L), with a mean of 0.027 $\mu\text{g/g}$ (0.027 mg/L) (Dabeka et al. 1992). Drinking water from 35 cities and villages in Galicia, northwest Spain were analyzed for dissolved aluminum during 1997 to 2003; an average aluminum concentration of 0.126 mg/L was reported, with concentrations ranging from 0.008 to 0.650 mg/L (Rubinos et al. 2007).

Aluminum has been measured in atmospheric precipitation (i.e., rain and snow) in the United States at concentrations up to 1.2 mg/L (Dantzman and Breland 1970; DOI 1971; Fisher et al. 1968; USGS 1964). Aluminum has been measured in rainwater samples collected during the Global Change Expedition in the North Atlantic Ocean (Lim and Jickells 1990). These authors reported that comparisons between acid-leachable and total (dissolved plus particulate) trace aluminum concentrations suggest that the acid-leachable fraction of aluminum can significantly underestimate total concentrations of aluminum in rainwater. Acid-leached mean concentrations of aluminum in rainwater collected during three rainfall events in the North Atlantic were 33.7, 12.2, and 1.99 $\mu\text{g/L}$. Overall, the acid-leached concentrations of aluminum in rainwater for seven rainfall events ranged from 1.14 to 35.2 $\mu\text{g/L}$. These values were compared with acid-leachable aluminum concentrations in precipitation from remote areas which ranged from 2.1 to 15.44 $\mu\text{g/L}$. Total (dissolved plus particulate) aluminum concentrations in North Atlantic precipitation samples collected in 1988 ranged from 6.1 to 827 $\mu\text{g/L}$ (Lim and Jickells 1990).

Aluminum concentrations in groundwater wells at neutral pH generally fall below 0.1 mg/L (Brusewitz 1984). In areas receiving acid precipitation, aluminum concentrations in groundwater may be >10 times the concentrations found in areas with neutral pH levels in the water (Brusewitz 1984), possibly due to precipitation of aluminum compounds in the more alkaline medium, or the reaction of aluminum with available silicates. In another study, Miller et al. (1984a) reported that the median concentration of aluminum in finished water obtained from groundwater was 0.031 mg/L (range, 0.014–0.290 mg/L) as compared to the median concentration in surface water of 0.043 mg/L (range, 0.016–1.167 mg/L). These authors also reported that, while 55% of the raw surface waters sampled contained aluminum concentrations >0.05 mg/L, only 4% of the raw groundwater samples contained aluminum concentrations >0.05 mg/L.

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6.4.3 Sediment and Soil

Aluminum is the most abundant metal and the third most abundant element in the earth's crust, comprising about 8.8% by weight (88 g/kg) (Staley and Haupin 1992). Its concentration in soils varies widely, ranging from about 0.07% by weight (0.7 g/kg) to over 10% by weight (100 g/kg) (Sorenson et al. 1974; USGS 1984c). Varying concentrations are found in different soil samples taken from the same area and in areas with different vegetation types (Brusewitz 1984; Sorenson et al. 1974). In Hawaii, aluminum contents were much higher with concentrations ranging from 79 to 317 g/kg (Moomaw et al. 1959). Soils in Florida and parts of Georgia, Texas, Oklahoma, and Michigan contain <20 g/kg of soil, whereas soils from portions of the Pacific Northwest, New England, Colorado, and Nevada have concentrations >80 g/kg (Sparling and Lowe 1996). Mean aluminum concentrations in cultivated and uncultivated soil samples collected during a number of field studies were 33 g/kg (range 7–>100 g/kg) for subsurface soils in the eastern United States, 54 g/kg (range 5–>100 g/kg) in subsurface soils in the western United States, and 57 g/kg (range 13–76 g/kg) in surface soils collected in Colorado (Connor and Shacklette 1975). Ma et al. (1997) reported a mean aluminum concentration of 0.730 g/kg (range 0.01–4.300 g/kg) in 40 surface soil samples from Florida. Aluminum concentrations in 1,903 soils samples collected from the United States, as well as the Virgin Islands, Guam, and Puerto Rico, were reported to range from 0.5 to 142 g/kg, with a median value of 46 g/kg (Burt et al. 2003). A median aluminum concentration of 1.8 mg/kg was reported in soils collected from 25 playgrounds located in urban Uppsala, Sweden's fourth largest city (Ljung et al. 2006).

Aluminum concentrations in soil also vary with different vegetation types. For example, aluminum concentrations in the soils of coniferous forests are often higher than in soils of beech forests since coniferous forests tend to have more acid soils (Brusewitz 1984). Alternate views of the data are that the acidic soil produced by conifers can preferentially mobilize aluminum from deeper layers toward surface soil, or that conifers over beech preferentially grow in soils rich in aluminum and it is their metabolic processes which produce more acidic soil.

Concentrations of various elements in 541 streambed-sediment samples collected from 20 study areas in the conterminous United States (1992–1996) were analyzed as part of the National Water-Quality Assessment Program of the U.S. Geological Survey. Aluminum was present in all samples; concentrations ranged from 1.4 to 14% by weight (14–140 g/kg), with a median of 6.4% by weight (64 g/kg) (Rice 1999). Mean aluminum concentrations in sediments collected in 1993 and 1994 from Lake Erie, Lake Ontario, and the Niagara River ranged from 1.339 to 13.823 g/kg dry weight (Lowe and

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Day 2002). Mean aluminum concentrations in sediments collected from three lakes in central Texas near a coal-fired power plant were 5.32, 8.16, and 8.64% in the Gibbons Creek Reservoir, Hall Lake, and Yarboro Lake, respectively (Menounou and Presley 2003). A mean aluminum concentration of 56.1 g/kg was reported in sediments from Terra Nova Bay, Antarctica (Giordano et al. 1999).

6.4.4 Other Environmental Media

Aluminum occurs naturally in many edible plants and is added to many processed foods. The concentrations in foods and beverages vary widely, depending upon the food product, the type of processing used, and the geographical areas in which food crops are grown (Brusewitz 1984; Sorenson et al. 1974). In general, the foods highest in aluminum are those that contain aluminum additives (e.g., processed cheese, grain products, and grain-based desserts) (Greger 1992; Pennington 1987; Saiyed and Yokel 2005). Because of the variability of reported concentrations of aluminum in foods, the many new manufactured food products on the market, and the increasing use of aluminum as a packaging material, a wide range of beverages and foods have been analyzed. The aluminum concentrations in a number of beverages, foods, and food products are listed in Table 6-4. Most unprocessed foods, (with the exception of some herbs and tea leaves) typically contain <5 mg/kg aluminum (Greger 1992; MAFF 1999; Pennington 1987; Schenk et al. 1989). Concentrations of aluminum in foods generally ranged from <0.15 mg/kg in eggs, apples, raw cabbage, corn, and potatoes to 695 mg/kg in American cheese (Greger 1992; MAFF 1999; Pennington 1987; Schenk et al. 1989). López et al. (2000) measured aluminum concentrations in 17 different spices and aromatic herbs widely consumed in Spain and in the Mediterranean diet; concentrations ranged from 3.74 to 56.50 mg/kg dry weight in cinnamon and oregano, respectively.

The high aluminum concentrations seen in some processed foods (e.g., processed cheeses, baked goods, and nondairy cream substitutes) are likely to have been introduced into the foods as additives, such as the anti-caking agent, sodium aluminosilicate, which is present in salt, nondairy creamers, and many other powdered materials (Table 6-4) (Saiyed and Yokel 2005; Schenk et al. 1989). The most commonly used food additives containing aluminum are: acidic sodium aluminum phosphate (leavening agent in baked goods); basic sodium aluminum phosphate (emulsifying agent in processed cheese); aluminum sulfates (acidifying agents); bentonite (materials-handling aid); aluminum color additives (dyes) from various food dyes; and aluminum silicates (anti-caking agents) (Greger 1992; Saiyed and Yokel 2005).

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Table 6-4. Estimated Aluminum Concentrations of Selected Foods

Foods	Aluminum concentration	Reference
Beverages (mg/L)		
Fruit juices (e.g., orange, reconstituted lemon, peach)	0.043–4.130	Schenk et al. 1989
Soft drinks (e.g., ginger ale, diet cola)	0.103–2.084	Schenk et al. 1989
Alcoholic beverages (e.g., beer, wine, wine coolers, champagne)	0.067–3.20	Schenk et al. 1989
Spirits (e.g., brandy, vodka, whiskey)	0.148–0.635	Schenk et al. 1989
Tea, steeped from tea bags	0.424–2.931	Schenk et al. 1989
Teas (1% extract)	0.378–3.55	Schenk et al. 1989
Herbal teas (1% extract)	0.14–1.065	Schenk et al. 1989
Instant coffee (1% solution)	0.02–0.581	Schenk et al. 1989
Whole coffee (3% extract)	0.235–1.163	Schenk et al. 1989
Beverages	1.3 ^a	MAFF 1999
Animal products (mg/kg)		
Beef, cooked ^a	0.2	Greger et al. 1985
Cheese (e.g., Swiss, cheddar, bleu)	3.83–14.1	Schenk et al. 1989
Cheese, (e.g., cottage, cheddar, Swiss)	0.12–19	Pennington 1987
Cheese, American	411–695	Pennington 1987
Cheese, processed	297	Greger et al. 1985
Chicken, with skin, cooked ^a	0.7	Greger et al. 1985
Egg, chicken	0.107	Schenk et al. 1989
Eggs, scrambled	2.865	Schenk et al. 1989
Eggs, cooked ^a	0.1	Greger et al. 1985
Eggs	0.14	MAFF 1999
Fish (cod), cooked ^a	0.4	Greger et al. 1985
Fish, salmon	5.44	Schenk et al. 1989
Fish, herring	0.127	Schenk et al. 1989
Fish	6.1	MAFF 1999
Ham, cooked ^a	1.2	Greger et al. 1985
Meat products	1.9	MAFF 1999
Milk, whole	0.06–2	Pennington 1987
Milk (skim, whole, and powdered)	0.028–7.9	Schenk et al. 1989
Milk	0.07	MAFF 1999
Poultry	0.3	MAFF 1999
Salami	1.12	Pennington 1987
Yoghurt, plain low-fat	1.12	Pennington 1987
Yoghurt, strawberry, sweetened	0.63	Pennington 1987
Fruits (mg/kg)		
Apple, fresh	0.14	Pennington 1987

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Table 6-4. Estimated Aluminum Concentrations of Selected Foods

Foods	Aluminum concentration	Reference
Banana, fresh	0.05	Pennington 1987
Grapes, fresh	1.81	Pennington 1987
Peaches, fresh	0.51	Pennington 1987
Raisins, dried	3.08	Pennington 1987
Strawberries, fresh	2.25	Pennington 1987
Fresh fruit	0.29	MAFF 1999
Fruit products	0.82	MAFF 1999
Grains (mg/kg)		
Biscuits, baking powder, refrigerated type	16.3	Pennington 1987
Bread, white	0.351	Schenk et al. 1989
Bread, white	2.33	Pennington 1987
Bread, whole wheat	2.91	Pennington 1987
Bread, pumpernickel	13.2	Schenk et al. 1989
Bread	6.6	MAFF 1999
Cereal (e.g., Post Raisin Bran®, Malt-o-Meal Wheat Cereal®)	0.040–29.33	Schenk et al. 1989
Miscellaneous cereals	5.2	MAFF 1999
Corn chips	1.23	Pennington 1987
Cornbread, homemade	400	Pennington 1987
Muffin, blueberry	128	Pennington 1987
Oatmeal, cooked	0.68	Pennington 1987
Oats	2.21–4.18	Schenk et al. 1989
Rice, cooked ^a	1.7	Greger et al. 1985
Rice, yellow, Rice-a-Roni®	1.97	Schenk et al. 1989
Spaghetti, cooked ^a	0.4	Greger et al. 1985
Vegetables and legumes (mg/kg)		
Asparagus	4.4	Greger et al. 1985
Beans, green, cooked ^a	3.4	Greger et al. 1985
Beans, navy, boiled	2.06	Pennington 1987
Cabbage, raw	0.1	Greger et al. 1985
Cauliflower, cooked ^a	0.2	Greger et al. 1985
Corn, boiled	0.1	Pennington 1987
Cucumber, fresh, pared	0.11	Pennington 1987
Green vegetables	3.1	MAFF 1999
Lettuce	0.6	Greger et al. 1985
Lettuce	0.08	Schenk et al. 1989
Peanut butter	2.0	Greger et al. 1985
Peanut butter, natural	6.29	Schenk et al. 1989
Peas, cooked ^a	1.9	Greger et al. 1985

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Table 6-4. Estimated Aluminum Concentrations of Selected Foods

Foods	Aluminum concentration	Reference
Potatoes, unpeeled, boiled ^a	0.1	Greger et al. 1985
Potatoes, unpeeled, baked	2.4	Greger et al. 1985
Potato, red	3.63	Schenk et al. 1989
Potato, sweet	1.01	Schenk et al. 1989
Potatoes	0.9	MAFF 1999
Spinach, cooked ^a	25.2	Greger et al. 1985
Tomatoes, cooked ^a	0.1	Greger et al. 1985
Other vegetables	2.7	MAFF 1999
Canned vegetables	0.97	MAFF 1999
Herbs and spices (mg/kg dry weight)		
Basil	24.80–27.30	López et al. 2000
Cinnamon	18.54–56.50	López et al. 2000
Garlic	13.60–15.25	López et al. 2000
Mustard	30.40–38.56	López et al. 2000
Nutmeg	22.81–24.80	López et al. 2000
Oregano	3.74–40.41	López et al. 2000
Pepper, black	5.79–24.41	López et al. 2000
Thyme	6.35–7.90	López et al. 2000
Other food products (mg/kg)		
Baking powder, commercial (Na Al sulfate containing)	20,000–26,000	Sorenson et al. 1974
Candy, milk chocolate	6.84	Pennington 1987
Chocolate cookie, Oreo®	12.7	Schenk et al. 1989
Cocoa	45	Greger et al. 1985
Nondairy creamer	25.7–94.3	Schenk et al. 1989
Nuts	4.0	MAFF 1999
Oils and fats	1.1	MAFF 1999
Pickles with aluminum additives	39.2 ^b	Greger et al. 1985
Pickles	0.126–9.97	Schenk et al. 1989
Salad dressing, Kraft Miracle Whip®	3.7	Schenk et al. 1989
Salt with aluminum additives	164	Greger et al. 1985
Salt	31.3–36.6	Schenk et al. 1989
Soup	0.032–3.6	Schenk et al. 1989
Sugars and preserves	2.7	MAFF 1999

^aFood not cooked or stored in aluminum pans, trays, or foil.

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Commercially available teas contain high concentrations of aluminum; 30–45% of this aluminum may be dissolved into an infusion of tea (Dong et al. 1999). Aluminum concentrations ranging from 0.2 to 9.5 mg/L have been reported in tea (Baxter et al. 1989; Flaten and Odegard 1988; Koch et al. 1989; Schenk et al. 1989; Müller et al. 1998; Pennington 1987; Pennington and Jones 1989; Kralj et al. 2005; Mehra et al. 2007). Fairweather-Tait et al. (1987) reported that approximately one-third of the aluminum in commercially available tea leaves was extracted into the tea (1.0 g tea/100 mL water); aluminum concentrations ranged from 2.7 to 4.9 mg/L in the tea after 5 minutes. Fimreite et al. (1997) reported aluminum concentrations of 4–5 mg/L in tea after 10 minutes. Schenk et al. (1989) reported that herbal teas contain lower concentrations of aluminum than ordinary tea (0.140–1.065 mg/L). Total aluminum concentrations in black, green, and red tea infusions ranging from 0.5 to 4.0 mg/L, with 10–35% of the total aluminum from an anion-exchange column as aluminum citrate. The remaining aluminum, which was strongly retained by the column, likely corresponds to aluminum species bound to phenolic compounds. Addition of lemon or milk was found to change the distribution of the aluminum species in the tea infusions (Kralj et al. 2005).

Brewed coffee (3% extract) and instant coffee (1% solution) contain aluminum concentrations of 0.235–1.163 and 0.02–0.581 mg/L, respectively (Schenk et al. 1989). Aluminum concentrations ranging from 0.1 to 0.34 mg/L have been reported in coffee (Koch et al. 1989; Müller et al. 1998). Another report provided aluminum concentration in coffee beans ranging from 11 to 21 mg/kg (Koch et al. 1989). The aluminum content of ground coffee beans has been measured at 51.8 mg/kg (Lione et al. 1984). López et al. (2000) reported aluminum concentrations in coffee ranging from 25.60 to 29.08 mg/kg dry weight. Müller et al. (1998) reported an aluminum concentration of 19 mg/kg dry weight in ground coffee. Lione et al. (1984) estimated that brewing coffee in a new aluminum pot can add from 0.88 mg (immediately after brewing) to 1.18 mg aluminum (after 12-hour storage in the pot and subsequent reheating) to each cup.

Aluminum concentrations in wines and spirits were 0.388–3.2 and 0.148–0.635 mg/L, respectively (Schenk et al. 1989). Lopez et al. (1998) reported mean aluminum concentrations in alcoholic beverages consumed in Spain; concentrations were 94.8–1,682.6, 36.5–795.2, and 15.7–739.6 µg/L in wine, beer, and other alcoholic beverages (cider, brandy, rum, whisky, gin, anisette, and liquor), respectively.

In fiscal years 1985/1986, the FDA conducted a survey of elements in fresh clams and oysters collected from U.S. coastal areas in use for shellfish production (Capar and Yess 1996). The average concentration (wet weight basis) of aluminum found in the four shellfish categories surveyed were: clams (hardshell),

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23±23 mg/kg (n=74); clams (softshell), 115±110 mg/kg (n=59); Eastern oyster, 33±26 mg/kg (n=104); and Pacific oyster, 30±28 mg/kg (n=46). Cod and bluefin tuna from the Northwest Atlantic Ocean contained an average of 1 and 0.4 mg/kg of aluminum, respectively, in muscle tissue (Hellou et al. 1992a, 1992b).

Cooking foods in aluminum pots and pans or storing foods in aluminum foil or cans may increase the aluminum content in some foods since aluminum may dissolve when in contact with a salty, acidic, or alkaline food (Abercrombie and Fowler 1997; Greger et al. 1985; King et al. 1981; Muller et al. 1993b; Nagy and Nikdel 1986). Aluminum concentrations in precooked foods (e.g., applesauce, green beans, beef, eggs, ham, pudding, rice, and tomato sauce) ranged from <0.1 to 21.6 mg/kg, while concentrations in the foods after cooking in conditioned aluminum pans and stainless steel pans ranged from 0.24 to 125 mg/kg and from <0.1 to 3.4 mg/kg, respectively (Greger et al. 1985). Acidic foods, such as tomatoes, tomato sauce, and applesauce, especially when cooked for >15 minutes, tended to accumulate more aluminum than other foods (Greger et al. 1985). Greger et al. (1985) also reported that foods cooked in new aluminum cookware had higher aluminum concentrations than foods cooked in old aluminum cookware or aluminum cookware that had been treated to simulate use. In addition, the aluminum concentrations in the foods prepared in any aluminum cookware (old, new, or treated to simulate use) had higher aluminum concentrations than the same foods cooked in stainless steel cookware. A study by Lin et al. (1997) noted that cooking with aluminum utensils may be an important aluminum exposure source for patients with chronic renal disease.

Abercrombie and Fowler (1997) reported in a small sampling of canned drinks stored at 15–20 °C, the aluminum content ranged from <0.1 to 74 mg/kg depending on the product and storage time. This study concluded that there appeared to be little basis for concern about the ingestion of aluminum when the internal protective coating of cans remains intact, the cans are stored properly, and the contents are consumed in a reasonable period of time. Fairweather-Tait et al. (1987) reported mean aluminum concentrations in Coca-Cola® and Pepsi-Cola® of 0.09 and 0.05 µg/g, respectively. Average aluminum concentrations in various beverages purchased in Australia, New Zealand, and Thailand were 0.90 and 0.15 in non-cola soft drinks in aluminum cans and glass bottles, respectively, and 0.66 and 0.24 in cola drinks in aluminum cans and glass bottles, respectively. Aluminum concentrations averaged 0.16 mg/L in beer in either aluminum cans or glass bottles (Duggan et al. 1992). Muller et al. (1993b) reported migration of aluminum from aluminum cans (unlacquered) into Coca-Cola® (pH 2.5) and diet Coca-Cola® (pH 3.0), and that the concentration of aluminum increased as the storage period increased. Concentrations of aluminum ranged from 46 to 170 µg/L in Coca-Cola® (storage for 40–101 days) and

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from 14 to 250 $\mu\text{g/L}$ in diet Coca-Cola[®] (storage for 44–173 days), respectively. Vela et al. (1998) examined the change in aluminum concentration in beer packaged in aluminum cans over time. Two brands of beer stored at 5 °C showed little change in aluminum concentration over 5 months. However, when stored at 23 °C, the concentrations increased from 50.0 to 546.5 $\mu\text{g/L}$ and from 108.0 to 414.0 $\mu\text{g/L}$ for the two brands of beer after 5 months. Joshi et al. (2003) studied the potential for the migration of aluminum into commercial sauces packaged in aluminum pouches. The results of this study indicated that after 45 days at 22 and 50 °C samples showed only minor changes in aluminum content as compared to fresh samples.

Aluminum concentrations of 0.6–3.7 and 0.1–0.4 mg/g were reported in four different types of tobacco and two samples of cannabis, respectively (Exley et al. 2006). Various elements were determined in tobacco used in the manufacture of 12 brands of cigarettes in the United States; aluminum concentrations ranged from 0.699 to 1.2 mg/g (Iskander et al. 1986).

Aluminum compounds are also used extensively in the manufacture of cosmetics (e.g., aluminum hexahydrate in deodorants) and in medical treatments (e.g., aluminum hydroxide in antacids to control gastric hyperacidity or aluminum oxide in dental ceramic implants) (Brusewitz 1984; FDA 2002; NIH 2004; NRC 1982). Many antacids contain 300–600 mg aluminum hydroxide (approximately 104–208 mg of aluminum) per tablet/capsule/5 mL dose (Zhou and Yokel 2005). Lione (1985a) reported aluminum content/dose (single tablet or 5 mL liquid) for antacids, internal analgesics (buffered aspirins), antidiarrheals, and anti-ulcerative drugs. The aluminum content per dose (single tablet or 5 mL liquid) ranged from 35 to 208 mg for antacids, 9–52 mg for buffered aspirins, 36–1,450 mg for antidiarrheal drugs, and 207 mg for an anti-ulcerative drug. Potential daily aluminum dosage ranged from 126 to 5,000 mg for these medications (Lione 1985a). Aluminum hydroxide (1–5%) is found in car polishes and paints and aluminum chlorohydrate (>1–20%) is found in antiperspirants and deodorants (NIH 2004).

Fernandez-Lorenzo et al. (1999) reported mean aluminum concentrations of 225.9 (8–1,149), 69.0 (20–204), and 152.5 (104–201) $\mu\text{g/L}$ in infant formulas, whole cow's milk, and soy milk, respectively, in a study in Spain. Ikem et al. (2002) reported mean aluminum concentrations of 58, 92, and 150 $\mu\text{g/L}$ in milk-based powdered formulas from Nigeria, the United Kingdom, and the United States, respectively. Mean aluminum concentrations of 101 and 460 $\mu\text{g/L}$ were reported for milk-based liquid formulas from the United Kingdom and soy-based powder formulas from the United States, respectively. Daily intakes of aluminum for infants in the United States were estimated to be 97, 573, and 361 $\mu\text{g/day}$ for milk-based powder formulas, soy-based powder formulas, and hypoallergenic powder formulas from the United

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States (Ikem et al. 2002). Navarro-Blasco and Alvarez-Galindo (2003) reported aluminum concentrations in soy-based infant formulas from Spain that ranged from 313 to 3,479 $\mu\text{g/L}$, with a mean of 930 $\mu\text{g/L}$. Mean aluminum concentrations in other types of Spanish infant formula were 499, 237, 252, 292, 574, 687, and 453 $\mu\text{g/L}$ for preterm formula, non-adapted starter formula, adapted starter formula, follow-up formula, lactose-free formula, hypoallergenic formula, and infant error diet formula, respectively. Aluminum concentrations were determined in infant formulas and food in Turkey (Sipahi et al. 2006). Aluminum concentrations in cereal-, milk-, cereal plus milk-based baby food were reported to be 6.43, 8.02, and 7.43, 3.33 and 13.15 $\mu\text{g/g}$, respectively. Aluminum concentrations in starches and rice flours, traditionally used in baby foods, were also reported as 3.33 and 13.15 $\mu\text{g/g}$, respectively (Sipahi et al. 2006).

Older reports on aluminum concentrations in infant formulas are also available; however, it is not known if these values would be necessarily representative of aluminum levels in infant formulas currently on the market and available to consumers. Aluminum concentrations in cow's milk-based infant formulas generally ranged from 4 to 700 $\mu\text{g/L}$ and from 5 to 2,500 $\mu\text{g/L}$ in soy-based infant formulas (Baxter et al. 1989, 1990, 1991; Bloodworth et al. 1991; Simmer et al. 1990). Average aluminum concentrations in infant formula from Canada were 0.129, 0.217, and 0.717 $\mu\text{g/g}$ in ready-to-use, concentrated, and powder milk-based infant formulas, respectively. Aluminum concentrations in Canadian soy-based formulas were 1.98, 1.41, and 9.44 $\mu\text{g/g}$ in ready-to-use, concentrated, and powder milk-based infant formulas, respectively (Dabeka and McKenzie 1990).

The median aluminum level in breast milk collected from 12 Canadian women was reported to be 14 $\mu\text{g/L}$ (range <5–45 $\mu\text{g/L}$) (Koo et al. 1988). In an Australian study, Weintraub et al. (1986) reported human breast milk concentrations of 30 $\mu\text{g/L}$. Simmer et al. (1990) reported a mean aluminum concentration of 49 $\mu\text{g/L}$ in breast milk collected from Australian women. Hawkins et al. (1994) reported a mean breast milk aluminum concentrations of 9.2 $\mu\text{g/L}$ collected from 15 nursing mothers in the United Kingdom. In a study of Croatia women, an average aluminum concentration in breast milk was 380 $\mu\text{g/L}$, with a range of 4 to 2,670 $\mu\text{g/L}$ (Mandić et al. 1995). Fernandez-Lorenzo et al. (1999) reported mean aluminum concentrations of 23.9 $\mu\text{g/L}$ (range 7–42 $\mu\text{g/L}$) in human milk in a study in Spain. Baxter et al. (1991) reported a mean aluminum concentration of 27 $\mu\text{g/L}$ (range 3–79 $\mu\text{g/L}$) in a study in the United Kingdom.

Concentrations of aluminum in whole blood and plasma have been reported to range from 0.14 to 6.24 mg/L and from 0.13 to 0.16 mg/L , respectively (Sorenson et al. 1974). Aluminum concentrations in

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serum have been reported as 1.46 and 0.24 mg/L, using neutron activation and atomic absorption analysis, respectively (Berlyne et al. 1970). An aluminum concentration in serum of 0.037 mg/L was reported using flameless atomic absorption analysis (Fuchs et al. 1974). Versieck and Cornelis (1980) discussed the possibility of aluminum contamination in blood and plasma samples from some of these early studies. This may question the reliability of aluminum levels reported in some older reports. House (1992) reported a geometric mean aluminum concentration of 0.0267 mg/L in serum and plasma for 71 office employees who were not occupationally exposed to aluminum. Mean plasma or serum aluminum concentrations were reported from various studies ranging from 0.0016 to 0.035 mg/L (House 1992). Drablos et al. (1992) analyzed aluminum serum concentrations in 230 nonexposed workers (controls) and reported a mean aluminum serum concentration of 0.005 mg/L. Nieboer et al. (1995) reviewed 34 studies on aluminum concentrations in serum or plasma, and also reported that aluminum serum concentrations in the general population were typically <0.01 mg/L. In an investigation of workers at an open bauxite mine in Surinam, serum aluminum concentrations of 24 men working in the mine for an average of 24 years were low and not statistically different from controls (de Kom et al. 1997). Razniewska and Trzcinka-Ochocka (2003) reported mean aluminum concentrations of 0.99 and 9.75 µg/L in serum and urine, respectively, in 18 healthy subjects not using medications containing aluminum.

A mean aluminum concentration of 23.21 µg/L (range 5.98–206.93 µg/L) was reported in serum samples collected from 533 female children (6–8 years old) living in Riyadh City, Saudi Arabia (Al-Saleh and Shinwari 1996). Hawkins et al. (1994) reported plasma aluminum concentrations in infants fed various formulas and breast milk. A mean plasma aluminum concentration of 8.6 µg/L was reported in breast fed infants; mean aluminum concentrations in plasma of infants fed various formulas ranged from 9.2 to 15.2 µg/L. Mean aluminum plasma concentrations of 9.9, 8.4, and 13.4 µg/L in breastfed infants at birth, 1 month, and 3 months of age, respectively. Infants on soy-based infant formulas, containing 1,600–1,700 µg/L of aluminum, were reported to have mean aluminum plasma concentrations of 8.2–12.4, 7.6–8.5, and 10.8–12.4 µg/L at birth, 1 month, and 3 months of age, respectively (Litov et al. 1989).

Aluminum concentrations in the urine can serve as an indicator of increased exposure to aluminum because a large proportion of ingested aluminum passes quickly through the body. Drablos et al. (1992) analyzed aluminum urine concentrations in 230 nonexposed workers (controls) and reported a mean aluminum urine level of 0.005 mg/L (range, 0.001–0.037 mg/L). Nieboer et al. (1995) reviewed eight studies on aluminum concentrations in urine and reported that aluminum urine concentrations in healthy individuals typically ranged from 0.0027 to 0.0081 mg/L. In a Finnish study of aluminum in urine from 3,212 occupationally exposed workers, mostly aluminum welders, between 1993 and 1996, the average

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annual urinary aluminum level was 1.4 $\mu\text{mol/L}$ (0.038 mg/L) and the range was 1.08–2.04 $\mu\text{mol/L}$ (0.029–0.055 mg/L) (Valkonen and Aitio 1997). The samples, collected as part of a routine occupational health program, were collected after the weekend as a morning specimen. The mean urinary aluminum concentration in 44 nonexposed persons, who did not use antacid preparations, was 0.33 $\mu\text{mol/L}$ (0.0089 mg/L), and the range and standard deviation were 0.07–0.82 $\mu\text{mol/L}$ (0.002–0.022 mg/L) and 0.18 $\mu\text{mol/L}$ (0.0022 mg/L), respectively. The mean serum aluminum concentration of 21 of these nonexposed individuals was 0.06 $\mu\text{mol/L}$ (0.0016 mg/L), and the range and standard deviation were 0.02–0.13 $\mu\text{mol/L}$ (0.0005–0.0035 mg/L) and 0.03 $\mu\text{mol/L}$ (0.0008 mg/L), respectively. Drablos et al. (1992) studied aluminum concentrations in workers at an aluminum fluoride plant. Mean aluminum concentrations in urine were 0.011 mg/L (range, 0.002–0.046 mg/L) for 15 plant workers, 0.032 mg/L (range, 0.006–0.136 mg/L) for 7 foundry workers, and 0.054 mg/L (range, 0.005–0.492 mg/L) for 12 potroom workers as compared to 0.005 mg/L (range, 0.001–0.037 mg/L) for 230 unexposed controls. Mean aluminum concentrations were 5.06 and 3.74 $\mu\text{g/L}$ in blood, and 6.56 and 6.35 $\mu\text{g/L}$ in urine of 103 workers in the optoelectronic industry and 67 controls, respectively (Liao et al. 2004). Pre- and postshift average aluminum concentrations in urine ranging from 0.13 to 0.153 mg/L were reported in welders from the construction industry (Buchta et al. 2005). Aluminum concentrations in human breast tissue and breast tissue fat of 4–437 nmol/g (0.1–12 $\mu\text{g/g}$) dry weight and 3–192 nmol/g oil (0.08–5.18 $\mu\text{g/g}$ oil), respectively, have been reported (Exley et al. 2007).

Nieboer et al. (1995) reported background concentrations of aluminum in bone of 1–3 $\mu\text{g/g}$ dry weight. Background aluminum concentrations in brain tissues (primarily grey matter) of healthy individuals typically ranges from 1 to 3 $\mu\text{g/g}$ dry weight or <0.5 $\mu\text{g/g}$ wet weight (Nieboer et al. 1995). Markesbery et al. (1984) determined trace element concentrations in various human brain regions in infants through adults. Aluminum concentrations were shown to increase with increasing age. Mean aluminum concentrations in adults were 0.467 $\mu\text{g/g}$ wet weight, as compared to 0.298 $\mu\text{g/g}$ wet weight in infants. Overall aluminum concentrations ranged from ≤ 0.050 to 3.05 $\mu\text{g/g}$, with the highest mean aluminum concentrations in the globus pallidus (0.893 $\mu\text{g/g}$) and the lowest in the superior parietal lobule (0.282 $\mu\text{g/g}$).

Metal concentrations were determined in spermatozoa and seminal plasma from men working in two industrial companies, a refinery and a polyolefin factory, 40 km east of Helsinki, Finland, and from sperm bank donor candidates from Helsinki, Finland in 1994. Aluminum concentrations in the factory employees were 0.93 and 0.54 mg/kg in spermatozoa and seminal plasma, respectively, and were 2.52 and 0.87 mg/kg in spermatozoa and seminal plasma, respectively, in the donor candidates. The

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authors attributed the lower concentrations in the factory workers to good quality of occupational protection in the factories. In addition, the factory employees lived in the countryside as compared to the donor candidates, who lived in a more urban area (Hovatta et al. 1998). Mean aluminum concentrations in seminal plasma of 2,200, 1,530, and 270 $\mu\text{g/L}$ were reported in samples collected from men working in smelter, refinery, and chemical industries respectively. A mean concentration of 460 $\mu\text{g/L}$ was reported in hospital workers (control group) (Dawson et al. 2000). Mean aluminum concentrations ranged from 18.0 to 101.0 $\mu\text{g/L}$ in seminal plasma collected from 64 apparently healthy men (21–35 years of age) recruited from the University of Texas (Dawson et al. 1998). A mean aluminum concentration of 15.0 $\mu\text{g/L}$ was reported in sweat collected from the arms of 15 normal, healthy subjects while exercising (Omokhodion and Howard 1994). Sighinolfi et al. (1989) reported aluminum concentration ranging from 25 to 102 $\mu\text{g/L}$ in human saliva.

Aluminum concentrations in hair ranging from 0.1 to 36 $\mu\text{g/g}$ have been reported (Alder et al. 1976; Caroli et al. 1994). Imahori et al. (1979) measured various elements in 202 human hair samples collected from a local population in the Tokyo metropolitan area. Aluminum was detected in 95 and 99 of the male and female hair samples, respectively. Mean aluminum concentrations were 13.7 mg/kg (range <0.24–65.0 mg/kg) and 13.6 mg/kg (<1.93–67.1 mg/kg) in male and female hair samples, respectively. Kobayashi et al. (1989) reported mean hair aluminum concentrations of 3.9 and 6.2 $\mu\text{g/g}$ in patients with senile dementia of Alzheimer type and a control group, respectively. Shore and Wyatt (1983) reported aluminum concentrations of 7.5 and 6.2 ppm ($\mu\text{g/g}$) in hair from patients with Alzheimer's disease and age-matched (nondemented) controls, respectively. Elemental concentrations were determined in hair from children (6–15 years old) living in environmentally degraded districts of the East Aral Sea region (Kazakhstan and Uzbekistan). Mean aluminum concentrations were 89.5 and 113.6 mg/kg in samples collected from two regions, Kazalinsk and Zhanakorgan, respectively (Chiba et al. 2004). Wilhelm et al. (1989) reported that use of hair analysis as an indicator of systematically incorporated metals may not be reliable, since endogenous metal concentrations in hair may be masked by the uptake of metals, including aluminum, from exogenous sources.

Human albumin solutions and other biological products intended for human use may contain aluminum because aluminum compounds are used in their manufacture or as a result of contamination. In albumin products, aluminum is generally introduced as a contaminant from filters, filter aides, buffer solutions, and anticoagulants, as well as the container itself. The aluminum level in a 5% pooled human albumin solution was 0.507 $\mu\text{g/mL}$ (Progar et al. 1996).

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Metal concentrations were measured in two lichen species (*Parmelic conspersa* and *Xanthoria calcicola*) from the island of Vulcano and around Mt. Etna, Sicily. Aluminum concentrations were 14,619 and 17,964 mg/kg dry weight in lichens collected near Mt. Etna and Vulcano, respectively (Varrica et al. 2000).

Mean aluminum concentrations in the soft tissues of zebra mussels (*Dreissena polymorpha*) collected in 1993 and 1994 from Lake Erie, Lake Ontario, and the Niagara River ranged from 232 to 5,030 mg/kg dry weight (Lowe and Day 2002). Whole fish composites were analyzed for various metals as part of a survey of 167 lakes in the northeastern United States as part of the Environmental Monitoring and Assessment Program (1992–1994); a mean aluminum concentration of 8.26 mg/kg wet weight (range 0.26–114.5 mg/kg wet weight) was reported (Yearley et al. 1998). Aluminum concentrations ranged from 2 to 4 mg/kg dry weight in the livers of various seabirds collected from the northern Pacific Ocean in 1992 (Elliott 2005). Mean aluminum concentrations in the feathers of nestling black-crowned night-herons in the Chesapeake and Delaware Bays ranged from 9.18 to 78.85 mg/kg dry weight (Golden et al. 2003).

An aluminum concentration of 25,948 mg/kg was reported in house dust from residences in Ottawa, Canada (Butte and Heinzow 2002).

6.5 GENERAL POPULATION AND OCCUPATIONAL EXPOSURE

Since aluminum is ubiquitous in the environment, the general population will be exposed to aluminum by inhalation of ambient air and the ingestion of food and water. Pennington and Schoen (1995) reported average daily intakes of 8–9 and 7 mg/day for adult men and woman, respectively, based on an FDA Total Diet Study. According to the 1997 total diet study conducted by the Food Standards Agency, the average U.K. population dietary exposure to aluminum was estimated to be 3.4 mg/day (MAFF 1999). Greger (1992) estimated that most adults consume 1–10 mg aluminum per day from natural sources. Biego et al. (1998) reported a daily average intake for aluminum of 4.2 mg in a study in France. Food additives containing aluminum, including preservatives, coloring agents, anticaking agents, and leavening agents are major dietary sources of aluminum in the United States (Saiyed and Yokel 2005; Soni et al. 2001).

In a report on FDA's Total Diet Study, the foods highest in aluminum were those suspected of containing aluminum additives (e.g., processed cheese, grain products, and grain-based desserts) (Pennington 1987).

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Measured daily dietary intakes of aluminum were reported to range from 2 to 14 mg/day. The major contributors to aluminum in the diet are grain products (24–49%), dairy products (17–36%), desserts (9–26%), and beverages (5–10%) (Pennington 1987). FDA revised their Total Diet Study in 1991 to reflect current food consumption patterns and to include additional sex-age groups (Pennington and Schoen 1995). Dietary intakes ranged from 0.7 mg/day for infants to 11.5 mg/day for 14–16-year-old males. The aluminum intake of adult males ranged from 8 to 9 mg/day and that for adult females was about 7 mg/day. Dietary intakes for 2-, 6-, and 10-year-old children were 4.6, 6.5, and 6.8 mg/day, respectively. Aluminum intakes per kilogram of body weight were 0.10 mg/kg for infants, 0.35 mg/kg for 2-year-old children, and 0.30 mg/kg for 10-year-old children. The other sex age groups had aluminum intakes of 0.10–0.15 mg/kg, except for 14–16-year-old males who had an aluminum intake of 0.18 mg/kg. Principal sources for aluminum were milk and dairy products (36%), fish and crustaceans (29%), cereals (16%), and vegetables (8%).

Saiyed and Yokel (2005) reported the aluminum content in various foods in the United States with aluminum food additives. Cheese from a frozen pizza was reported to contain up to 14 mg of aluminum from basic sodium aluminum phosphate. An equivalent amount of cheese from a ready-to-eat restaurant pizza contained 0.03–0.09 mg of aluminum. Up to 1.5 mg of aluminum were found in single serving packets of nondairy creamer containing sodium aluminosilicate. Products such as baking powder, pancake and waffle mixes, and ready-to-eat pancakes contained up to 180 mg of aluminum per serving (Saiyed and Yokel 2005).

Cooking in aluminum containers often results in statistically significant, but not biologically important, increases in the aluminum content of some foods. In one study, increases in the aluminum content of foods after contact with aluminum utensils were <1 mg/kg for 47% of the food examined and <10 mg/kg for 85% of the food examined (Pennington and Schoen 1995). The migration of aluminum from cookware into food will increase with the acidity of the food and the duration of exposure. For example, red current juice was prepared by boiling berries for 3 hours in either an aluminum or stainless steel pot. The aluminum concentrations of the juice prepared in the aluminum pot was 89.1 mg/L, whereas the juice prepared in the stainless steel pot was 1.83 mg/L (Valkonen and Aitio 1997). Aluminum was also shown to migrate into fish baked on aluminum foil. Increases in aluminum concentration ranged from a factor of 2 for saithe fillets baked on aluminum foil without added ingredients (0.10–0.21 mg/kg) to a factor of about 70 for mackerel fillets grilled on aluminum foil with onion rings and mixed spices (0.07–5.04 mg/kg). The migration of aluminum into foods appeared to be dependent on factors such as temperature, duration of cooking, the composition and pH of the food, and the presence of other

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substances (e.g., organic acids and salt) (Ranau et al. 2001). A study by Lin et al. (1997) noted that cooking with aluminum utensils may be an important aluminum exposure source for patients with chronic renal disease.

The intake of aluminum in foods is low compared with the amount of aluminum consumed when taking aluminum-containing medication, such as antacids, buffered aspirins, antidiarrheal agents, and certain anti-ulcer drugs at their recommended dosages (Lione 1983, 1985a; Pennington and Schoen 1995; Soni et al. 2001; Zhou and Yokel 2005). Antacids and buffered aspirin, which are often taken in multiple daily doses for prolonged periods, contain 4–562 mg/kg of aluminum (Lione 1983; Schenk et al. 1989; Shore and Wyatt 1983). For example, according to Pennington and Schoen (1995), buffered aspirin may contain 10–20 mg of aluminum per tablet. Many antacids contain 300–600 mg aluminum hydroxide (approximately 104–208 mg of aluminum) per tablet/capsule/5 mL dose (Zhou and Yokel 2005). Other exposures to aluminum can occur through the use of cosmetics and other consumer products containing aluminum compounds (Lewis 2001; NIH 2004; O'Neil et al. 2001).

Pennington and Schoen (1995) reported average daily intakes of 8–9 and 7 mg/day for adult men and woman, respectively, based on an FDA Total Diet Study. According to the 1997 total diet study conducted by the Food Standards Agency, the average U.K. population dietary exposure to aluminum was estimated to be 3.4 mg/day (MAFF 1999). Biego et al. (1998) reported a daily average intake for aluminum of 4.2 mg in a study in France.

Lione (1985a) estimated that 126–728 and 840–5,000 mg were possible daily doses of aluminum consumed in buffered aspirins and antacids products, respectively. These doses are from 6 to almost 40 times and 42–250 times greater, respectively, than aluminum doses obtained from consumption of food. When large oral loads of aluminum (1,000–4,000 mg/day) in the form of antacids are ingested, some of this excess aluminum is absorbed, usually <1% of the intake amount in healthy individuals (Gorsky et al. 1979; Kaehny et al. 1977; Reiber et al. 1995).

Median concentrations of aluminum in drinking water not receiving coagulation treatment and that receiving coagulation treatment have been reported as 0.043 and 0.112 mg/L, respectively (Miller et al. 1984a). If the total dose of aluminum obtained from water is calculated based on an estimated consumption of 1.4 L/day, the amount of aluminum ingested would respectively be 0.06 and 0.16 mg/day or roughly 1% of the 7–9 mg/day for adults from dietary sources.

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While the intake of aluminum is mainly through the ingestion of food and drinking water, inhalation of ambient air represents a small contribution to an individual's exposure to aluminum (Browning 1969). Background concentrations of aluminum in the atmosphere generally range from 0.005 to 0.18 $\mu\text{g}/\text{m}^3$ in the United States (Hoffman et al. 1969; Sorenson et al. 1974). If the inhalation rate is taken to be 20 m^3/day , then the total amount of aluminum obtained from inhalation of 0.18 $\mu\text{g}/\text{m}^3$ would be 3.6 $\mu\text{g}/\text{day}$, suggesting that ambient air is not normally a major exposure pathway for aluminum. This is negligible compared with the estimated dietary intake for adults of 7–9 mg/day. However, the aluminum content of air in urban and industrial areas has been reported to be considerable higher, ranging from 0.4 to 8.0 $\mu\text{g}/\text{m}^3$ (Cooper et al. 1979; Dzubay 1980; Kowalczyk et al. 1982; Lewis and Macias 1980; Moyers et al. 1977; Ondov et al. 1982; Pillay and Thomas 1971; Sorenson et al. 1974; Stevens et al. 1978). If the inhalation rate is taken to be 20 m^3/day , then the total amount of aluminum inhaled would range from 8 to 160 $\mu\text{g}/\text{day}$, which is still negligible compared with the aluminum intake from dietary sources. Dusts arising from soil, especially in industrial or agricultural areas (Eisenreich 1980), and from the metal surfaces of air conditioners can contain large amounts of aluminum (Crapper McLachlan 1989), resulting in high localized concentrations and, subsequently, in higher exposures. Typically, however, for the general population, inhalation is likely to be less important as an exposure pathway than is dietary exposure to aluminum, but may represent a source of greater exposure in some urban environments.

Occupational exposure to aluminum occurs not only in the refining of the primary metal, but also in secondary industries that use aluminum products (e.g., aircraft, automotive, and metal products), and aluminum welding (Nieboer et al. 1995). Three major steps are involved in primary aluminum production. Aluminum is first extracted with caustic soda from bauxite ore, precipitated as aluminum hydroxide, and subsequently converted to aluminum oxide in a calcination process. In the second step, the oxide is dissolved in molten cryolite (Na_3AlF_6) and electrolyzed to yield the pure molten metal. The electrolytic cells are called pots and the work area is called the potroom. Casting is the final step in the process where molten aluminum is poured into ingots in the foundry. Exposure is primarily to aluminum hydroxide and oxide in the initial extraction and purification process, to aluminum oxide and aluminum fluoride in the potroom (as well as to tar-pitch volatiles including PAHs), and to partially oxidized aluminum metal fumes in the foundry (Drablos et al. 1992; IARC 1984; Nieboer et al. 1995).

Most of the studies of occupational exposure (aluminum refining and metal industry workers) to aluminum have dealt with inhalation of aluminum-containing dust particles. Rarely is a worker exposed solely to aluminum-containing dust; exposure to mixtures of aluminum with fine respirable particles or other toxic chemicals is more prevalent. For example, it had been observed that the incidence of bladder

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cancer was unusually high among aluminum reduction workers. An epidemiological study showed that volatile PAHs in coal tar pitch, however, were the actual causative agents (Theriault et al. 1984a).

Synergism among metal dusts, fine particles, toxic chemicals including PAHs, and cigarette smoke is a highly plausible cause of skin irritation and cancers appearing in workers for many industrial processes involving aluminum.

According to the National Occupational Exposure Study (NOES) conducted by NIOSH from 1981 to 1983, the industries with the largest numbers of workers potentially exposed to aluminum and aluminum compounds include: plumbing, heating, and air conditioning; masonry and other stonework; electrical work; machinery except electrical; certified air transportation equipment; electrical components; fabricated wire products; general medical and surgical hospitals; industrial buildings and warehouses; and special dies, tools, jigs, and fixtures (NIOSH 1991).

A group of 44 aluminum welders in the train body and truck trailer construction industry were monitored for aluminum exposure (Buchta et al. 2005). Median aluminum concentrations of 5.6 mg/m³ (range: 0–31.5 mg/m³) and 4.5 mg/m³ (range: 1.3–15.6 mg/m³) in respirable dust in air were reported in welding fumes in 1999 and 2001, respectively. Median aluminum concentrations in aluminum welders were 152.7 µg/L (range: 2.9–656.3 µg/L) and 145.5 µg/L (range: 5.0–656.3 µg/L) in urine in pre- and post-shift samples in 2001, respectively. Median aluminum concentrations in aluminum welders were 10.6 µg/L (range: 3.3–40.3 µg/L) and 14.3 µg/L (range: 3.8–51.0 µg/L) in plasma in pre- and post-shift samples in 2001, respectively (Buchta et al. 2005).

6.6 EXPOSURES OF CHILDREN

This section focuses on exposures from conception to maturity at 18 years in humans. Differences from adults in susceptibility to hazardous substances are discussed in Section 3.7, Children's Susceptibility.

Children are not small adults. A child's exposure may differ from an adult's exposure in many ways. Children drink more fluids, eat more food, breathe more air per kilogram of body weight, and have a larger skin surface in proportion to their body volume. A child's diet often differs from that of adults. The developing human's source of nutrition changes with age: from placental nourishment to breast milk or formula to the diet of older children who eat more of certain types of foods than adults. A child's behavior and lifestyle also influence exposure. Children crawl on the floor, put things in their mouths,

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sometimes eat inappropriate things (such as dirt or paint chips), and spend more time outdoors. Children also are closer to the ground, and they do not use the judgment of adults to avoid hazards (NRC 1993).

As with adults, exposures of children to aluminum from breathing air, drinking water, and eating food is generally low. As aluminum is part of the natural environment and found widely in soils, rocks, and foods, exposure to low levels of aluminum is unavoidable. Children are likely to ingest dirt from their unwashed hands or when playing with soils and may be exposed to aluminum in this manner. Children living in proximity to hazardous waste sites or industries that release aluminum to the environment may be exposed to higher levels of aluminum than are found in the natural environment via ingestion of aluminum contained in soil, or via inhalation of aluminum from soil that is entrained in air. While aluminum contained in dirt may be in many forms, some of these forms may be embedded in minerals not bioavailable even in the acid environment of the stomach.

When FDA revised their Total Diet Study in 1991, several sex-age groups relating to children were included (Pennington and Schoen 1995). Average dietary intakes of aluminum in children are shown in Table 6-5. Dietary intakes of aluminum for children ranged from 0.7 mg/day for infants to 11.5 mg/day for 14–16-year-old males. Aluminum intakes per kilogram of body weight for children ranged from 0.10 mg/kg for infants to 0.35 mg/kg for 2-year-old children. The major sources of aluminum in food by age-sex group are shown in Table 6-6. Processed foods containing aluminum additives such as processed cheese and grain-based products constitute the foods with the largest quantities of aluminum and the largest components of the dietary intake of children. Soy-based formula may contain high quantities of aluminum and infants on such formula would have much higher dietary intakes of aluminum than other infants. Pennington and Schoen (1995) reported that soy-based infant formula was a major contributor to aluminum for infants, contributing 0.161 mg/day.

As with adults, aluminum intake from aluminum-containing medication, such as antacids, buffered aspirins, and antidiarrheal agents would overwhelm ordinary dietary intakes (Pennington and Schoen 1995). Children may be exposed to aluminum from aluminum-containing medications, vaccinations, parenteral feeding, dialysis fluids, and treatment for hyperphosphatemia (Advenier et al. 2003; Andreoli et al. 1984; Baylor et al. 2002; Bougle et al. 1991; Bozynski et al. 1989; Chedid et al. 1991; Goyens and Brasseur 1990; Griswold et al. 1983; Klein et al. 1989; Koo et al. 1986, 1992; Malakoff 2000; Milliner et al. 1987; Moreno et al. 1994; Naylor et al. 1999; Offit and Jew 2003; Randall 1983; Robinson et al. 1987; Salusky et al. 1990; von Stockhausen et al. 1990; Warady et al. 1986). Advenier et al. (2003) reported a mean aluminum concentration of 1.6 $\mu\text{mol/L}$ (0.043 mg/L) in parenteral nutrition solutions, resulting

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Table 6-5. Dietary Intakes of Aluminum in Children

Age-sex group	Aluminum intake	
	(mg/day)	(mg/kg)
6–11-Months	0.7	0.10
2-Years	4.6	0.35
6-Years	6.5	0.30
10-Years	6.8	0.11
14–16-Years (females)	7.7	0.15
14–16-Years (males)	11.5	0.18

Source: Pennington and Schoen 1995

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Table 6-6. Major Sources of Aluminum in Food by Age-Sex Group

Foods by age-sex group (Al/day)	Aluminum/day	
	mg	Percent of total intake
6–11-month-old infants (0.7 mg)		
Soy-based formula	0.161	23.0
American processed cheese	0.122	17.4
Yellow cake with icing	0.088	12.6
Green beans, strained	0.038	5.4
Pancakes	0.029	4.1
Total	0.438	62.6
2-year-old children (4.6 mg)		
Cornbread	1.580	34.3
American processed cheese	1.037	22.5
Yellow cake with icing	0.384	8.3
Fish sticks	0.173	5.4
Pancakes	0.113	2.5
Tortillas	0.093	2.0
Muffins	0.093	2.0
Fruit drink from powder	0.079	1.7
Taco/tostada	0.071	1.5
Tea	0.061	1.3
Total	3.684	80.1
6-year-old children (6.5 mg)		
American processed cheese	1.382	21.3
Yellow cake with icing	1.091	16.8
Pancakes	0.752	11.6
Fish sticks	0.529	8.1
Cornbread	0.450	6.9
Tortillas	0.297	4.6
Taco/tostada	0.209	3.2
Muffins	0.202	3.1
Hamburger	0.104	1.6
Fruit drink from powder	0.105	1.6
Total	5.121	78.8
10-year-old children (6.8 mg)		
American processed cheese	1.498	22.0
Cornbread	1.105	16.3
Pancakes	0.858	12.6
Tortillas	0.344	5.1
Yellow cake with icing	0.350	5.1
Fish sticks	0.280	4.1
Taco/tostada	0.259	3.8

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Table 6-6. Major Sources of Aluminum in Food by Age-Sex Group

Foods by age-sex group (Al/day)	Aluminum/day	
	mg	Percent of total intake
Muffins	0.207	3.0
Chocolate cake with icing	0.141	2.1
Chocolate snack cake	0.144	2.1
Total	5.186	76.3
14–16-year-old females (7.7 mg)		
American processed cheese	2.139	27.8
Yellow cake with icing	0.906	11.8
Cornbread	0.781	10.1
Taco/tostada	0.682	8.9
Pancakes	0.668	8.7
Tortillas	0.325	4.2
Muffins	0.219	2.8
Cheeseburger	0.183	2.4
Tea	0.159	2.1
Fish sticks	0.125	1.6
Total	6.187	80.4
14–16-year-old males (11.5 mg)		
Cornbread	4.209	36.6
American processed cheese	1.978	17.2
Pancakes	1.038	9.0
Yellow cake with icing	0.925	8.0
Taco/tostada	0.398	3.5
Tortillas	0.398	3.5
Cheeseburger	0.310	2.7
Tea	0.225	2.0
Hamburger	0.211	1.8
Fish sticks	0.170	1.5
Total	9.862	85.8

Source: Pennington and Schoen 1995

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in a mean aluminum daily intake of 0.08 $\mu\text{mol/kg/day}$ (0.002 mg/kg/day). An upper limit of 0.90 $\mu\text{g/L}$ for aluminum in all large-volume parenteral solutions used in total parenteral nutrition therapy was set by the FDA (Advenier et al. 2003). Aluminum compounds such as aluminum hydroxide, aluminum phosphate, or aluminum sulfate (alum) are commonly used as an adjuvant in many vaccines licensed by the FDA; the amount of aluminum in vaccines is limited to no more than 0.85 mg/dose (Baylor et al. 2002).

Elevated levels of aluminum may be found in the tissues and fluids of children undergoing treatments, such as parenteral feeding or dialysis, or if they are receiving aluminum-containing medications (Advenier et al. 2003; Andreoli 1990; Andreoli et al. 1984; Bougle et al. 1991; Bozynski et al. 1989; Chedid et al. 1991; Goyens and Basseur 1990; Griswold et al. 1983; Klein et al. 1989; Koo et al. 1986, 1992; Milliner et al. 1987; Moreno et al. 1994; Naylor et al. 1999; Robinson et al. 1987; Roodhooft et al. 1987; Salusky et al. 1986, 1990; von Stockhausen et al. 1990); however, these levels are atypical of the general population.

6.7 POPULATIONS WITH POTENTIALLY HIGH EXPOSURES

In addition to individuals who are occupationally exposed to aluminum (see Section 6.5), there are several groups within the general population that have potentially higher exposures (higher than background) than the general population. These populations include members of the general population living in the vicinity of industrial emission sources and hazardous waste sites, individuals with chronic kidney failure requiring long-term hemodialysis treatment, infants fed a formula diet containing high levels of aluminum, and individuals consuming large quantities of antacid formulations for gastric disorders, anti-ulcerative medications, buffered analgesics for arthritis, or antidiarrheal medications. Furthermore, the elderly are at risk because of multiple chronic diseases including ulcers and other gastrointestinal diseases, rheumatoid arthritis, and renal disorders. Aluminum has been detected in virtually all food products (especially plant-derived and processed foods), ambient air, drinking water, and soils. Substantially higher concentrations of aluminum have been detected in localized areas around some industrial and hazardous waste disposal sites.

Individuals living or working in proximity to aluminum production facilities may be exposed to higher concentrations of aluminum in the ambient air than members of the general population. In addition, individuals living in proximity to hazardous waste sites may be exposed to aluminum via ingestion of aluminum contained in soil from their unwashed hands when working or playing with contaminated soils

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and sediments. Children in particular are likely to ingest dirt from their unwashed hands, or inhale resuspended dust during near-ground activities. If residential wells are the primary source of drinking water, this may also pose a risk to human health via consumption of contaminated drinking water.

Individuals with chronic renal failure requiring long-term hemodialysis treatment are another group within the general population that may be exposed to greater than background levels of aluminum (Alfrey 1987; Chappuis et al. 1988, 1989; Chedid et al. 1991; Fernández-Martin et al. 1998; Griswold et al. 1983; Lione 1985a; Marumo et al. 1984; Muller et al. 1993b; Salusky et al. 1990; Winterberg et al. 1987). Elevated levels of aluminum may be found in the tissues and fluids of individuals undergoing treatments, such as hemodialysis, or if they are receiving aluminum-containing medications (Chappuis et al. 1988, 1989; Chedid et al. 1991; Griswold et al. 1983; Marumo et al. 1984; Salusky et al. 1990; Tsukamoto et al. 1979; Winterberg et al. 1987); however, these levels are atypical of the general population. Aluminum levels in virtually every body tissue are significantly higher in this group of patients if aluminum is present in the dialysate (Alfrey et al. 1980; Cooke and Gould 1991). In addition, Main and Ward (1992) reported a 10-fold increased serum aluminum concentration in a hemodialysis patient after she was prescribed effervescent analgesic tablets containing citrate. This patient was already taking aluminum hydroxide capsules. Once the effervescent analgesic tablets were discontinued, the patient's serum aluminum levels fell to acceptable levels within 3 weeks. Since citrate appeared to enhance aluminum absorption, these authors stated that patients with renal failure taking aluminum compounds should not be prescribed citrate-containing preparations. In a study by Fernández-Martin et al. (1998), a decrease in serum aluminum concentrations in patients on hemodialysis over the past 10 years was observed, from 61.8 µg/L in 1988 to 25.7 µg/L in 1996. These reductions have been achieved due to the restriction of the use of oral aluminum hydroxide, as well as to the use of adequate water treatment systems.

The oral intake of aluminum tends to be higher for children than for adults (Greger 1992). Calculations based on the FDA's Total Diet Study suggest that 2-year-old children (13 kg body weight) consumed almost 3 times as much aluminum per kg body weight as adult males (75 kg body weight) or adult females (60 kg body weight), respectively (0.48 versus 0.18 and 0.15 mg aluminum/kg body weight, respectively) (Greger 1992). Infants fed milk-based or soy-based infant formulas can be exposed to higher concentrations of aluminum than infants fed breast milk or cows' milk (see Section 6.4.4). Within this group, the infants believed to be most at risk would be preterm infants with impaired renal function because they would be less able to excrete the absorbed aluminum (Bishop 1992; Greger 1992; Koo et al. 1988, 1992; Weintraub et al. 1986).

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As discussed in Section 6.4.4, individuals consuming large quantities of antacid formulations, anti-ulcerative medications, buffered analgesics, or antidiarrheal medications are exposed to higher than background doses of aluminum in their diet. Lione (1985a) estimated that 126–728 and 840–5,000 mg were possible daily doses of aluminum consumed in buffered aspirins for rheumatoid arthritis and antacid products, respectively. These doses are 6–40 and 42–250 times greater, respectively, than aluminum doses obtained from consumption of foods (3.4–9 mg/day) (Biego et al. 1998; MAFF 1999; Pennington and Schoen 1995).

6.8 ADEQUACY OF THE DATABASE

Section 104(i)(5) of CERCLA, as amended, directs the Administrator of ATSDR (in consultation with the Administrator of EPA and agencies and programs of the Public Health Service) to assess whether adequate information on the health effects of aluminum is available. Where adequate information is not available, ATSDR, in conjunction with NTP, is required to assure the initiation of a program of research designed to determine the health effects (and techniques for developing methods to determine such health effects) of aluminum.

The following categories of possible data needs have been identified by a joint team of scientists from ATSDR, NTP, and EPA. They are defined as substance-specific informational needs that if met would reduce the uncertainties of human health assessment. This definition should not be interpreted to mean that all data needs discussed in this section must be filled. In the future, the identified data needs will be evaluated and prioritized, and a substance-specific research agenda will be proposed.

6.8.1 Identification of Data Needs

Physical and Chemical Properties. The physical and chemical properties of aluminum and various aluminum-containing compounds are sufficiently well defined to allow an assessment of the environmental fate of these compounds (HSDB 2008; Lewis 2001; Lide 2005; O'Neil et al. 2001). No additional data are needed at this time.

Production, Import/Export, Use, Release, and Disposal. According to the Emergency Planning and Community Right-to-Know Act of 1986, 42 U.S.C. Section 11023, industries are required to submit substance release and off-site transfer information to the EPA. The TRI, which contains this information for 2005, became available in May of 2007. This database is updated yearly and should provide a list of industrial production facilities and emissions.

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Because aluminum compounds occur naturally (Browning 1969; Dinman 1983; IARC 1984; NRC 1982) and are widely used in industry, in the manufacture of household products, and in processing, packaging, and preserving food (Browning 1969; Lewis 2001; O'Neil et al. 2001; Stokinger 1981; Venugopal and Luckey 1978), the potential for human exposure to these compounds through ingestion of food and water and inhalation of airborne particulates is substantial. Recent data on production, import/export, and use are available (Lewis 2001; O'Neil et al. 2001; USGS 2007a, 2007c). Information on disposal of aluminum compounds is limited. In the United States, about 3 million metric tons of aluminum was recovered from purchased scrap in 2005 (USGS 2007b, 2007c). TRI data are available for releases of aluminum, as fume or dust and as aluminum oxide (fibrous forms) (TRI05 2007). Additional information on disposal would be useful in assessing the potential for the release of and exposure to aluminum compounds.

Environmental Fate. Aluminum partitions to air, water, soil, and plant material. As an element, aluminum cannot be degraded in the environment; it can undergo various precipitation or ligand exchange reactions in the environment. Its partitioning to various media is determined by the physical and chemical properties of the aluminum compound and the characteristics of the environmental matrix that affects its solubility (Brusewitz 1984; Dahlgren and Ugolini 1989; Filipek et al. 1987; Goenaga and Williams 1988; James and Riha 1989; Litaor 1987; Mulder et al. 1989; Wangen and Jones 1984). Aluminum is transported through the atmosphere primarily as a constituent of soil and other particulate matter (Eisenreich 1980). Transformations are not expected to occur during transport of aluminum through the atmosphere. Aluminum partitions between solid and liquid phases by reacting and complexing with water molecules, anions, and negatively charged functional groups on humic materials and clay (Bodek et al. 1988). Information on the environmental fate of aluminum is sufficient to permit a general understanding of transport and transformation in all environmental media. No additional information is needed at this time.

Bioavailability from Environmental Media. Aluminum compounds are deposited in the lungs following inhalation (Christie et al. 1963; Steinhagen et al. 1978; Stone et al. 1979; Thomson et al. 1986) and are poorly absorbed following ingestion (Hohl et al. 1994; Priest et al. 1998; Stauber et al. 1999; Steinhausen et al. 2004). A fractional absorption of 1.5–2% was estimated based on the relationship between urinary aluminum excretion and the airborne soluble aluminum to which workers were exposed (Yokel and McNamara 2001). Very limited information is available regarding absorption following dermal contact; however, this pathway of exposure is not expected to be significant. Additional

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information on absorption following ingestion of soils contaminated with aluminum compounds and dermal contact would be useful in assessing bioavailability following exposure via these routes, particularly at hazardous waste sites.

Food Chain Bioaccumulation. Little information is available on the uptake of aluminum into food crops. Uptake into root crops is of particular importance, since many plant species concentrate aluminum in their roots (DOE 1984; Kabata-Pendias and Pendias 1984; Vogt et al. 1987). The limited information available on bioconcentration in animals appears to indicate that aluminum is not significantly taken up by livestock (DOE 1984). The fact that in studies dealing with aluminum in food, aluminum is generally present in low concentrations in fruit, vegetables, and meat products that do not contain aluminum additives or have other contact with aluminum (e.g., cooked in aluminum pots) (Greger et al. 1985; MAFF 1999; Pennington 1987; Pennington and Schoen 1995; Schenk et al. 1989; Sorenson et al. 1974), would support a conclusion that aluminum does not bioaccumulate in the food chain. Because of its toxicity to many aquatic species, aluminum does not bioconcentrate appreciably in fish and shellfish and therefore, it would not be a significant component of the diet of animals that feed upon them (Rosseland et al. 1990). Further studies on the uptake of aluminum by plants, especially those grown on acid soils, would be useful in expanding a limited database and characterizing the importance of food chain bioaccumulation of aluminum as a source of exposure for particular population groups.

Exposure Levels in Environmental Media. Reliable monitoring data for the levels of aluminum in contaminated media at hazardous waste sites are needed so that the information obtained on levels of aluminum in the environment can be used in combination with the known body burden of aluminum to assess the potential risk of adverse health effects in populations living in the vicinity of hazardous waste sites.

Estimates of human exposure to aluminum from food (Biego et al. 1998; Greger 1992; MAFF 1999; Pennington 1987; Pennington and Schoen 1995; Saiyed and Yokel 2005; Schenk et al. 1989; Sorenson et al. 1974), drinking water (Cech and Montera 2000; DOI 1970; Letterman and Driscoll 1988; Miller et al. 1984a; Schenk et al. 1989), and air (Browning 1969; Crapper McLachlan 1989; Sorenson et al. 1974) are available, as are estimates from exposure from antacids, buffered analgesics, antidiarrheal and anti-ulcerative compounds (Lione 1983, 1985a; Schenk et al. 1989; Shore and Wyatt 1983; Zhou and Yokel 2005). Information on the intake of aluminum from vitamins and other dietary supplements is lacking and would be useful in estimating human exposure. Additional information on the occurrence of

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aluminum in the atmosphere, surface water, groundwater, and soils surrounding hazardous waste sites would be helpful in updating estimates of human intake.

Exposure Levels in Humans. Measurements of the aluminum content in human tissues, especially in blood (Berlyne et al. 1970; de Kom et al. 1997; Drablos et al. 1992; Fuchs et al. 1974; House 1992; Liao et al. 2004; Nieboer et al. 1995; Razniewska and Trzcinka-Ochocka 2003; Sorenson et al. 1974), urine (Buchta et al. 2005; Drablos et al. 1992; Liao et al. 2004; Nieboer et al. 1995; Razniewska and Trzcinka-Ochocka 2003; Valkonen and Aitio 1997), and breast milk (Baxter et al. 1991; Fernandez-Lorenzo et al. 1999; Hawkins et al. 1994; Koo et al. 1988; Mandić et al. 1995; Simmer et al. 1990; Weintraub et al. 1986), are available. However, Versieck and Cornelis (1980) discussed the possibility of aluminum contamination in blood and plasma samples from some of early studies. This may question the reliability of aluminum levels reported in some older reports.

Measurements of aluminum in other human tissues and fluids, such as bone, brain, saliva, spermatozoa, and seminal fluid are also available (Dawson et al. 1998, 2000; Hovatta et al. 1998; Markesbery et al. 1984; Nieboer et al. 1995; Sighinolfi et al. 1989). However, recent biological monitoring data, particularly for aluminum in blood and urine, are limited. More recent information would be useful in assessing current exposure levels. Additional biological monitoring data for populations surrounding hazardous waste sites would be useful in helping to better characterize human exposure levels.

This information is necessary for assessing the need to conduct health studies on these populations.

Exposures of Children. Measurements of the aluminum content in tissues, blood, and urine of children who have been exposed to aluminum, as well as unexposed children, are limited. Chiba et al. (2004) reported aluminum concentrations in hair of children. Al-Saleh and Shinwari (1996) reported aluminum concentrations in serum samples of girls aged 6–8 years. Hawkins et al. (1994) and Litov et al. (1989) reported plasma aluminum concentrations in infants fed various formulas and breast milk. Studies measuring aluminum concentrations in tissues, blood, and urine of specialized groups of children (e.g., infants with renal failure or on parenteral nutrition) have also been reported (Advenier et al. 2003; Andreoli 1990; Andreoli et al. 1984; Bougle et al. 1991; Bozynski et al. 1989; Chedid et al. 1991; Goyens and Brasseur 1990; Griswold et al. 1983; Klein et al. 1989; Koo et al. 1986, 1992; Milliner et al. 1987; Moreno et al. 1994; Naylor et al. 1999; Robinson et al. 1987; Roodhooft et al. 1987; Salusky et al. 1986, 1990; von Stockhausen et al. 1990).

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Additional information monitoring aluminum concentrations in children would be useful in assessing both the normal aluminum content of children and the effect of exposure on aluminum concentrations in children. This information would also be useful in assessing differences in the effect of aluminum exposure on children to that of adults. While the largest source of aluminum exposure in adults is from aluminum-containing medications and cosmetics, we do not know the amount of such products that may be given to children. Additional information on the intake of available aluminum from soil during childhood activities, or the placental transfer to fetal blood, especially among pregnant women taking antacids as a result of abdominal upsets, would be useful in assessing exposure levels in children.

Data are available on the intake of aluminum in food eaten by children and from their diet (Dabeka and McKenzie 1990; Koo et al. 1988; Pennington and Schoen 1995; Pennington 1987; Simmer et al. 1990; Weintraub et al. 1986). Aluminum concentrations in human breast milk, infant formula, and cow's milk have been reported. The aluminum content of human breast milk generally ranged from 9.2 to 49 µg/L, lower than that reported in infant formulas (Fernandez-Lorenzo et al. 1999; Hawkins et al. 1994; Koo et al. 1988; Simmer et al. 1990; Weintraub et al. 1986). Soy-based infant formulas contain higher concentrations of aluminum, as compared to milk-based infant formulas or breast milk. Recent reports provide average aluminum concentrations ranging from 460 to 930 µg/L for soy-based infant formulas and from 58 to 150 µg/L for milk-based formulas (Fernandez-Lorenzo et al. 1999; Ikem et al. 2002; Navarro-Blasco and Alvarez-Galindo 2003). Infant formulas are much higher in aluminum than human breast milk. Daily intakes of aluminum for infants in the United States were estimated to be 97, 573, and 361 µg/day from milk-based powder formulas, soy-based powder formulas, and hypoallergenic powder formulas, respectively (Ikem et al. 2002).

Child health data needs relating to susceptibility are discussed in Section 3.12.2, Identification of Data Needs: Children's Susceptibility.

Exposure Registries. No exposure registries for aluminum were located. This substance is not currently one of the compounds for which a sub-registry has been established in the National Exposure Registry. The substance will be considered in the future when chemical selection is made for sub-registries to be established. The information that is amassed in the National Exposure Registry facilitates the epidemiological research needed to assess adverse health outcomes that may be related to exposure to this substance.

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6.8.2 Ongoing Studies

The Federal Research in Progress (FEDRIP 2006) database provides additional information obtainable from a few ongoing studies that may fill in some of the data needs identified in Section 6.8.1. These studies are summarized in Table 6-7.

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Table 6-7. Ongoing Studies on Aluminum

Investigator	Affiliation	Research description	Sponsor
Longnecker, M	Not provided	This research proposes to study elemental concentrations in toenails, which may provide a good measure of exposure for various elements, including aluminum.	NIH
Yokel, RA	University of Kentucky, Lexington, Kentucky	The overall objective of the proposed research is to test the null hypothesis that the bioavailability of aluminum is comparable from foods and from drinking water.	NIH

NIH = National Institutes of Health

Source: FEDRIP 2006