

Polychlorinated Biphenyl (PCB) Degradation Processes: State of Science

1.0 Introduction to Polychlorinated Biphenyls

The family of polychlorinated biphenyls (PCBs) contains 209 theoretically possible molecular conformations, called congeners. Each congener consists of a biphenyl molecule substituted with from one to ten chlorine substituents. PCBs were produced as mixtures during the mid-twentieth century and sold for industrial applications under trade names such as Aroclor, Clophen, Fenclor, or Kanechlor. PCB mixtures are identified with 4-digit numerical designations, such as Aroclor 1242, which provide information about a mixture's specific chlorine content. In general, the first two digits describe the number of carbon atoms in the biphenyl molecule (i.e., 12) and the last two digits give the specific chlorine content of the mixture in percent by weight. For example, Aroclor 1242 is a mixture of PCB congeners that contains 42% chlorine by weight. One exception to this convention is Aroclor 1016, which contains approximately 41% chlorine by weight. The approximate molecular composition and physical properties of these and other common PCB mixtures are given below in Table 1.

Table 1. Approximate Molecular Composition (%) and Selected Physical Properties of Aroclor PCBs (Source: Cohen et al., 1993).

Composition and Properties	Aroclor						
	1221	1232	1016	1242	1248	1254	1260
Biphenyl	11.0	6.0	< 0.01	-	-	-	-
Monochlorobiphenyl	51.0	26.0	1.0	1.0	-	-	-
Dichlorobiphenyl	32.0	29.0	20.0	17.0	1.0	-	-
Trichlorobiphenyl	4.0	24.0	57.0	40.0	23.0	-	-
Tetrachlorobiphenyl	2.0	15.0	21.0	32.0	50.0	16.0	-
Pentachlorobiphenyl	0.5	0.5	1.0	10.0	20.0	60.0	12.0
Hexachlorobiphenyl	-	-	< 0.01	0.5	1.0	23.0	46.0
Heptachlorobiphenyl	-	-	-	-	-	1.0	36.0
Octachlorobiphenyl	-	-	-	-	-	-	6.0
Specific Gravity (@ 25/15.5° C)	1.18	1.27	1.37	1.38	1.41	1.50	1.56
Absolute Viscosity (cp @ 38° C)	5	8	20	24	70	700	resin
Solubility (: g/L @ 25° C)	200	-	240	240	54	12	2.7
Vapor Pressure (mm @ 25° C)	0.0067	0.0046	0.0004	0.0004	0.0004	0.00008	0.00004
Log K _{ow}	2.8	3.2	4.4	4.1	6.1	6.5	6.9

PCB mixtures are nonflammable, exhibit a high degree of chemical and thermal stability, and have desirable dielectric properties. These characteristics made PCBs well suited for a variety of industrial applications. Although the principle use of Aroclor mixtures was as dielectric fluid in electrical transformers and capacitors, PCBs were also used in heat transfer and hydraulic systems as well as in inks, lubricants, paints, adhesives and dust control agents. Approximately 1.25 billion lbs. of PCBs had been sold in the United States by the time their production and sale was restricted in 1977 (Cohen et al., 1993).

The widespread use and chemical stability of PCBs has led to extensive environmental contamination through accidental releases and inappropriate disposal techniques. In 1977 it was estimated that approximately 150 million lbs. of PCBs had been released into the environment, and that an additional 750 million lbs. were still in use and a possible source of future contamination (Cohen et al., 1993). PCBs have been found in at least 216 of 1177 hazardous waste sites on the National Priorities List (ATSDR, 1989).

When comparing PCB remediation technologies it is important to note the units used to describe PCB concentrations. The congeners contained in PCB mixtures have molecular weights ranging from 154.1 g/mole for biphenyl to 498.7 g/mole for decachlorobiphenyl; consequently, the removal of one mole of biphenyl from a PCB mixture results in significantly less mass removal than one mole of decachlorobiphenyl. Comparisons are further complicated by the fact that oxidative and reductive processes attack PCB mixtures in different ways, and by the fact that the toxicity of a mixture may not be significantly reduced unless specific highly chlorinated "dioxin-like" congeners are destroyed.

Oxidative processes destroy PCBs via ring cleavage and are generally more effective against the less chlorinated (and therefore less oxidized) congeners. This is particularly true for aerobic biodegradation. Oxidative processes therefore typically demonstrate significant reductions in both mass and molar concentrations of PCB mixtures; nonetheless, direct comparisons of results between independent studies are difficult unless both studies use the same unit conventions (i.e., mass units or molar units) or provide congener specific concentrations.

Processes that reductively dechlorinate PCB mixtures (e.g., anaerobic biodegradation, zero-valent iron) may significantly affect mass concentrations (e.g., ppm) through the removal of chlorine atoms, while the overall molar concentration (e.g., moles/L) remains relatively constant. This results from the fact that chlorine atoms make up a relatively large fraction of the overall mass of a PCB molecule. The removal of one chlorine atom will lower the mass of a decachlorobiphenyl molecule by 7%. However, the same dechlorination of decachlorobiphenyl to nonachlorobiphenyl does not change the molar concentration of a PCB mixture. Therefore, reductive dechlorination results reported in molar concentrations of PCB mixtures may show very low percent reductions despite a significant decrease in toxicity and an increase in the mixtures susceptibility to aerobic biodegradation.

2.0 Health Effects of PCB Contamination

The primary concern with PCB contamination stems from the fact that PCBs are known to cause cancer in laboratory animals, and are suspected of being human carcinogens. In addition to carcinogenicity, PCBs exhibit a variety of other detrimental health effects including, skin disorders, liver damage, and reproductive and developmental disorders. The most toxic PCB congeners have a coplanar (flat) conformation and chlorine substituents at the para- and meta- positions making them structurally similar to dioxins. These generally highly chlorinated congeners tend to demonstrate "dioxin-like" toxicity and represent the primary health threat to humans.

The hydrophobic nature of PCBs causes them to bioaccumulate in the adipose tissues of exposed animals and individuals; consequently, eating fish and shellfish from PCB-contaminated water is a primary route of human exposure (ATSDR, 1989). Similarly, PCBs bioaccumulate in the breast milk of nursing mothers whom then pass the PCB-contaminated milk to infants. In addition to exposure through ingestion, PCBs may be absorbed through the skin or inhaled as vapors or on contaminated dust particles.

3.0 Pertinent Regulations

Several government agencies have adopted regulations or guidelines governing PCB exposure to reduce the risk of human health effects. The maximum contaminant level (MCL) for PCBs in drinking water has been set by legislation at 0.0005 mg/L (EPA 822-B-96-002, 1996). The Food and Drug Administration (FDA) issued PCB concentration limits between 0.2 and 3 ppm in infant foods, eggs, milk (milk fat), and poultry (fat). The Occupational Safety and Health Administration (OSHA) permissible occupational exposure limits for selected PCBs range from 0.5 to 1.0 mg/m³ for an 8-hour workday. In contrast, the National Institute for Occupation Safety and Health (NIOSH) recommends an occupational exposure limit for all PCBs of 0.001 mg/m³ of air in a 10-hour workday, 40-hour workweek.

The risk posed to human health and the environment by PCB contamination has driven the search for cost effective remedial technologies. The Code of Federal Regulations (CFR) addresses PCB treatment/disposal methodologies in 40 CFR 761.65. The CFR requires non-liquids (e.g., soils, rags or other debris) with a PCB concentration greater than 50 ppm be incinerated or sent to a chemical waste landfill. Similarly, liquids (other than mineral oil) with PCB concentrations between 50 and 500 ppm are to be disposed of in a chemical waste landfill or treated in an incinerator or high efficiency boiler. The relatively high costs associated with these disposal options have driven the development of other methods to lower PCB levels in contaminated materials.

4.0 Physical PCB Treatment Processes

A number of physical processes have been proposed for the remediation of PCB contaminated soils. The following subsections provide brief discussions about the treatment of PCBs with incineration, in-situ vitrification, solvent extraction, stabilization, and base catalyzed decomposition.

4.1 Incineration. The Toxic Substances Control Act (TSCA) requires that 99.9999% of PCBs be destroyed if incineration is to be used for PCB remediation. To achieve this destruction and removal efficiency, the incinerator must operate at a high temperature (>1000°F) and be capable of holding the PCB contaminated waste for long residence times. There are fewer than 10 TSCA permitted PCB incinerators in the USA. Some of these incinerators are mobile and can be brought to the waste site without additional TSCA permitting, but air discharge and other permits may still be required depending on the location and type of waste. It costs between \$500 to \$800 per ton to incinerate PCB contaminated solid waste and about \$3.25 to \$5.00 per gallon to incinerate liquid PCB wastes (Amend and Lederman, 1992). This is a conventional but expensive technology for PCB remediation.

4.2 In-situ vitrification. In-situ vitrification (ISV) is an emerging remedial technology that does not destroy the contaminants, but rather uses electrical current to melt contaminated soils in situ, thus immobilizing any contaminants not volatilized by the heat of the process. Off-gases must be treated for organic contaminants. The volume of contaminated waste is reduced 20-40% by the ISV process since pore spaces in the soil are eliminated (Amend and Lederman, 1992). The resulting vitrified material may be left in place and covered with clean fill or soil. Treatability tests are required before using this

technology for PCB contaminated soils because the process may not provide adequate immobilization in all cases. Currently there is only 1 vendor of this technology, and the cost is similar to incineration.

4.3 Solvent extraction. Solvent extraction (SE) is another non-destructive method of PCB remediation. The process makes use of the preferential solubility of PCBs in oily solvents to concentrate the contaminants in a single phase. One study achieved a 98% removal of approximately 20 ppm of PCB contamination in oily soils (Amend and Lederman, 1992). The cost of this process is very site and waste specific. Once the PCBs are concentrated in the solvent stream, they must still be destroyed by incineration or disposed in a proper manner. Solvent extraction is an emerging technology.

4.4 Stabilization. Stabilization is another non-destructive, emerging technology for PCB remediation. In this process, the PCB contaminated soil is excavated and mixed with amendments and binders that immobilize PCBs within the matrix. Volume increases of up to 120% have been reported. The stabilized waste is then landfilled. The procedure costs approximately \$250 per ton to stabilize the waste. Landfill costs are additional. At least one vendor has experienced significant leaching from stabilized waste matrices, raising concerns about the effectiveness of the stabilization process (Amend and Lederman, 1992).

4.5 Base Catalyzed Decomposition. Base catalyzed decomposition (BCD) is a somewhat complicated two-stage process that had been proposed for the treatment of PCB contaminated soils and sediments. In the first stage contaminated materials are mixed with NaHCO_3 and heated to 340°C . Vapors resulting from this treatment are condensed, separated and treated with either carbon adsorption or continue on to the second stage of the process. In Stage 2, the condensate is heated to 340°C and mixed with a hydrogen donor oil, NaOH , and a catalyst. Each stage of the process requires the treatment of off-gas and the disposal of treated soils and residues, thus raising the overall cost of the effort considerably. Although greater than 97% reductions in the PCB mass concentration have been reported after first stage treatment, treated materials still require disposal (Chen et al., 1997). In addition, little is known about the PCB removal mechanisms, leading to questions about the formation of toxic by-products.

5.0 Biological Treatment Processes

In addition to physical treatment processes, a number of studies have been performed to evaluate the effectiveness of biological treatment methodologies on PCB contaminated materials. In general, researchers have attempted to stimulate microbially catalyzed PCB-degrading activity through nutrient addition, genetic engineering, bioaugmentation, cometabolite addition (e.g., adding biphenyl), and microbial enrichments. Other methods for improving biological PCB destruction include using any of several physical, chemical or biological methods (e.g., surfactants) to increase PCB bioavailability. In general, a wide variety of biological treatment processes have been investigated, each with various advantages and disadvantages, but no one treatment offers a clearly superior solution to the PCB problem.

Initially, researchers focused on the ability of aerobic microorganisms to metabolize PCBs. It soon became clear that most aerobic microorganisms were limited to the metabolism of PCBs with fewer than 4 chlorine substituents. The discovery of weathered Aroclor 1242 in Hudson River sediments suggested that the more highly chlorinated PCBs were susceptible to reductive dechlorination by anaerobic microorganisms. Following this discovery, the ability of anaerobic microorganisms to degrade PCBs was investigated. Although it was found that anaerobic microorganisms could indeed reduce the number of chlorine substituents from highly chlorinated PCBs they were unable to completely degrade PCB mixtures. Researchers then examined the effectiveness of using anaerobic and aerobic biodegradation in sequence. Each of these approaches and have been discussed in the following sections along with a brief

account of published field demonstrations.

5.1 Aerobic Biodegradation. In general, aerobic biodegradation of PCBs is limited to the congeners containing fewer than 4 chlorine atoms, but a few strains of aerobic bacteria have demonstrated the ability to degrade tetra-, penta-, and hexa- chlorobiphenyls. Because the majority of congeners in Aroclors 1221, 1232, 1016 and 1242 contain fewer than 4 chlorine atoms, it is possible to demonstrate significant levels of PCB mass removal with aerobic biodegradation. In fact, mass reductions between 50 and 85% have been commonly reported in materials contaminated with up to 1,000 ppm of Aroclors 1221 through 1248. One recent study showed a 67% drop in the molar concentration of weathered Aroclor 1248 from soil slurry microcosms with a low organic carbon content (Evans et al., 1996). The main disadvantage of aerobic PCB biodegradation is its inability to breakdown the more highly chlorinated and more highly toxic congeners. Consequently, the overall reduction in risk to human health and the environment may not be as significant as the overall PCB mass removal.

Aerobic bacteria attack less chlorinated PCBs via aerobic oxidative processes, predominately by the enzymatic 2,3-dioxygenase pathway, followed by oxidation through a second dioxygenase and ring cleavage (Harkness et al., 1993). This pathway leads to the production of chlorobenzoic acid intermediates that can build-up and inhibit pure cultures of PCB degrading organisms, but are readily hydrolyzed by other aerobic bacteria in diverse microbial communities.

In addition to the bioremediation of PCBs with aerobic bacteria, the ability of various species of white rot fungi to biodegrade PCBs has also been investigated. These fungi are the primary degraders of lignin, a non-repetitive polymer consisting of numerous substructures of which the biphenyl group is included. In nature, these organisms secrete a series of peroxidase type enzymes that break up the lignin in wood to free up cellulose that the fungi use for energy and growth. These enzymes are produced in response to nutrient stresses such as carbon and nitrogen limitation, and they have been implicated in the breakdown of environmental contaminants including PCBs. One of the primary differences between bacterial and fungal degradation of PCBs is that the fungi tend to degrade a wider range of PCB congeners than do bacteria. *Phanaerochaete chrysosporium*, the most commonly studied species of white rot fungi, has been shown capable of degrading a wide range of Aroclors from 1242 to 1260. Like aerobic bacteria, the rate and extent of degradation is inversely proportional to the chlorine content, with little metabolism of the tetrachlorinated and/or hexachlorinated congeners. Other species including *Trametes versicolor* and *Pleurotus ostreatus* were very effective at degrading these congeners.

Although white rot fungi have been shown to be capable of degrading PCBs in flask studies, there has been limited success applying these organisms in engineered applications for soil remediation. White rot fungi grow naturally on decaying wood, not in soil, and treatment of contaminated soil requires development of an inoculum consisting of the organism and a suitable growth support (i.e. wood or some cellulose based matrix). The challenge with PCB remediation is to balance the nutritional requirements and metabolism of the fungi on the support matrix, while assuring the survival of the fungus in the soil environment.

5.2 Anaerobic Biodegradation/Reductive Dechlorination. In contrast to aerobic biodegradation which destroys PCBs via ring cleavage, microbially catalyzed reductive dechlorination simply removes chlorine substituents from the more highly chlorinated congeners. By shifting the congener distribution to less chlorinated analogs, the overall toxicity of the mixture is typically reduced and the mixture becomes more susceptible to aerobic degradation. In general, reductive dechlorination preferentially removes chlorines from the meta and para positions and replaces them with hydrogen atoms, resulting in substantial reductions in carcinogenicity and "dioxin-like" toxicity. In addition to lowering the overall toxicity of PCB contaminated materials, the tendency of the PCB mixture to bioaccumulate is also

reduced. For example, 2-chlorobiphenyl and 2,2-bichlorobiphenyl display an approximately 450-fold decrease in the tendency to bioaccumulate in fish compared with tri- and tetra-chlorinated PCBs (Abramowicz and Olson, 1995). Most importantly, the resulting less chlorinated PCBs are susceptible to aerobic biodegradation process.

Several approaches have been attempted to enhance the microbially catalyzed reductive dechlorination of PCBs. Researchers have attempted to stimulate dechlorination by amending microcosms with carbon substrates (e.g., fatty acids). Although in some cases this resulted in shortened lag times or increased initial rates of dechlorination, the overall extent of PCB dechlorination was not significantly increased (Abramowicz and Olson, 1995). Others have attempted to stimulate dechlorination by adding individual polychlorinated or polybrominated congeners to microcosms. The process is designed to selectively enhance populations of organisms that can use the supplied congener as an electron acceptor. In one instance, this strategy reduced 79% of hexa- through nonachlorobiphenyls in sediments contaminated with Aroclor 1260; the resulting dechlorination products were predominately tri- to pentachlorobiphenyls (Abramowicz and Olson, 1995). This approach may not be applicable to in situ remediation efforts due to the potential regulatory resistance encountered at the prospect of adding polyhalogenated biphenyls to a contaminated site.

The use of anaerobic biodegradation as a stand alone treatment for PCB contaminated sites would likely be hampered by regulatory treatment goals which are generally based on mass removal, not toxicity reduction. Although anaerobic dechlorination may provide greater toxicity reduction than aerobic PCB biodegradation, it is less likely to be used at a contaminated site because it does not produce the same level of PCB mass removal. The discrepancy in mass removals can be partially explained by the fact that a mass reduction of only 34.45g is observed for each mole of chlorine atoms removed via reductive dechlorination, while a 257.5g mass reduction is observed for each mole of trichlorobiphenyl destroyed aerobically. To optimize the reduction of both PCB toxicity and mass removal, researchers have begun investigating the utility of following anaerobic biodegradation with an aerobic biodegradation step.

5.3 Sequential Anaerobic-Aerobic Biodegradation. Sequential anaerobic-aerobic biodegradation is a two step process in which PCB contaminated soils/sediments are first incubated anaerobically to reductively dechlorinate the more heavily chlorinated PCB congeners (i.e., congeners with more than 3 chlorine substituents). The anaerobic incubation is followed by an aerobic incubation intended to degrade the resulting mass of less-chlorinated congeners. In principle this should permit a significantly greater PCB mass removal, particularly of the more highly chlorinated Aroclor mixtures.

One sequential study using soils contaminated with 1240 ppm of Aroclor 1248 demonstrated a 9% decrease in the PCB mass concentration after 12 weeks of anaerobic incubation, followed by an additional 72% decrease after aerobic incubation resulting in a total mass reduction of 81%. The same study showed that the aerobic treatment alone resulted in PCB mass removals as high as 80%, but more commonly aerobic reductions were around 72% (Shannon et al., 1994). Although these numbers may cause one to question the utility of the anaerobic incubation, it is important to remember that only 1% of the congeners in Aroclor 1248 contain six or more chlorine atoms and that anaerobic dechlorination is more effective against the more highly chlorinated congeners.

A second investigation examined the aerobic and the sequential anaerobic-aerobic treatment of weathered Aroclor 1248 in soil slurry microcosms. Microcosms had an initial Aroclor concentration of 100 ppm and an organic carbon content of 0.6%. Microcosms used in the sequential treatment process demonstrated dechlorination for the first 19 weeks of a 79-week anaerobic incubation. After week 79, the microcosms were amended with *Pseudomonas* sp. LB400, a known PCB degrading bacterium, and aerobically incubated for an additional 19 weeks. The sequential anaerobic-aerobic incubation produced

a 70% reduction in PCB molar concentration; similarly, aerobically incubated microcosms exhibited a 67% decrease in PCB concentration (Evans et al., 1996). Again, the fact that the vast majority of congeners in Aroclor 1248 are susceptible to aerobic degradation skews data in favor of the aerobic incubation. Nonetheless, results did show that microcosms undergoing aerobic treatment alone contained a higher proportion of penta- and hexachlorobiphenyls.

5.4 Field Demonstrations.

Although no full-scale field demonstrations of PCB bioremediation have been documented; a few pilot-scale field demonstrations have been conducted with varying degrees of success.

In 1987, General Electric (GE) made the first field scale attempt to bioremediate PCBs at a racetrack in New York which had been sprayed with PCB oil for dust control. Initial PCB concentrations at the site ranged from 50-525 ppm. GE bioaugmented the site with *Pseudomonas* sp. LB400, a known PCB degrading bacterium, and achieved an approximately 20% reduction in PCB concentrations in 4 months. This reduction was only about half of what was expected based on bench-scale treatability studies on site soil. Researchers speculated that environmental factors such as temperature and moisture content in the soil hampered the treatment effectiveness (McDermontt et al., 1989; Harkness and Bergeron, 1990).

GE attempted another PCB bioremediation field study with Hudson River sediments in a bioslurry with added inorganic nutrients, biphenyl, and oxygen. The initial concentration of PCBs in the sediments was 39 ppm, and from 37-55% reduction was achieved. A possible explanation for the low destruction was low bioavailability of the PCBs, which can be strongly sorbed onto organic matter in soil (Harkness et al., 1993; 1994).

Woods Pond Massachusetts was the site chosen for GE's attempt at anaerobic, field-scale bioremediation of PCBs. In this study, 2,6-dibromobiphenyl was added as a co-substrate, and the following reductions were observed: hexachlorobiphenyl – 70%, heptachlorobiphenyl – 57%, and octachlorobiphenyl – 27%. The degradation products of these reactions, tri- and tetrachlorobiphenyls, increased accordingly. It appeared that the reaction mechanism favored *meta* dechlorination followed by *para*. No *ortho* dechlorinations were observed (Bedard et al., 1995).

Land application biotreatment of PCB contaminated sludge achieved an 85% reduction in lower congeners for the Madison Metropolitan Sewerage District (Gan & Berthouex, 1994). The sludge had an initial PCB concentration of 50 ppm. The land was tilled, and commercial fertilizer was applied to the sludge/soil mixture. Half lives for the lower PCB congeners were measured: dichlorobiphenyl – 7 to 11 months, trichlorobiphenyl – 5 to 17 months, tetrachlorobiphenyl – 11 to 58 months with an overall half life of 19 months for PCBs in general.

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