POLYCHLORINATED BIPHENYL USAGE AND SOURCES OF LOSS TO THE ENVIRONMENT IN MICHIGAN

John L. Hesse*

Abstract

Polychlorinated biphenyls (PCB's) are used in a wide variety of applications including dielectric fluids in transformers and capacitors; plasticizers in paints, inks, plastics, and coatings; hydraulic systems; heat transfer systems; investment casting waxes; and many more. This paper describes industrial uses of PCB's in Michigan and focuses on those uses which were found to be sources of environmental contamination. It describes PCB use figures supplied by several industries and concentrations detected in industrial and municipal effluents and sludges.

INTRODUCTION

Polychlorinated biphenyls (PCB's) are complex mixtures of chlorine-substituted biphenyl compounds and have been produced commercially in the United States since 1929. Their properties of nonflammability, stability, resistance to acids, alkalis, and other caustic chemicals, and low volatility under prolonged heating have made them useful in a wide range of industrial products.

Monsanto Chemical Company, the sole U.S. manufacturer of PCB's, recently released production and sales figures through 1974 (ref. 1). These showed that production and sales roughly doubled between 1960 and 1970, with nearly 80 million pounds sold annually in the United States by 1970.

Broadhurst (ref. 2) has summarized many of the uses of PCB's. The five largest uses for PCB's prior to 1970 were dielectric fluids in capacitors, plasticizer applications, transformer fluids, hydraulic fluids and lubricants, and heat transfer fluids. The list of other uses is lengthy.

Because of the widescale usage of PCB's, they have become distributed throughout the world. Since 1967, reports of PCB residues in fish, birds, water, sediments, and other environmental samples have been common. They are of concern in the environment because of their persistence, potential for biological magnification, and chronic toxicity.

By 1971, Monsanto recognized the environmental problems which had developed because of PCB's and initiated a program to phase out sales for all open-ended uses, finally limiting sales to uses in electrical capacitors and transformers. Many officials believed this action would result in rapid reductions in environmental contamination but little evidence of this has yet been documented.

Michigan officials have been studying the PCB situation since 1969 when excessive PCB residues were detected in Great Lakes fish. After development of adequate laboratory capabilities, we instituted a monitoring program for PCB's early in 1971 consisting of a statewide water sampling survey ~~in inland~~ waters and tributaries to the Great Lakes. Supplementary sampling has since been conducted on fish from inland and Great Lakes waters, stream sediments, potable water intakes, sanitary landfill runoff, municipal wastewater treatment plant effluents, and industrial discharges. Limited questionnaire surveys of industrial usage have also been conducted. This paper will report briefly on the industrial uses of PCB's that we have identified in Michigan and focus on those uses which were found to be sources of environmental contamination.

Electrical Applications

Prior to the 1971 Monsanto limitation of sales to the so-called "closed-system" uses, approximately 60 percent of U.S. sales were for electrical capacitor and transformer applications. In 1968, annual capacitor and transformer sales totaled approximately 30 and 16.8 million pounds, respectively (ref. 3).

While losses from the use of PCB's in these applications have been estimated as minimal (ref. 3), incorrect disposal practices and accidental losses from equipment and storage areas do occur and can result in environmental contamination.

In response to an industrial questionnaire sent to a cross section of Michigan industries in 1971, one major power company in Michigan reported an annual use of 562,000 pounds of PCB's (250,000 in precipitator transformers and 312,000 in capacitors). Company records showed annual losses of approximately 300 pounds of PCB liquids to soils from burst capacitors and 12,000 pounds from salvaged capacitors. This fluid from the capacitors was reportedly disposed of through waste

*Supervisor, Toxic Material Unit, Department of Natural Resources, Lansing, Michigan.

haulers. Waste haulers then dispose of these wastes in many ways, including dust control on roads, sale of oils as fuel in low-temperature boilers, and treatment and discharge to municipal sewerage systems. Fluid recovered from precipitator transformers was returned to Monsanto for incineration. In the same industrial questionnaire survey, a smaller power company reported that it had been disposing of approximately 700 pounds of PCB's per year by mixing with other waste oils and using the oils for dust control on their driveway and parking lot.

In October 1975, a 55-gallon drum of PCB transformer oil on inventory at a Michigan power company developed a leak from a defective seam. Approximately 45 gallons of the fluid soaked into the ground, resulting in more than 100 cubic yards of contaminated soil having to be removed and disposed of in an approved landfill. This situation illustrates the lack of such proper safeguards as diking around storage areas and represents an example of environmental loss from a so-called "closed-system" use.

Most industries other than utility companies using electrical transformers containing PCB's contract either the transformer manufacturer or smaller companies for servicing and do not concern themselves with the problem of disposal of the waste fluids. Frequency of servicing appears to differ between industries, varying anywhere from every 6 months to 10 years.

While water sampling programs in Michigan have failed to identify any electrical applications of PCB's as major point sources of loss to aquatic situations, environmental losses do occur which have the potential of indirectly reaching watercourses through atmospheric fallout or leaching from contaminated soils at spill or disposal sites. Losses directly to water may occur on occasion but would be difficult to detect because of their intermittent nature.

Plasticizer Applications

Plasticizer applications represent the single largest "open-ended" or dissipative use of PCB's. PCB's are or have been used as plasticizers in most countries in a wide variety of consumer products, including paints, inks, copying paper, adhesives, sealants, plastic products, and textile coatings, many of which are traded internationally (ref. 4). Monsanto's U.S. sales figures for 1970 showed a volume of nearly 20 million pounds for plasticizer applications (ref. 1). Sales for this use were discontinued in 1971. Environmental losses from past sales will likely continue for a long period of time.

Papers used in the thermographic, xerographic, or pressure-sensitive copying processes have had PCB's added as plasticizers either in the ink or paper coating. Recycling of these papers can result in contamination of food packaging materials and other paper products (ref. 3).

Effluents from paper recycling or deinking plants also become contaminated. In Michigan, paper industry effluents have been found to commonly contain from to 10 μg/l of PCB's. Contamination of fish has been identified as high as 110 mg/kg in the Kalamazoo River downstream from Kalamazoo, Michigan, where Michigan's paper industry is primarily centered. Stream sediments in the Kalamazoo area contain as much as 36 mg/kg PCB's; this is believed to be a result of past deinking processes in the area. Lower level contamination continues from processing of recycled paper but most deinking mills have ceased operation over the past 10 years.

While Monsanto stopped sales of PCB's for paper applications in 1971, one Michigan paper company recently reported to the Michigan Department of Natural Resources that several raw products that they purchase still contain PCB's. Two coloring compounds reported have 23 and 500 mg/kg PCB's and nearly all of the wood pulp contains from 0.5 to 1.0 mg/kg.

Burning of waste papers containing PCB's, which occurs every day, undoubtedly results in atmospheric losses. PCB's have been detected in snowfall samples in Wisconsin (ref. 5).

The largest quantities of PCB's in plasticizer applications end up in dumps and landfills. Much of the material is in sealed containers or impregnated in plastics and is slowly released to the environment. Nisbet and Sarofim (ref. 6) reported that vaporization directly from paints, coatings, and plastics does occur, with losses as great as 20 percent. Open burning in landfills also releases PCB's to the atmosphere. Another source is loss to leachate from landfills. Samples of surface runoff water from nine landfills in Michigan showed five of the nine samples having PCB concentrations ranging from 0.04 to 0.30 μg/l, while the others contained less than 0.01 μg/l.

In 1970, the U.S. Food and Drug Administration identified milk contamination in Ohio, Georgia, and Florida resulting from use of a PCB-containing sealant in silos (ref. 3). In 1975 the Michigan Department of Agriculture investigated problems from this usage and found 76 dairy herds in Michigan with PCB's in milk ranging from a trace to 14 mg/l on a fat basis. Scrapings from silos on these farms contained PCB's up to 10,000 mg/kg. Eighty additional silos have been identified as having the PCB sealant and have been removed from use until a protective coating can be applied to eliminate transfer of PCB's to the cattle feed.

Hydraulic Fluids

PCB's have found wide usage in hydraulic systems involving extreme pressure and high-temperature conditions. Annual U.S. domestic sales of PCB's for hydraulic uses totaled approximately 8 million pounds in 1970 (ref. 1). In Michigan, the automotive and metal finishing industries have used PCB-containing hydraulic fluids extensively. Industrial inspections and questionnaires documented past usage to have been as much as 878,000 pounds/year in individual plants, with most being collected and disposed of through waste haulers. Large losses of PCB's from these plants to municipal sewers or directly to surface waters have also been identified. Wastewater discharges from identified users of PCB-hydraulic fluids have had concentrations as high as 7.1 mg/l PCB's, indicating losses as great as 30 pounds per day. In a materials balance study, one industry which showed in its records that it was replacing approximately 30,000 pounds PCB's/year in hydraulic systems calculated that 10,000 pounds/year were entering the atmosphere through vaporization losses, 8,000 pounds/year in absorbents placed in landfills, 6,000 pounds/year lost to soils and drainage systems on plant property, 5,000 pounds/year going to waste haulers, with the remainder incinerated.

All industries that we have identified as using PCB's in hydraulic systems have converted to substitute products, mostly phosphate esters. Complete elimination of PCB residues from the systems has proved ineffective even with repeated flushings. Sampling of replacement fluids indicates PCB contamination as high as 100 mg/l. Residues in discharge lines combined with loss of contaminated replacement fluids appear to result in continued low-level discharges. Because of existing inventories and possible purchase from foreign sources or reprocessing companies, it is unlikely that all hydraulic systems not included in our surveys or questionnaires have been switched to replacement fluids. One facility which we investigated had an inventory of 9,000 gallons of PCB fluids in storage.

In summary, Michigan's surveys of hydraulic system usage of PCB's suggests that this has been a major source of environmental contamination and may continue to contribute to the problem to a lesser but still significant degree.

Heat Transfer Systems

PCB's heat transfer fluids have been used in place of steam or superheated water in many industries and large building complexes. They provide advantages of reduced explosion hazards, absence of corrosion problems, and have good heat transfer characteristics (ref. 4). In 1970, Monsanto sold approximately 4 million pounds of PCB's for this application but phased out all sales in 1972 (ref. 3).

From 1971 through 1973, Michigan identified at least 12 facilities discharging PCB's to surface waters or municipal sewers from this usage. Concentrations in some discharges were as high as 5.2 mg/l.

One automotive company in Michigan reported using 34,000 pounds of PCB's per year in heat transfer systems at three of its plants. The waste fluids were reportedly disposed of by industrial waste haulers. Another smaller plant reported loss of 800 pounds in 1972 and that approximately 700 pounds of this total was incinerated, 75 pounds disposed of in dumps, and 25 pounds lost to sewers and drains.

In another Michigan survey, a chemical company complex was found to have an effluent concentration of 35 μg/l resulting from heat transfer system losses. Heat transfer losses are not all from industrial facilities. Sampling of interceptor lines of the Detroit, Michigan, sewerage system resulted in the identification of heat transfer usage from two residential building complexes and one hospital as large sources of PCB input to the system.

Miscellaneous Uses

In 1971, Michigan officials found a PCB concentration of 6.6 μg/l in the waste lagoon of a soap and detergent company. The source was traced back to the company's usage of PCB's as a dedusting agent at 0.7 percent in one of its powdered products. The company had discontinued this usage in June 1970 and the PCB's were apparently leaching from contaminated lagoon sediments.

The investment casting wax industry has been recently identified as another user of PCB's (ref. 7). This casting method, which has largely replaced sand casting for production of low-tolerance-quality metal castings, uses formulated waxes as a basic molding mode. The waxes are injected into metallic dies to form the part shape and then coated with ceramic to form a final mold. The wax is removed by insertion of the mold in an autoclave, which melts the wax. Residues of wax are also secondarily removed by insertion of the molds in a furnace at 1,600° to 1,800° F. During these two procedures, the bulk of the wax is collected and usually recycled but a portion is lost to vaporization, cooling water contact, and poor housekeeping practices.

The waxes in use are obtained from a few major suppliers, one of whom is a major importer of decachlorobiphenyl from an Italian source. We have found twelve firms in Michigan using this casting process and several firms that reprocess their waxes. Our preliminary investigations indicate that most of the waxes contain approximately 20 percent of PCB's or polychlorinated terphenyls (PCT's); one recent sample indicates the range may extend as high as 60 percent content in the wax. The nature of these casting operations, including both operational and storage modes, at many plants indicates strong potential for environmental loss. An effluent sample from one of the Michigan companies using investment casting wax contained 2.5 μg/l of PCB's.

During a telephone survey of 39 industries in Michigan during May 1975, several industries reported use of a toilet hand soap containing 0.05 percent PCB's by weight. Followup on these reports indicated that this product actually contains a chlorinated diphenyl oxide rather than PCB's.

Three industries in the telephone survey reported use of a hydrated lime product that contained about 0.5 mg/kg PCB's. This product was being used for water treatment and in production of glass pellets. No explanation was available on why the lime contained PCB's but it is assumed to be a contaminant rather than an additive.

Concentrations in Municipal Wastes

Because of the diversity of PCB usage, municipal wastewater is likely to receive PCB's from numerous domestic and industrial sources, many of which are listed above. Buckley (ref. 3) estimated that municipal outfalls would likely contribute less than 5 percent (300 tons) of total annual loadings (6,000 tons) to aquatic environments of the North American continent. His estimate was based on an assumed average PCB concentration of 10 μg/l in wastes serving a sewered population of 150 million people producing 130 gallons of sewage per person per day.

Sampling of 58 municipal wastewater treatment plant (WWTP) effluents throughout Michigan in 1973 showed an average concentration of 0.52 μg/l (table 1). This is considerably lower than Buckley's value of 10 μg/l used in his calculations but still exemplifies widespread contamination in municipal wastes, and documents that they are indeed a source of PCB loss to surface waters.

Much of the PCB's entering municipal waste treatment facilities are removed and become incorporated into the waste sludge. Table 2 shows PCB concentrations in the sludges from Michigan municipal plants sampled in 1973. Concentrations are much higher than in the effluents from the same plants. The average for all plants was 15.6 mg/kg, with individual values as high as 350 mg/kg. Disposal of these sludges may add to environmental PCB levels. Sewage sludges are commonly disposed of by incineration, spreading on agricultural land, or placing in landfills.

Discussion

While I have characterized in this report possible and proven sources of environment loss of PCB's in Michigan from several applications, I want to acknowledge that not all industrial effluents tested in Michigan contain measureable PCB levels. We have tested over 9[illegible] industrial samples, with approximately 40 percent of effluents sampled containing PCB's above our 0.1 μg/l laboratory sensitivity limit. Twenty-three percent of the industries tested had greater than 0.5 μg/l, 18 percent greater than 1 μg/l, 6 percent greater than 10 μg/l, and [illegible] percent greater than 100 μg/l. This represents a diversity of industries showing contamination and in many cases the source of effluent contamination could not be determined.

Since new industrial sources of loss are continuously being found, we feel we are far from eliminating the PCB inputs to surface waters by searching for point sources and taking corrective measures on a case-by-case basis. We are far from understanding the impact or comparative contribution of atmospheric losses but we feel these are probably very significant also.

In light of considerable environmental damage and economic impact of PCB contamination, an absence of any apparent decline in environmental concentrations with current control methods, the ubiquitous nature of usage, the general lack of adequate disposal facilities, and the indication that atmospheric transport and deposition is also significant, the Michigan Department of Natural Resources urges that a national ban be pursued on all domestic and imported PCB's destined for use other than in transformers and capacitors, and that the critical or essential use of PCB's in transformers and capacitors be immediately and critically reviewed in light of current potential replacement products.

Feeling that existing legislation was inadequate to control PCB's, a group of Michigan legislators introduced a bill on August 14, 1975, to prohibit the manufacture, sale, and use of PCB's in Michigan for all applications other than for transformers and capacitors. The bill would provide for labeling, reporting, and disposal requirements and specifies penalties for violations. The bill calls for a stepwise elimination of PCB's with the sale, manufacture, and use of products containing 25 percent or more PCB's forbidden by January 1, 1976; 10 ppm or more forbidden after January 1, 1977; and 5 ppm or more forbidden after January 1, 1978. Public hearings on the bill are currently being conducted by the Michigan House Committee on Marine Affairs. [The bill was amended and voted out of committee on December 11, 1975. The amendment changed the maximum allowable concentration after January 1, 1977, to 1,000 ppm and 100 after January 1, 1978.]

Table 1. Concentrations of PCB's in the final effluent from wastewater treatment plants throughout Michigan, Spring, 1973. Concentrations in micrograms per liter, dry weight.

City	Date	PCB's 1242	PCB's 1254	PCB's 1242:1254
Adrian	3/7/73			0.26
Albion	2/22/73		0.34	
Ann Arbor	3/1/73		0.25	
Bay City	3/1/73	3.20		
Battle Creek	2/28/73		<0.10	
Benton Harbor-St. Joseph	2/28/73		0.20	
Brighton	3/1/73			0.33
Cadillac	3/28/73		0.53	
Charlotte	5/1/73		0.34	
Constantine	2/28/73		0.46	
Detroit	2/28/73	2.15		
Dexter	3/1/73		0.23	
East Lansing	5/2/73		0.18	
Escanaba	3/28/73			0.28
Essexville	3/1/73		0.10	
Flint	2/22/73		<0.10	
Gladstone	3/28/73		0.44	
Grand Haven	2/21/73		<0.10	
Grand Rapids	2/21/73		1.05	
Holland	2/21/73		<0.10	
Houghton-Hancock	3/27/73		0.18	
Iron Mountain-Kingsford	3/28/73		0.69	
Ironwood	3/27/73			0.29
Jackson	2/22/73		<0.1	
Kalamazoo	2/29/73			1.12
L'Anse	3/27/73			0.31
Lansing	4/18/73		0.22	
Manistique	3/28/73		0.63	
Marquette	3/27/73		0.29	
Marshall	2/22/73		0.15	
Menominee	2/28/73		0.57	
Midland	3/1/73		<0.10	
Milford	3/8/73			2.20
Monroe	5/4/73		<0.10	
Mt. Clemens	3/8/73			2.90
Mt. Pleasant	3/28/73		0.29	
Muskegon	2/28/73		0.88	
Muskegon Heights	2/28/73		0.18	
Niles	2/20/73		0.48	
Norway	3/28/73		<0.10	
Owosso	2/21/73		0.12	
Pontiac (Auburn)	3/12/73			0.60
Pontiac (E. Blvd.)	3/12/73			0.54
Port Huron	3/8/73			1.80
Saginaw	3/1/73			0.77
Sault Ste. Marie	3/28/73		0.73	
St. Ignace	3/28/73		0.31	
Three Rivers	2/21/73		<0.10	
Traverse City	3/7/73		<0.10	
Trenton	2/28/73		<0.10	
Warren	2/27/73		<0.10	
Wayne County	2/28/73		0.40	
Wyoming	2/21/73		0.22	
Ypsilanti	2/28/73			0.31
Ypsilanti Twp #1	3/1/73		<0.10	
Ypsilanti Twp #2	3/1/73		<0.10	

[a]Concentrations based on best fit of three standards: Aroclor 1242, Aroclor 1254, or a mixture of Aroclor 1242 and 1254 in a 1:1 ratio.

Table 2. Concentrations of PCB's in the sludge from fifty-seven municipal wastewater treatment plants in Michigan, Spring 1973. Concentrations in milligrams per kilogram, dry weight.

City	Date	PCB's 1242	PCB's 1254	PCB's 1242:1254
Adrian	3/7/73			24.0
Albion	2/22/73		1.5	
Ann Arbor	3/1/73		1.1	
Bay City	3/1/73	352.0		
Battle Creek	2/28/73		2.8	
Benton Harbor-St. Joseph	2/28/73		13.8	
Brighton	3/1/73		<0.1	
Cadillac	3/28/73		<0.1	
Charlotte	5/1/73		6.8	
Constantine	2/28/73		2.1	
Detroit	2/28/73	32.1		
Dexter	3/1/73		3.2	
E. Lansing	5/2/73		4.6	
Escanaba	3/28/73		5.9	
Essexville	3/1/73		3.9	
Flint	2/22/73		6.3	
Gladstone	3/28/73		4.1	
Grand Haven	2/21/73		4.1	
Grand Rapids	2/21/73		11.8	
Holland	2/21/73		0.8	
Howell	4/26/73		15.0	
Houghton-Hancock	3/27/73		5.5	
Iron Mountain-Kingsford	3/28/73		9.5	
Ironwood	3/27/73		5.2	
Jackson	2/22/73		3.0	
Kalamazoo	2/29/73	23.3		
L'Anse	3/27/73		4.4	
Lansing	4/18/73		5.3	
Manistique	3/28/73		1.5	
Marquette	3/27/73		2.8	
Marshall	2/22/73		3.9	
Menominee	2/28/73		4.2	
Midland	3/1/73		2.9	
Milford	3/8/73			50.5
Monroe	5/4/73		3.3	
Mt. Clemens	3/8/73	175.0		
Mt. Pleasant	3/28/73		6.5	
Muskegon	2/28/73		12.7	
Muskegon Heights	2/28/73		11.0	
Niles	2/20/73		7.8	
Norway	3/28/73		<0.1	
Owosso	2/21/73		2.0	
Pontiac (Auburn)	3/12/73			12.1
Pontiac (E. Blvd.)	3/12/73			9.2
Port Huron	3/8/73			9.2
Saginaw	3/1/73		5.0	
Sault Ste. Marie	3/28/73		2.4	
St. Ignace	3/28/73		1.5	
Three Rivers	2/21/73		4.1	
Traverse City	3/7/73		1.8	
Trenton	3/28/73		<0.1	
Warren	2/27/73		<0.1	
Wayne County	2/28/73	2.0		
Wyoming	2/21/73		0.58	
Ypsilanti	2/28/73		2.0	
Ypsilanti Twp #1	3/1/73		<0.1	
Ypsilanti Twp #2	3/1/73		<0.1	

[a]Concentration based on best fit of three standards: Aroclor 1242, Aroclor 1254, or a mixture of Aroclor 1242 and 1254 in a 1:1 ratio.

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