

4.0 Polychlorinated Biphenyls

Polychlorinated biphenyls (PCBs) consist of a mixture of chlorinated biphenyls that contain a variable number of substituted chlorine atoms on the aromatic rings. There are 209 possible congeners, all of which have varying physical and chemical properties as well as differing degrees of toxicity (Hutzinger et al., 1974). PCBs are a probable carcinogen and have been shown to produce developmental and reproductive effects in experimental animals. Because PCBs are lipophilic, they tend to bioaccumulate in lipids and biomagnify up through the food chain. In general, PCBs are resistant to biodegradation and the more highly chlorinated congeners are more persistent in the environment than the less-chlorinated congeners.

4.1 Natural Sources

PCBs do not occur naturally in the environment (Eisler, 1986b).

4.2 Anthropogenic Sources

Due to their low flammability, PCBs were manufactured for use in insulating and cooling equipment such as electrical transformers and large capacitors. PCBs were also used in vacuum pumps, electromagnets, lubricants, plastics, paints, inks, and carbonless copy paper. The Monsanto Company, the sole manufacturer of PCBs in the United States, produced 630,000 tonnes of PCBs (NAS, 1979) until The Toxic Substances Control Act of 1978 banned the manufacture of most PCBs in 1979. Of the 630,000 tonnes produced by Monsanto, 67,500 tonnes were exported from the United States. Today, PCBs continue to be used in existing equipment that was in operation before the ban, especially in capacitors and transformers.

PCBs are no longer manufactured, but are still prevalent in some equipment. They can be released into the environment from items that contain PCBs and are still in use, or from the storage and disposal of these contaminated items. Additionally, PCBs can still be generated inadvertently via chemical processes.

PCBs were never manufactured in the Lake Superior basin. Their presence in the lake may be attributed in large part to deposition from the atmosphere (Jeremiason et al., 1994). Concentrations of PCBs in sediments and fish tissue have decreased since production of PCBs ceased in 1979. Concentrations in the atmosphere have remained constant for about the last 10 years, suggesting that evaporation from contaminated soils and lakes continues to provide PCBs to the atmosphere. The flux of PCBs from land to air tends to be greater in urban areas or in areas having PCB-contaminated soils (Panshin and Hites, 1994). For example, in northern Wisconsin the atmospheric PCB flux is 1-3 mg/(m² yr) (Swackhammer and Armstrong, 1986). The flux in Bloomington, Indiana which is surrounded by PCB-contaminated Superfund sites, was estimated to be between 16 to 160 mg/(m² yr) (Panshin and Hites, 1994).

Management efforts have focused on reducing the stocks of PCBs in storage and use. The goal of this report is to develop an estimate of potential releases of PCBs to the U.S. Lake Superior basin by calculating PCB-contaminated equipment storage in the adjacent three states and estimating potential PCB releases from the use of PCB-contaminated equipment and from accidental spills.

4.2.1 Electrical Equipment Use

PCBs were used as electric current insulating material and as a coolant in electrical equipment due to their fire-resistant and insulating properties. According to Toxic Substances Control Act (TSCA) regulations, electrical equipment containing PCBs may still be used if the equipment is "totally enclosed" such that no leaking will occur (i.e., in transformers, capacitors, electromagnets, switches, voltage regulators, etc.). Additionally, activities specifically authorized based on the finding that they will not present unreasonable risk and those which petition and receive an exemption may also use PCB-contaminated equipment.

A survey of three industrial categories in Michigan gave some indication of the way in which PCBs are being used and stored at plant facilities (Spitzley, 1989). The survey covered natural gas production plants, motor vehicle/car body plants, and nonferrous foundries. The amount of PCB equipment in use was related to electricity use; i.e., facilities using large amounts of electricity tended to have more PCB-contaminated equipment. Fifty-three percent of the facilities surveyed used PCB-contaminated transformers. Thirty-three percent of industries surveyed reported using transformers having greater than 500 ppm PCBs, and 21 percent reported using PCB capacitors. In addition, several facilities reported having PCB-contaminated soils on site (20 percent of natural gas and 8.6 percent of car body manufacturers).

The Michigan survey indicated wide disparity among industries in terms of their progress toward phasing out PCB electrical equipment (Table 25). For example, nearly 90 percent of capacitors in use in the motor vehicle industry are filled with PCBs versus 57 percent of the capacitors in the natural gas industry. The average level of PCB use among the industries surveyed in Michigan (around 75 percent of capacitors and transformers) was significantly greater than that in the utilities industry, which has made exemplary progress in phasing out PCBs (USEPA, 1994b). For example, only about 9 percent of transformers from Great Lakes Utilities had transformers having 50 to 500 ppm PCB mineral oil

and about 0.74 percent had transformers with greater than 500 ppm PCBs.

Both the utilities and industry sectors represent possible sources for PCB contamination. These sectors will be discussed in the following sections.

Utilities. It has been suggested that 87 percent of the original stock of PCBs has been removed from service by the utilities industry since 1979 (Lingle and Wilson, 1993). If this were true for industries across the board, there would be 73,125 tonnes of PCB remaining in the United States ($0.13 \cdot 562,500$ tonnes of PCB produced and not exported from the United States). A population extrapolation of the entire United States (pop. = 248,710,000) to the U.S. Lake Superior basin (pop. = 425,548) would result in an estimate of 125 tonnes of PCBs remaining today in U.S. Lake Superior basin utilities.

Large Utilities

By far the most complete information on PCB use in the U.S. Lake Superior basin covers the large utilities in the adjacent states. The Great Lakes Utility Report, in which this information is summarized, was prepared for U.S. EPA Region 5 by the utility industry in response to EPA's voluntary PCB phasedown program. The information was compiled by 12 major electric utilities (known as the Great Lakes Utilities or GLU) in Region 5 that worked together to gather extensive data on electrical equipment currently in service.

Table 25. Results of PCB Use Survey in Michigan Involving Four Industrial Sectors Showing Relative Progress in Phasedown.

Industrial category	Total number of capacitors	Number of PCB capacitors	Percent PCB capacitors	Total number of transformers	Number of transformers > 500 ppm PCB	Percent transformers > 500 ppm PCB	Number of transformers 50-500 ppm PCB	Percent transformers 50-500 ppm PCB
Natural gas ^a	809	463	57.2%	13,255	4	0.03%	10,283	77.6%
Nonferrous minerals ^a	582	444	76.3%	164	45	27.4%	24	15.2%
Motor vehicle/acrbody ^a	4509	4053	89.9%	870	445	51.1%	29	3.3%
G.L. utilities ^a (large)	700,000 ^c	83,000	11.9%	4.87 x 10 ⁶	35,884	0.74%	435,000	8.9%

^a From Spitzley (1989).

^b From Lingle and Wilson (1993).

^c From USEPA (1994b).

Three of the major electric utilities—Minnesota Power, Northern States Power, and Wisconsin Electric Power—operate in the U.S. Lake Superior basin.

- Minnesota Power submitted a report to U.S. EPA Region 5 on PCB use and management (MP, 1994). The report covered a 26,000-square-mile area of northeastern Minnesota including the Minnesota portion of the Lake Superior drainage basin and data from Superior Light & Power, a subsidiary of Minnesota Power serving the community of Superior, Wisconsin.

The majority of PCB-contaminated equipment in service today with Minnesota Power is located in substations where the risk of environmental contamination due to failure or accident is much less than for equipment on distribution lines. The PCB-related electrical equipment at the substations fall into two categories: (1) PCB substation capacitors and (2) PCB-contaminated substation equipment. PCB capacitors contain 100 percent PCB and make up the bulk of PCB mass in use. Other electrical equipment in substations can contain PCB-contaminated oil. All equipment containing greater than 500 ppm PCB has been replaced or retrofilled. Minnesota Power inventories all PCB-contaminated equipment at substations, and distribution transformers are tested for PCBs when they are taken in for service. Although transformers may contain PCB-contaminated oil, no current inventory has been performed due to the prohibitive cost of sampling. In addition, PCB transformers in distribution lines and PCB-contaminated equipment indoors in hospitals, schools, office buildings, and power plants have all been replaced.

Minnesota Power published its PCB capacitor and transformer decommissioning schedule in the Minnesota Power Report (MP, 1994). The schedule to decommission and retrofit equipment in the Lake Superior basin is in Tables 26 and 27.

Table 26. PCB Capacitor Decommissioning Schedule at Minnesota Power.^a

Year	Number	Quantity of PCB fluid (gallons)	Mass (kg)

			@9.85 lb/gal density ^b	@13.50 lb/gal density ^b
1994	877	2,390	10,594	14,519
1995	733	1,956	8,670	11,883
1996	582	1,412	6,259	8,578
1997	210	560	2,482	3,402
1998	22	52	230	316

^a MP (1994).

^b Hutzinger et al. (1974).

Table 27. PCB Contaminated Equipment Retrofit Schedule at Minnesota Power.^a

Year	Capacity (gallons)	Mass (kg)
1993	56,018	41.1
1994	52,719	38.6
1995	48,799	37.7
1996	15,872	7.6
1997	7,794	3.7
1998	2,474	1.8

^a MP (1994).

- Northern States Power, Eau Claire, Wisconsin is the second participating Great Lakes Utility serving the U.S. Lake Superior basin. The Wisconsin branch of Northern States Power has removed all known network and vault PCB transformers, all known PCB capacitors, and all network and plant transformers and substation equipment having > 500 ppm PCBs. Northern States Power's Wisconsin Division has phased out all of its PCB transformers and capacitors. A total of 71,655 kg was removed since 1978 (Tina Ball, Northern States Power Company, personal communication, July 5, 1995). The only PCB items remaining in Northern States Power's Wisconsin Division are two large-capacity storage tanks containing PCB-contaminated oil and 36 surge arrestors at hydro plants. The storage tanks, containing a total of 29,830 kg PCBs, are located at the Wheaton Plant, which is not in the U.S. Lake Superior basin.
- Wisconsin Electric Power Company, Presque Isle, Michigan, is the third Great Lakes Utility in the U.S. Lake Superior basin. Information to estimate its PCB use was obtained from the Michigan Critical Materials Register annual wastewater report (Christopher Hull, Michigan Department of Natural Resources, personal communication, June 12, 1995). Wisconsin Electric Power Company reported having 89,000 lb of PCBs on site in 1991. This same amount (all known quantities), was reported as disposed of as hazardous waste that same year.

Small Utilities

Small utilities and rural electric cooperatives are not represented by the GLU. Small utilities might still be using PCB capacitors since they possibly have fewer resources to invest in phasedown programs. A state-wide search of the 1991 Michigan Critical Materials Register annual wastewater report data base (last year for which complete information is available) identified some small electric utilities using PCBs (Table 28). Based on the number of customers, PCB use by small utilities in Michigan can be extrapolated to the U.S. Lake Superior basin. No small utilities were identified in the Critical Materials Register Wastewater Report for Michigan's Lake Superior basin. According to the Great Lakes Utilities report (Lingle and Wilson, 1993), about 16 percent of the population of USEPA's Region 5, which includes the Lake Superior basin, is served by municipal systems or rural electric cooperatives. This fraction applied to the U.S. Lake Superior basin translates into 68,100 customers serviced by small utilities. A population extrapolation, based on the Michigan data using a ratio of kg PCB in use to customers serviced multiplied by the population serviced in the Lake Superior basin (68,100), estimates a total of 15,985 kg PCBs in use in the U.S. Lake Superior basin.

Industry. Information to estimate the use of PCBs by industries came from the Michigan Critical Materials Register annual wastewater report for 1991 (Christopher Hull, Michigan Department of Natural Resources, personal communication, June 12, 1995). Four industries in the U.S. Lake Superior basin, including two mining and two forest products facilities, use PCB-contaminated equipment. These industries, located in Michigan, are required to report their wastewater discharges, residuals for disposal, and use of toxic pollutants, including toxics that contaminate equipment and process materials (Table 29). Some of these industries also reported removing some PCBs from their sites as hazardous waste. Michigan industries in the Lake Superior basin were using roughly 213,582 kg (using the midpoint of the range) of PCBs at the end of 1991. Based on a population extrapolation from Michigan (pop. = 142,606; Allardice and Thorp, 1994), to the rest of the U.S. Lake Superior basin (pop. = 425,548), it is estimated that there are 637,346 kg PCBs in use by industries in the U.S. Lake Superior basin.

A summary of PCB use estimates for utilities and industries is presented in Table 30.

Table 28. 1991 Estimated PCB Use in Michigan Municipal Systems and Rural Electric Cooperatives

Small utility name	Population served	Original quantity in use at start of in 1991 (kg)	Quantity disposed of in 1991 (kg)	Quantity remaining in use (kg)
Fruit Belt Electric Cooperative	22,311	815	0.0	2815
Lansing Board of Water and Light	86,237	37,353	3,402	33,951
Cloverland Electric Coop.	13,884	None Reported	None Reported	None Reported
Hillsdale Board of Public Utilities	5,333	None Reported	None Reported	None Reported
Thumb Electric Cooperative	9,991	None Reported	None Reported	None Reported
Western Michigan Electric Coop.	10,352	None Reported	None Reported	None Reported
TOTAL	148,108	38,168	3,402	14,766

4.2.2 Additional Sources of PCBs

PCBs can be released through discarded PCB-contaminated equipment residing in landfills, or via spills.

Landfills. Several landfills, including municipal, industrial, and demolition landfills, are located in the U.S. Lake Superior basin (LSBP, 1995b). It is unknown at this time whether PCB-contaminated equipment is contained within any of these landfills, although some Superfund sites in Minnesota's portion of the basin are contaminated with PCBs (LSBP, 1995b).

Table 29. 1991 Estimated PCB Use (in kg) for Michigan Industries.

Industry	Original quantity in use January 1991	Quantity disposed of in 1991	Quantity remaining in use
Copper Range	173,952	0	173,952
Cleveland Cliffs Iron Co.	5,334	1,447	3,887
Kimberly Clark Corp.	46-227	0	46-227
Stone Container	39,916	4,309	25,607
TOTAL	219,248-219,429	5,756	213,492-213,673

Table 30. Summary of Estimated PCB Use Data for Utilities and Industries in the U.S. Lake Superior Basin

Source/use category	In use (kg)
Large Utilities	
Capacitors	10,594 ^a
Transformers	38.6 ^a
Small Utilities	15,985 ^b
Industries	637,346 ^c
TOTAL	663,964

^a Data from the 1994 schedule for Minnesota Power (Tables 26 and 27).

^b Data based on Michigan small utilities information of 34,766 kg PCB/148,108 persons served · 68,100 persons in the U.S. Lake Superior basin estimated serviced by small utilities

^c Data based on Michigan industries information of 213,582 kg PCB/142,606 population of Michigan · 425,548 persons in the U.S. Lake Superior basin.

Spills. Most spills (92 percent) are from transformers, half of which do not contain PCBs, and 3.7 percent of spills are from large capacitors in substations. About 1.7 percent of all spills involve waterways, of which 80 percent of those reported are cleaned up within one day with a 50 percent cleanup efficiency (Lingle and Wilson, 1993). Assuming 0.4 percent per year of PCBs are spilled (number for capacitor spills from Alliance technologies, as cited in Thompson, 1994), and a total of 663,964 kg PCBs in use, 2,656 kg of PCBs are spilled each year in the U.S.

Lake Superior basin. If 99.7 percent of the amount spilled from capacitors is cleaned up (clean-up efficiency factor for oil spills, Lingle and Wilson, 1993), the amount remaining in the ground is roughly 8.0 kg/yr. A quantity of 7.2 mg/kg of the amount spilled was reported to become an atmospheric emission (Alliance Technologies, 1987, as cited by Thompson, 1994). If 1.7 percent of all spills involve waterways and there is a 50 percent clean-up efficiency, an estimated 22.6 kg/yr of PCBs are released from spills to waterways. From these emission factors a total of approximately 31 kg might spill irrecoverably in the U.S. Lake Superior basin (Table 31).

4.3 Total PCB Releases

A total of 31 kg/yr of PCBs were estimated to be released via spills and 663,964 kg of PCBs were estimated to be in use in U.S. Lake Superior basin in 1990 (Table 32).

Table 31. Estimated PCB Emissions in the U.S. Lake Superior Basin.

Estimated inventory	Estimated spills (kg/yr)	Estimated emissions (kg/yr)			
		Land	Air	Water ^a	Total
663,964	2,656	8.0	0.02	22.6	30.6

^a Water = Spills · 0.017 (fraction involving waterways) · 0.5 (clean-up efficiency).

Table 32. Summary of Estimated PCB Uses and Releases to the U.S. Lake Superior Basin

Source/use category	Use (kg)	Emissions (kg/yr)
Large Utilities		
Capacitors	10,594	NA ^a
Transformers	38.6	NA
Small Utilities	15,985 ^b	NA
Industries	637,346	NA
Spills		
Land	-	8.0
Air	-	0.02
Water	-	22.6
TOTAL	663,964	30.6

^a NA = Not applicable. It is assumed that the capacitors and transformers in use are contained in closed environments, and will not release unless a spill occurs.

4.4 Uncertainties in PCB Estimations

The largest area of uncertainty with PCB estimations is that there might be other sources of PCB-contaminated equipment not reported in the basin. Other equipment could be in use and/or stored or discarded, thereby having the potential to release PCBs to the environment. Given this uncertainty, PCBs releases to the U.S. Lake Superior basin could be underestimated. Another area of uncertainty is the estimate of PCBs remaining in Lake Superior industries. This estimate is based on the Michigan Critical Materials Register database. If Lake Superior basin industries are different from the state of Michigan as a whole, PCBs in the basin might be over- or underestimated.

5.0 Hexachlorobenzene

Hexachlorobenzene (HCB) is a compound that is formed as a by-product during the manufacture of chlorinated solvents and other chlorine-containing compounds, including pesticides. HCB was used as a pesticide (fungicide) until 1985, and it has been used in the production of synthetic rubber and military ordnance materials. Currently, there are no commercial uses for HCB. HCB has a low water solubility and resists degradation, and thus is persistent in the environment and partitions to soils and sediments. Human exposure to HCB can occur through contact with contaminated soils, dust particles, or industrial releases into the environment. HCB can bioaccumulate in the environment and biomagnify up through aquatic and terrestrial food chains. In laboratory animals exposed orally to HCB, developmental defects as well as adverse effects on the immune system and the liver were found; HCB is a suspected human carcinogen (Klaassen et al., 1986; USEPA, 1986, 1988; ATSDR, 1990).

5.1 Natural Sources

Hexachlorobenzene does not occur naturally in the environment.

5.2 Anthropogenic Sources

Hexachlorobenzene is not intentionally produced in the United States or Canada. Nevertheless, it continues to be released to the environment by several pathways:

- As a by-product of several industrial processes (i.e., pulp and paper mills, paint manufacturers, coal and steel products, mining, and wood preservation facilities).
- During fuel combustion (from coal, wood, and diesel fuel).
- During incineration (from medical and hazardous waste and sewage sludge).
- From contaminated sites and landfills.
- From use of HCB-contaminated pesticides (including chlorothalonil (5% HCB), dimethyltetrachloroterephthalate (3% HCB), and picloram (2% HCB) (ATSDR, 1990)).
- From use of HCB-contaminated commercial products (chlorinated solvents, including pentachlorophenol).

5.2.1 Wastewater Discharges

There is little information on the release of HCB to the waterways in the U.S. portion of the Lake Superior basin. Possible sources of discharge include pulp and paper mills, chemical manufacturers and other industrial plants, and sewage treatment facilities. No reliable data exist to indicate that HCB is generated and/or emitted from pulp and paper mills (Cohen et al., 1995). HCB was not detected in any of the pulp and paper mill effluents located in the U.S. Lake Superior basin. Additionally, there are no identified sources for industrial discharges, and data are limited and unreliable for WWTP discharges (Cohen et al., 1995; LSBP, 1995a, 1995b).

5.2.2 Atmospheric Emissions

HCB can be contributed to the atmosphere from mining, fuel combustion, and incineration.

Mining. The mining industry potentially releases HCB in copper smelting and iron sintering. Emission factors for copper smelting (Cohen et al., 1995) were used to estimate emissions from Copper Range (Table 33). Emissions estimates are unknown for the taconite pelletizing process.

Fuel Combustion. HCB is released in the combustion of coal, wood, gasoline, and diesel fuel. Emission factors and throughputs of HCB into the Lake Superior basin from various types of fuel combustion are shown in Table 34 (Cohen et al., 1995).

Incineration. Several types of incinerators are known to emit HCB, including:

- Municipal solid waste
- Sewage sludge
- Medical waste
- HCB waste
- Hazardous waste
- Cement kilns
- Small incinerators

Emission factors and throughput for the major incinerators in the Lake Superior basin were available from Cohen et al. (1995). Of the 11 municipal wastewater treatment facilities in the U.S. Lake Superior basin, only Western Lake Superior Sanitation District (WLSSD) in Duluth, Minnesota, incinerates both municipal waste and sewage sludge. Medical waste incinerators are present in the region. There were no known HCB waste incinerators, hazardous waste incinerators, or cement kilns in the Lake Superior basin. No emission factors were identified for small incinerators, which could release substantial quantities of HCB. Calculated emission estimates for WLSSD are shown in Table 35. HCB releases from medical waste incinerators were extrapolated to the U.S. Lake Superior basin based on population (Table 36).

Table 33. Estimated HCB Emissions from Mining Operations in the U.S. Lake Superior Basin in 1990.

Mining operation	Production rate (tons/day)	Production rate ^a (tonnes/day)	Production rate (g/yr)	Emission factor ^b (g HCB/g production)	Air Emissions (kg/yr)
Copper Range ^c	146	132.5	4.8 x 10	3.9 x 10 ⁻⁸	1.9 ^d

Talconite pelletizing	-	-	-	unknown	unknown
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^a From LaMP Stage II (LSBP, 1995a).

^b From Cohen et al. (1995).

^c The Copper Range smelter closed in September 1995.

^d $132.5 \text{ tonnes/day} \cdot 365 \text{ days/yr} \cdot 1,000 \text{ kg/tonne} \cdot 1,000\text{g/kg} \cdot 3.9 \times 10^{-8} \text{ g HCB/g production} \cdot \text{kg}/1,000\text{g}$

Table 34. Estimated HCB Emission from Fuel Combustion in the U.S. Lake Superior Basin.

Fuel type	Emission factor ^a (A)	Throughput ^b (B)			Throughput in the U.S. Lake Superior basin ^c (C)	Estimated air emissions in U.S. Lake Superior basin (kg/yr) ^d (D)
		MN	MI	WI		
Coal	$1.6 \times 10^{-11} \text{ g HCB/g fuel}$	$1.9 \times 10^9 \text{ g fuel/hr}$	$3.6 \times 10^9 \text{ g fuel/hr}$	$2.1 \times 10^9 \text{ g fuel/hr}$	$5.8 \times 10^7 \text{ g fuel/hr}$	8.1×10^{-3}
Wood	$6.0 \times 10^{-11} \text{ g HCB/g fuel}$	$7.8 \times 10^8 \text{ g fuel/hr}$	$7.6 \times 10^8 \text{ g fuel/hr}$	$3.0 \times 10^8 \text{ g fuel/hr}$	$1.40 \times 10^7 \text{ g fuel/hr}$	7.4×10^{-3}
Diesel fuel	$2.1 \times 10^{-8} \text{ g HCB/veh-km}$	$4.2 \times 10^9 \text{ veh-km/yr}$	$5.3 \times 10^9 \text{ veh-km/yr}$	$4.5 \times 10^9 \text{ veh-km/yr}$	$1.1 \times 10^8 \text{ veh-km/yr}$	2.3×10^{-3}
Leaded gasoline	$8.7 \times 10^{-10} \text{ g HCB/veh-km}$	$3.4 \times 10^9 \text{ veh-km/yr}$	$7.1 \times 10^9 \text{ veh-km/yr}$	$3.5 \times 10^9 \text{ veh-km/yr}$	$1.1 \times 10^8 \text{ veh-km/yr}$	9.6×10^{-5}
Unleaded gasoline	$2.4 \times 10^{-11} \text{ g HCB/veh-km}$	$6.4 \times 10^{10} \text{ veh-km/yr}$	$1.3 \times 10^{11} \text{ veh-km/yr}$	$6.6 \times 10^{10} \text{ veh-km/yr}$	$2.0 \times 10^9 \text{ veh-km/yr}$	4.8×10^{-5}
TOTAL						1.8×10^{-2}

^a From Cohen et al. (1995), Table I.

^b From Cohen et al. (1995), Appendix.

^c Based on 1990 U.S. census population of MN, MI, and WI of 18,562,711 persons. $C = (B \text{ (average of MN+MI+WI throughputs)})/18,562,711 \cdot \text{Population of U.S. Lake Superior basin (425,548)}$ (Allardice and Thorp, 1994).

^d For coal and wood: $D = A \cdot C \cdot 24 \text{ hr/day} \cdot 365 \text{ day/yr} \cdot \text{kg}/1,000\text{g}$.

For mobile sources: $D = A \cdot C \cdot \text{kg}/1,000\text{g}$.

Table 35. Estimated HCB Emissions from Municipal Incinerators in the U.S. Lake Superior Basin.

Incinerator	Type	Throughput ^a (g/hr) (A)	Emission factor (g HCB/g throughput) ^b (B)	Air emissions to U.S. Lake Superior basin (kg/yr) ^c (C)
WLSSD	Municipal solid waste refuse-derived fuel	7.3×10^6	1.0×10^{-8}	0.64
WLSSD	Sewage sludge	3.0×10^5	5.0×10^{-7}	1.3
TOTAL				1.9

^a Cohen et al. (1995), Appendix.

^b Cohen et al. (1995), Table I.

^c For municipal solid waste and sewage sludge; $C = A \cdot 24 \text{ hr/day} \cdot 365 \text{ day/yr} \cdot B \cdot \text{kg}/1,000\text{g}$.

Table 26. Estimated HCB Emissions from Medical Waste Incinerators in the U.S. Lake Superior Basin.

Type	Emission factor ^a (g HCB/g throughput) (A)	Average throughput in U.S. Lake Superior basin (g/hr) ^b (B)	Throughput in U.S. Lake Superior basin (g/hr) ^c (C)	Air emissions to U.S. Lake Superior basin (kg/yr) ^d (D)
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Medical waste	1.9×10^{-8}	3.4×10^7	7.8×10^5	0.13
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^a From Cohen et al. (1995), Table I.

^b From Cohen et al. (1995), Appendix; throughputs for MI = 1.3×10^7 , MN = 9.6×10^6 , WI = 1.1×10^7 g/hr.

^c Based on 1990 U.S. census population of MN, MI, and WI of 18,562,711 persons. $C = (B \text{ (average of throughputs MN+MI+WI)}/18,562,711) \cdot \text{Population of U.S. Lake Superior basin (425,548)}$.

^d $D = C \cdot A \cdot 24 \text{ hr/day} \cdot 365 \text{ d/yr} \cdot \text{kg}/1,000\text{g}$.

Dry Cleaning. The solvent perchloroethylene (PCE) is the most frequently used dry cleaning chemical. Dry cleaning accounts for 74 percent of its total use (Thompson, 1994). Perchloroethylene contains trace impurities of HCB at a level of about 5.0×10^{-6} kg/L (Thompson, 1994). About 70 percent of PCE used is lost to the atmosphere through volatilization, and the remaining 30 percent is disposed of off site and "recovered" or mixed with combustible hazardous waste and burned in incinerators and cement kilns, forming ultra-toxic organochlorine by-products (Cantin, 1992). CRA (1993, cited in Rice and Weinberg, 1994, p. 36) reported that the U.S. dry cleaning industry used 1.3×10^8 kg of PCE in 1990; thus, approximately 9.1×10^7 kg/yr of PCE was potentially released to the atmosphere in the United States.

Based on the U.S. 1990 Census, the atmospheric contribution of HCB to the U.S. portion of the Lake Superior basin from dry cleaning was estimated using the following equation:

$9.1 \times 10^7 \text{ kg PCE for U.S./yr} \cdot 1,000 \text{ g/kg} \cdot 1 \text{ mL}/1.6227 \text{ g (density of PCE)} \cdot 1 \text{ L}/1,000\text{mL} \cdot 5.0 \times 10^{-6} \text{ kg HCB/L PCE} \cdot 425,548 \text{ (population in basin)}$, 248,709,873 (1990 U.S. population)

$= 4.8 \times 10^{-1} \text{ kg/yr from dry cleaning}$

5.2.3 Other Hexachlorobenzene Uses and Releases

HCB is also present in pesticides and pentachlorophenol, which are used in the U.S. basin in agriculture and industries.

Pesticides. Use of contaminated pesticides has been implicated as the largest source of human exposure to HCB (ATSDR, 1990). HCB also can be inadvertently produced during the manufacture of several pesticides. Pesticides known or suspected to contain HCB are dimethyl tetrachloroterephthalate, chlorothalonil, pentachloronitrobenzene (PCNB), pentachlorophenol, pichloram, atrazine, and simazine (IARC, 1986). An emission factor of 7.5×10^{-1} g HCB/g HCB-contaminated pesticide and estimated application of pesticides to farms in Michigan, Minnesota, and Wisconsin were used in this analysis (Cohen et al., 1995) (Table 37). Approximately 0.07 kg/yr of HCB is applied as a contaminant of pesticides each year in the U.S. Lake Superior basin. This estimate is based on the assumption that the pesticides are applied for one hour per day for every person in the basin for 180 days per year.

Table 37. Estimated Application of HCB-Contaminated Pesticides in the U.S. Lake Superior Basin.

State	Emission factor ^a (g HCB/g contaminated pesticide used) (A)	Statewide throughput ^b (g pesticides applied/hour) (B)	Percent farmland in Lake Superior counties in 1987 ^c (C)	Superior basin throughput ^d (g/hr) (D)	Potential HCB released ^e (kg/yr) (E)
MI	0.75	14	2.2	0.31	0.04
MN	0.75	7	1.2	0.08	0.01
WI	0.75	6.3	1.5	0.09	0.01
				TOTAL=	0.07

^a From Cohen et al. (1995), Table I.

^b From Cohen et al. (1995), Appendix.

^c From County and City Data Books: 1988 and 1994.

^d $B \cdot C/100 = D$.

^e $D = C \cdot 1 \text{ hr/day} \cdot 180 \text{ day/yr} \cdot A \cdot \text{kg}/1,000 \text{ g} = E$.

Sewage Sludge. HCB may occur in municipal sewage sludge due to the use of PCP to treat foreign textiles (Horstmann and McLachlan, 1994). PCP is estimated to contain up to 200 mg HCB/kg and 11.5 mg TEQ dioxins/kg (Thompson, 1994). These estimates were used to convert the 0.04 g TEQ dioxins/yr found in sewage sludge in the U.S. portion of the basin to provide an estimate of the amount of HCB that might be present in the sludge:

$0.04 \text{ g TEQ dioxins/yr} \cdot 1 \times 10^6 \text{ mg/g} \cdot \frac{200 \text{ mg HCB/kg}}{11.5 \text{ mg TEQ dioxins/kg PCP}} \cdot \text{kg}/1 \times 10^6 \text{ mg}$

Sewage sludge was estimated to contain 0.7 kg HCB/yr in the U.S. Lake Superior basin. This HCB might volatilize or otherwise be released to air or water, but the possible pathways have not been examined. WLSSD was estimated to contribute 1.3 kg HCB/yr to the atmosphere by burning of sewage sludge (Table 35), using emission factors and throughput from Cohen et al. (1995). Since the contribution from municipal sewage sludge might be doubly counted in this situation, the 0.7 kg HCB/yr was not included in calculating the total.

Forest Products. The Koppers wood preserving plant in Superior, Wisconsin, does not use PCP but does have PCP-contaminated soils on site. The quantity of PCP in these soils is estimated to be 250 kg (see Section 3.2.3). Using the proportion of HCB estimated to be found in PCP (200 mg/kg), 50 g of HCB is estimated to be present in the soils at the Koppers site, but releases of this HCB to Lake Superior through volatilization or runoff are unknown. Additional soil monitoring under way in 1996 will provide a better indication of HCB distribution at this site.

5.3 Total Hexachlorobenzene Releases

A total of approximately 4.7 kg/yr of HCB is estimated to be released or disposed of from industrial, incineration, fuel combustion, municipal and residential, and consumer product sources (Table 38).

Table 38. Summary of Estimated HCB Releases in the U.S. Lake Superior Basin.

Source/Use Category	Emissions and Discharges (kg/yr)			Use, Disposal, Soils (kg/yr)
	Water	Air	Total Releases	
INDUSTRIAL				
Forest products	-	-	-	-
Other products	-	-	-	-
Mining	-	1.9 ^a	1.9	-
Wood preserving	-	-	-	-
<i>INDUSTRIAL TOTAL</i>	-	1.9	1.9	
INCINERATION				
Class III, medical waste	-	0.13	0.13	-
Class IV, small incinerators	-	-	-	-
WLSSD	-	1.9	1.9	-
<i>INCINERATION TOTAL</i>	-	2.0	2.0	-
FUEL COMBUSTION				
Coal	-	8.1 x 10 ⁻³	8.1 x 10 ⁻³	-
Wood	-	7.4 x 10 ⁻³	7.4 x 10 ⁻³	-
Diesel Fuel	-	2.3 x 10 ⁻³	2.3 x 10 ⁻³	-
Leaded gasoline	-	9.6 x 10 ⁻⁵	9.6 x 10 ⁻⁵	-
Unleaded gasoline	-	4.8 x 10 ⁻⁵	4.8 x 10 ⁻⁵	-
<i>FUEL COMBUSTION TOTAL</i>	-	1.8 x 10 ⁻²	1.8 x 10 ⁻²	-
MUNICIPAL/RESIDENTIAL				
Landfills	-	-	-	-
Wastewater treatment plants	-	-	-	-
Dry cleaners	-	0.48	0.48	0.21
<i>MUNICIPAL/RESIDENTIAL TOTAL</i>	-	0.48	0.48	0.21
COMMERCIAL PRODUCTS				
Pentachlorophenol use	-	-	-	0.7 DC ^b
Chlorinated solvent use	-	-	-	-
Pesticide use	-	-	-	0.07
<i>COMMERCIAL PRODUCTS TOTAL</i>	-	-	-	0.07
TOTAL	-	4	4.4	0.3

^a Copper Range smelter closed in September 1995.

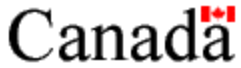
^b DC = double count and was not included in the total.

5.4 Uncertainties in Hexachlorobenzene Estimations

The major concerns in HCB estimations were identification of sources and accuracy of the emission factors and other values used in the calculations. Source identification for HCB is difficult because of the potential contamination in other organochlorine chemicals that might be used in the basin but could not be tracked down for this report. In addition, virtually nothing is known about contributions from small incinerators, manufacturing processes, and landfills in the U.S. portion of the Lake Superior basin.

For pesticides, no data were found on quantities of pesticides used per acre or what kinds of pesticides were applied. Data on nonfarm uses of pesticides (e.g., spraying of ditches, silviculture) were also not located.

Emission factors from Cohen et al. (1995) were based on U.S. EPA emission factors or were estimated using complex models of transport and measured emissions data from specific sources extrapolated to all facilities based on estimated annual throughput, i.e., the amount of waste burned by a type of incinerator, the quantity discharged in effluent, or the number of vehicle-kilometers traveled by diesel trucks. Cohen et al. (1995) noted that few data were available for existing individual facilities in each source class and that those data indicate that emission factors can vary greatly between facilities and within a particular facility over time. The reasons for such variations might be unknown or unreported. Comparisons of computed with actual values were generally in agreement, however. Cohen et al. (1995) also noted that the actual concentration of HCB in air over the Great Lakes was apparently the result of a mixture of globally distributed HCB and a smaller portion of HCB emitted from sites in the United States and Canada. Thus, HCB contributions to Lake Superior from atmospheric deposition could be higher than that estimated (56-95 pg/m³ measured at several sites, versus 1-27 pg/m³ estimated using the models developed by Cohen and colleagues).



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