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Environmental Fate and Global Distribution of Polychlorinated Biphenyls

Angelika Beyer and Marek Biziuk

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1 Introduction

Polychlorinated biphenyls (PCBs) are synthetic chlorinated organic chemicals. They are environmentally widespread and persistent and are routinely found in air, water, sediments, and soils. Moreover, they accumulate through the food chain from aquatic organisms to fish and to humans. PCBs are complex mixtures of individual chlorobiphenyls. The low reactivity and high chemical stability of the PCBs have made them useful for numerous industrial applications; the qualities that make many individual chlorobiphenyls industrially useful, however, render them more persistent and less environmentally desirable than many other organic chemicals.

There is growing concern about the trace quantities of *highly chlorinated organic compounds* (e.g., dioxins, *PCBs*, and certain pesticides) that exist in diverse environmental media (air, soil, water, and biota). Such halogenated organic compounds enter the food chain from environmental media, mainly through intake of animal or fish fats (meat, fish, and milk), and reach humans and wildlife.

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A common ingredient of these compounds is chlorine, one of the most abundant naturally occurring chemical elements on this planet. Since the discovery of chlorine, the chemical industry has developed many processes and products that either contain chlorine or are produced with its help. However, as the volume and diversity of chlorinated substances has increased, environmental concerns for them has also increased; many chlorinated moieties that are persistent, toxic, and bioaccumulative have been banned (Lopez 2003; Lohmann et al. 2007). Consequently, there is growing scientific, regulatory, and social interest in measuring the levels of chlorinated chemicals in environmental media, and in determining the environmental effects of such contamination.

PCBs are produced by chlorination of biphenyls and comprise a class of 209 individual discrete chemicals, each with 12 carbon atoms. The PCB structure is presented in Fig. 1. Chlorine is substituted for hydrogen atoms in any of the ten numbered positions of this structure.

Other than commercial production, PCBs are also created as thermodynamically stable by-products of certain combustion processes such as incineration of PCB-containing wastes. EPA regulations concerning the storage and disposal of PCBs have specified incineration as the only acceptable method of PCB disposal unless, by reason of the inability to dispose of the waste or contaminated materials in this manner, clearance is obtained from EPA to dispose of materials in another way (EPA 1978). However, the general acceptance of incineration as a means of disposal for PCB-contaminated materials has declined because of concerns about incomplete incineration and the possible formation of highly toxic by-products, such as hydrogen chloride, polychlorinated dibenzodioxins, and polychlorinated dibenzofurans, if the combustion temperature is not held sufficiently high (Arbon et al. 1994; Chuang et al. 1995).

Depending on the degree of PCB chlorination and their physicochemical properties (Fig. 2), PCBs have successfully been used as stable fluid insulators in high-voltage electric transformers, in high-capacity condensers, as heat exchangers, pesticide extenders, adhesives, dedusting agents, components of cutting oils, flame retardants, hydraulic lubricants, and components of plasticizers in paints, inks, toners, and printing inks (Erickson 1997).

As PCBs move through specific environmental compartments, or move from one environmental medium to another, the relative concentrations of individual chlorobiphenyls change. These changes result from physical and chemical processes and selective bioaccumulation and biotransformation of PCBs by living organisms. These processes produce mixtures that are substantially different from the ones

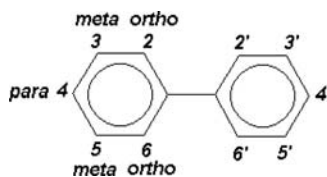


Fig. 1 Structure of polychlorinated biphenyls (PCBs)

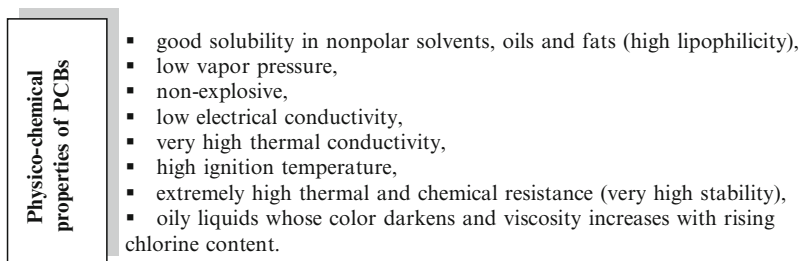


Fig. 2 Physicochemical properties of PCBs

originally deposited into the environment. The primary reason that aged environmental residues of PCB mixtures differ from the original forms deposited in the environment is that some congeners are more easily degraded than others. In particular, *ortho*-substituted congeners are readily degradable; therefore, they are not as abundant in aged environmental samples. In contrast, the so-called “dioxin-like” PCBs, namely the coplanar (= *non-ortho* substituted) and *mono-ortho*-substituted congeners, are very stable and resist biodegradation and metabolism; these congeners are present in commercial mixtures and in aged environmental samples as well. Therefore, identification and quantification of the PCB mixtures on which the environment has acted is complicated, as is the task of assessing the risk posed by these residual mixtures in the environment.

In this paper, we review the global distribution and environmental fate of PCBs. We emphasize aspects pertaining particularly to bioaccumulation, bioconcentration, and biomagnification of these environmental contaminants. Our review supports the conclusion that the presence of PCBs in the environment may pose long-term public health and ecosystem risks.

2 Global Distribution

Human activities constitute the only source of PCBs in the environment. Hence, sites that are highly contaminated by PCBs tend to exist in industrialized areas. PCBs are ubiquitous in the general environment (Fig. 3) and consequently are transported by wind and water. PCBs bind strongly to organic particles in the water column, atmospheric particulates, sediments, and soil. The deposition of particle-bound PCBs from the atmosphere and sedimentation from water are largely responsible for their accumulation in sediments and soils.

The principal transport route for PCBs through aquatic systems is from waste streams into receiving waters. Currently, the major source of PCB release to surface water is the environmental cycling process (Mackay 1989; Swackhamer and Armstrong 1986). Small amounts of PCBs may enter receiving waters by runoff from accidental spillage of PCB-containing hydraulic fluids, disposal of waste oils

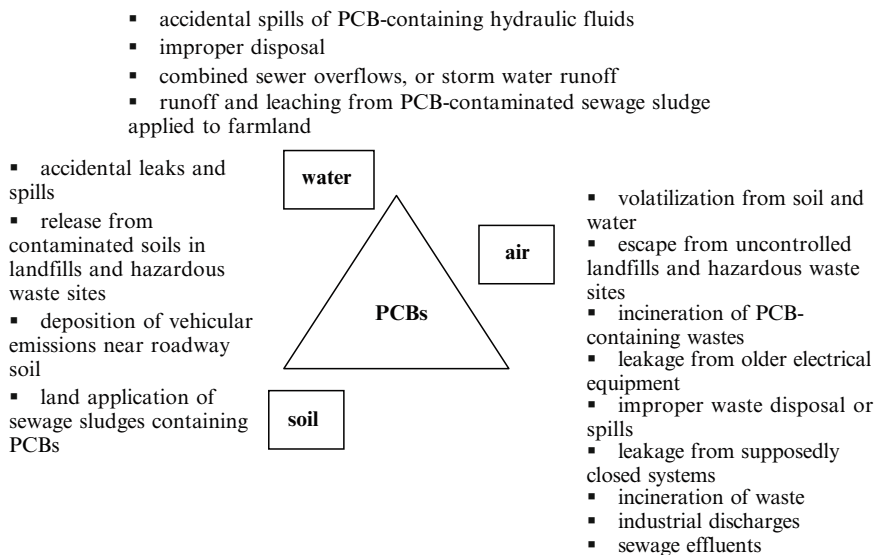


Fig. 3 How PCBs enter the general environment

into street drains, or from farmland to which sewage sludge containing small quantities of PCBs has been applied (Gan and Berthouex 1994; Gunkel et al. 1995). PCBs are persistent in waters where an equilibrium is established as PCBs are adsorbed from water onto particles or sediments or desorbed back into water or biota. The marine environment is one of the final sinks for PCBs. PCBs do not remain long dissolved in water. Therefore, they volatilize into the atmosphere, and large quantities volatilize from lakes and seas, even as PCBs from global air masses are deposited into them (U.S. Department of Health and Human Services et al. 2000; Wolska et al. 2003).

The importance of volatilization to atmospheric concentrations of PCBs is well established. This conclusion is also supported by the estimated Henry's law constants (HLCs) for individual PCB congeners (Thomas 1982). Henry's law constant is a fundamental property that describes the tendency for water to reach an equilibrium between the atmosphere and surface waters. The lack of accurate HLC values is a major problem in modeling the transport and fate of PCBs in the environment, a necessary step for performing risk assessment and developing environmental remediation strategies (Phillips et al. 2008). HLCs have been measured for selected congeners, commonly at 25°C. The largest experimental data sets at this temperature are those of Brunner et al. (1990), who measured HLCs for 58 congeners, and Bamford et al. (2000), who measured HLCs for 26 congeners. However, HLC is a highly temperature-dependent property, and transport and fate modeling requires knowledge of HLCs over a range of environmentally relevant temperature (Phillips et al. 2008).

The toxicity to animal life mainly results from the presence of PCBs in water. The PCBs that exist in water or sediments are filtered through fish, crustaceans, or

mollusks via their gills; because these xenobiotics are lipophilic, they make their way into the fatty tissues of these animals. These aquatic animals are, in turn, eaten by birds and mammals, which also accumulate PCBs in their fatty tissues. Such intake at successive trophic levels further concentrates the PCB residues. Such concentration from one trophic level to another is called biomagnification and is further explained later. Yolks rich in fatty material, from eggs laid by bird species near the top of the food chain, often have the highest concentrations of PCBs.

Several approaches are used to study the fate of PCBs in the environment. The main purpose of such studies is to understand contaminant concentrations and fluxes among environmental compartments under a variety of environmental scenarios. Traditional monitoring tools, such as measurements of chemical concentrations, are employed to verify the level of contaminants in a given environmental compartment (air, water, soil, sediment, biota; Borga and Di Guardo 2005). Since the 1970s, microecosystems (microcosms) have also been widely used to gain an understanding of the effects of toxic substances at the ecosystem level; microecosystems, based on this small prototype concept, were constructed for the study of pesticides (Kersting 1975, 1984, 1991; Kersting and Van Wijngaarden 1992). In these studies ideas about quantifying changes that occur in complex systems have evolved (Kersting 1984, 1991). Another approach is to mathematically model the contaminant distribution in the environment by using multimedia models. With this approach, an attempt is made to relate physicochemical properties of a chemical to an understanding of its probable partition and transport in the environment. Models can be purely empirical or mechanistic (Boese et al. 1995; Van Bavel et al. 1996). Borga and Di Guardo (2005), in their paper, illustrate and discuss some of the difficulties encountered when different approaches are used to study environmental distribution of contaminants. Breivik et al. (2007) present an update of a dynamic mass balance approach aimed at presenting the “big picture” of the global historical atmospheric emissions of selected PCBs.

3 Environmental Fate

As a result of their chemical stability, PCBs are environmentally persistent and are among the most prominent and widespread environmental contaminants. When these undesirable features are combined with the low water solubility of PCBs, great concern emerges, because these substances may be (and are) accumulated through the food chain and reach aquatic organisms, fish, and humans. However, it has been discovered, with great interest, that PCBs can be dehalogenated in freshwater and in estuarine sediments (Abramowicz 1994; Bedard et al. 1996; Berkaw et al. 1996; Brown et al. 1987; Brown and Wagner 1990; Fish and Principe 1994; Quensen et al. 1990). The deep interest in this discovery derives from the fact that dehalogenation is expected to detoxify these dangerous substances (Bedard and Van Dort 1997). Another important pathway for PCB loss is volatilization, and colloidal adsorption of PCBs to dissolved organic matter (DOM), which reduces bioavailability (Fig. 4).

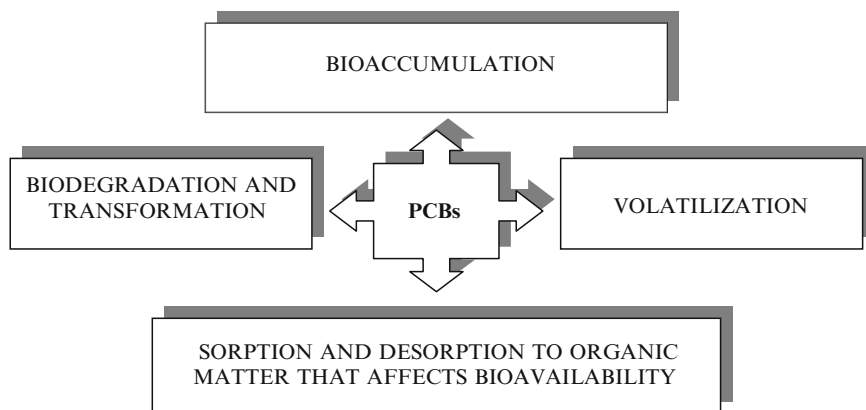


Fig. 4 Processes that affect the environmental fate of PCBs

3.1 Biodegradation and Transformation

Biodegradation occurs primarily through the action of bacteria or other microorganisms, either aerobically or anaerobically. It is the only process known to degrade PCBs in soil or in aquatic environments. The specific processes that achieve degradation are generally either aerobic oxidative dechlorination, or hydrolytic dehalogenation and anaerobic reductive dechlorination. Theoretically, the biological degradation of PCBs should produce CO_2 , chlorine, and water. This process involves the removal of chlorine from the biphenyl ring followed by ring cleavage and oxidation of the resulting compound (Boyle et al. 1992).

Different microbes have different capabilities to degrade PCBs. Microbial degradation processes comprise a set of dechlorination reactions that determine which PCB congeners (of the 209 congeners that are theoretically possible) are suitable substrates, which chlorines will be removed from those congeners, and the order in which these chlorines will be removed (Bedard and Quensen 1995). Results of several studies have demonstrated that different PCB dechlorination processes appear to be responsible for microbial dechlorination (Bedard and May 1996; Bedard and Quensen 1995; Brown and Wagner 1990; Sokol et al. 1994), but maximal chlorine removal requires the complementary action of two or more such dechlorination processes (Bedard and Quensen 1995; Quensen et al. 1990). It has been proposed that discrete dechlorinating microorganisms that harbor dehalogenases, with different regiospecificities, are responsible for the various dechlorination processes that have been described (Bedard et al. 1993, 1996, 1997; Bedard and Quensen 1995; Brown and Wagner 1990; Quensen et al. 1990).

For example, aerobic bacteria of the genus *Pseudomonas* degrade PCBs by aerobic oxidative dehalogenation. This process involves the addition of oxygen to the biphenyl ring. The metabolic pathway used by *Micrococcus* sp. resembles that described for *Pseudomonas* sp. and is illustrated in Fig. 5 (Benvinakatti and Ninnekar 1992).

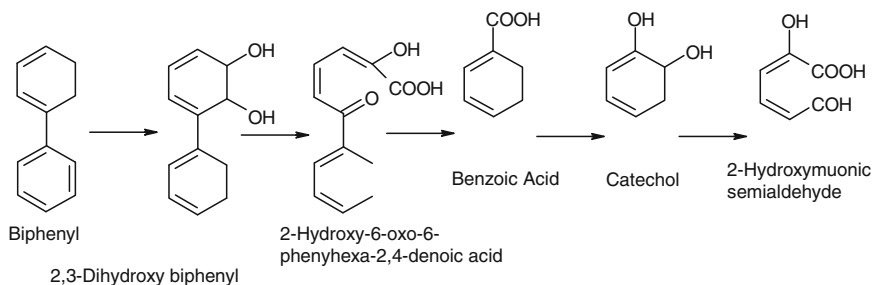


Fig. 5 A possible pathway for the aerobic oxidative dehalogenation of PCBs

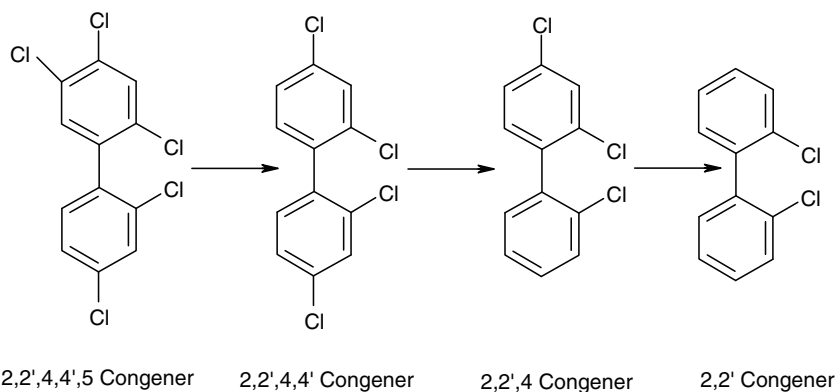


Fig. 6 A potential pathway for anaerobic degradation of highly chlorinated PCB congeners to less chlorinated ones

PCB congeners can be dechlorinated selectively by either aerobic or anaerobic bacteria. However, only the lower chlorinated congeners are amenable to aerobic degradation. Aerobic PCB biodegradation is a cometabolic process, in which another substrate such as biphenyl is required as a carbon and energy source. Alternatively, anaerobic reductive dechlorination occurs only for the most heavily chlorinated PCB congeners. This process may be of selective advantage to microorganisms because it produces a gain in energy for the organism (Jafvert and Rogers 1990).

The rate of anaerobic dechlorination is constrained mainly by rates of chlorine removal from *meta* and *para* positions. Some methanogenic bacteria, for example, preferentially dechlorinate PCBs at the *meta* and *para* positions, with a resulting enhancement of mono-, di-, and tri-chlorinated *ortho*-substituted PCBs (Alder et al. 1993). In anaerobic environments, highly chlorinated biphenyls can undergo reductive dechlorination to less chlorinated congeners. Specifically, monochlorobiphenyls and *ortho*-substituted dichlorobiphenyls degrade in this manner. This type of degradation involves the replacement of chlorine with a hydrogen atom on the biphenyl ring and is illustrated in Fig. 6 (Fish and Principe 1994). Byproducts are less toxic than the parent molecules and can be degraded by aerobic microbes.

Under anaerobic conditions, not only are PCBs with *para*- and *meta*-substituted rings more easily degraded than the *ortho*-substituted compounds, but PCBs containing all chlorines on one ring are biodegraded faster than those that contain chlorines throughout both rings (Abramowicz 1995).

The study of bacterial PCB biodegradation has been considerably assisted by use of tritium-labeled PCBs. High specific activity permits tracing all tritium-labeled PCB biodegradation products. Kim et al. (2004) found that four *Bacillus* sp. strains retained the ability to accumulate and metabolically destroy PCBs.

Different microbial populations have demonstrated differing congener specificity when dechlorinating PCBs or breaking their carbon-carbons bonds (Quensen et al. 1990). However, such bacterial populations appear to display similar degradation patterns. In Woods Pond, at least seven distinct microbial dechlorination activities have been identified and described (Wu et al. 1997a; Table 1). It was shown in this study that individual reactions in sediments exhibited strong temperature dependencies. Moreover, temperature influenced the timing and preferred sequence of dechlorination. Results indicated that several distinct PCB-dechlorinating microbial populations, with different temperature ranges and different dechlorination specificities, exist in these sediments, but no pure cultures have yet been isolated. Van Dort and Bedard (1991) reported the first experimental demonstration of biologically mediated *ortho*-dechlorination of a PCB, and the first stoichiometric conversion of a PCB congener to less-chlorinated forms. However, this process does not appear to be important under natural conditions. Bedard et al. (1997) stimulated a new *para*-dechlorination activity (Process LP) that caused further conversion of Process N products to tri- and tetra-chlorobiphenyls.

To date, there are at least eight distinct, documented reductive dechlorination pathways or processes, each resulting in a different congener distribution profile. These dechlorination processes have been identified from careful examination of congener loss and product accumulation patterns in different sediment samples (Bedard 2003; Wiegel and Wu 2000) and marked using letters: N, LP, M, Q, H, H', T, P. Apart from patterns shown in Table 1, H, H', M, and Q processes have been described (Bedard 2003; Wiegel and Wu 2000). The documented patterns include *meta*- and *para*-dechlorination and also reflect the relatively infrequent observation of *ortho*-dechlorination, in many of the initial sediment studies.

Anaerobic transformation of highly chlorinated congeners into lower ones, in a variable environment, renders them subject to later aerobic microbial degradation, which can oxidatively mineralize lower Cl congeners such as the homologs Cl₁ and Cl₂ to carbon dioxide and water (Bedard et al. 1987; Bedard and Quensen 1995).

A detailed description of the potential for microorganisms to transform PCBs can be found in numerous review articles (Field and Sierra-Alvarez 2008).

Fish are generally considered to have poor capability to biotransform PCBs (Boon et al. 1989; Matthews and Dedrick 1984). However, results of some work suggest that fish do have some capacity to biotransform PCBs (Wong et al. 2002, 2004), and the formation of hydroxylated PCBs (OH-PCBs) in fish has been reported (White et al. 1997). Hydroxylated PCBs were found in a number of fish species from the Great Lakes (Campbell et al. 2003; Li et al. 2003), which presents

Table 1 The microbial reductive dechlorination of 2,3,4,6-tetrachlorobiphenyl in PCB-contaminated sediments from Woods Pond (Lenox, MA)

Process name	Dechlorination reaction	Specificity of dechlorination	Temperature range (°C)	Temperature at which reaction was dominant (°C)	Short description	Refs.
Process T	2,3,4,6 → 2,4,6	<i>meta</i>	4–34 50–60	4–34	A very restricted <i>meta</i> dechlorination of specific hepta- and octa-chlorobiphenyls	(Wu et al. 1997b)
Process P	2,3,4,6 → 2,3,6	Flanked <i>para</i>	18 30–34	18	Highly selective and removes only <i>para</i> chlorines flanked by at least one <i>meta</i> chlorine	(Quensen et al. 1990; Wu et al. 1997b)
Process N	2,3,6 → 2,6	<i>meta</i>	18 30–34	34	Selectively removes <i>meta</i> chlorines, but only those that are flanked by at least one chlorine in either the <i>para</i> or the <i>ortho</i> position	(Bedard and May 1996; Wu et al. 1997b)
Process LP	2,4,6 → 2,6 2,4 → 2	Unflanked <i>para</i> Unflanked <i>para</i>	15–30 15–30	18–22 18–22	Unflanked <i>para</i> dechlorination	(Wu et al. 1997b)

new concerns, because the toxicity of the transformation products may be greater than that of the parent compounds (Purkey et al. 2004). The results of a study by Buckman et al. (2006) demonstrate that fish can possibly biotransform and hydroxylate PCBs. They also suggest that it may be true that the ability of fish to biotransform PCBs is restricted, compared with birds and mammals. However, the mechanisms involved appear to be similar in all species and are dependent upon the chlorine substitution pattern of the PCBs being metabolized.

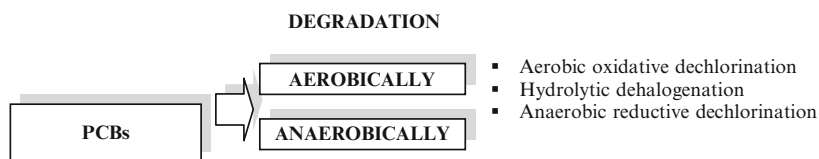
Fig. 7 summarizes the transformation processes associated with “higher chlorinated” biphenyls; potentially these are fully biodegradable, although anaerobic reductive dechlorination followed by aerobic mineralization of the lower chlorinated products is required (Field and Sierra-Alvarez 2008).

3.2 Volatilization

PCBs, especially those that have experienced significant microbial dechlorination, are very susceptible to volatilization. PCB volatilization from thin films on sand and soil was reported over 30 yr ago by Haque et al. (1974), who found minimal loss at ambient temperature. Even short drying times at room temperature resulted in significant (45–60% after 4 hr) PCB losses from sediments. Heat, air-flow (hood storage), coarse grain size, high water content, and enrichment in lower *ortho*-chlorinated congeners all were expected to increase the rate and extent of PCB volatilization (Chiarenzelli et al. 1996).

Clearly, volatilization results in differential loss of the lighter homologs, and the atmosphere is enriched by their volatilization, although subsequently they are subject to deposition from the atmosphere.

Apart from volatilization and biotransformation by microbes and higher organisms, PCBs are remarkably stable.



Factors that can affect PCB degradation:

- degree of chlorination – more highly chlorinated congeners are less readily degraded than less chlorinated ones
- the position of chlorine atoms on the rings
- concentration in environmental media of the congener
- type of microbial population present in media
- available nutrients, e.g., biodegradation rates decrease with high levels of organic carbon present
- pH and temperature

Fig. 7 Environmental factors that can affect the degradation of PCBs

3.3 *Adsorption to Organic Matter*

The environmental behavior of hydrophobic organic compounds in water is driven by partitioning between dissolved and sorbed phases. The partitioning behavior of a compound bears empirical relationships to other properties of the chemical, such as water solubility and the octanol-water partition coefficient (K_{ow}). However, actual partitioning behavior in the environment may differ significantly from such theoretical predictions (Butcher et al. 1998).

When sorbed to DOM, PCB contaminants are unavailable for uptake by organisms and, hence, become less bioavailable. In contrast, although PCBs sorbed to particulate organic matter (POM) prevent or constrain direct uptake of PCBs, these contaminants are still available to the detrital food web, which is an important pathway in rivers.

The fate of PCBs in detrital systems is dependent on the structure and origin of PCB congeners. In deeper bodies of water, fecal pellets representing POM rapidly sink to become bottom sediments (Baker et al. 1991). Cl_2 to Cl_4 PCB homologs may be present in higher concentrations in fecal material, but they are also released quickly. Baker et al. (1991) have shown that lower chlorinated PCB congeners are rapidly removed from surface waters and settle out from the water column to the sediment–water interface. At the lake floor, a large fraction of the recently settled out PCBs is released and mixed back into the water column.

Planar PCBs bind strongly to POM and are less bioavailable (Van Bavel et al. 1996). Highly chlorinated homologs sorb strongly to POM and are not assimilated easily by detritus feeders (Boese et al. 1995).

It is known that a combination of binding processes (sorption) and mass-transport processes (diffusion) is responsible for the partitioning of PCBs between aqueous and solid phases and for their transport between these phases. These processes are also directly involved in, and affect the environmental fate PCBs. Precise quantitative predictions of phase speciation may allow an a priori estimate of the directly bioavailable and dissolved fractions of pollutants and their tendency for long-term dispersion in the environment. Such predictions are critical in assessing the environmental risk from PCB contamination (Gdaniec-Pietryka et al. 2007).

3.4 *Bioaccumulation, Bioconcentration, and Biomagnification*

Organisms can accumulate high concentrations of PCBs relative to concentrations of these substances in nonbiotic portions of the environment. This phenomenon is variously referred to as bioconcentration, bioaccumulation, and biomagnification – terms, which we try to define more precisely later (Mackay and Fraser 2000).

Some confusion exists in the literature about definitions. We adopt and follow the terms set out by Gobas and Morrison (2000).

3.4.1 Bioaccumulation

Bioaccumulation is a selective process that causes an increased chemical concentration in an organism than in the surrounding medium, and it results from uptake by all exposure routes including transport across respiratory surfaces, dermal absorption (bioconcentration), and dietary absorption (biomagnification). Bioaccumulation can thus be viewed as a combination of bioconcentration and food uptake (biomagnification).

The bioaccumulation factor (BAF) in fish is the ratio of the concentration (C_B) of the chemical in the organism to that in water, similar to that of bioconcentration factor (BCF).

$$\text{BAF} = C_B/C_{\text{WT}} \text{ or } C_B/C_{\text{WD}}$$

The most common approach for evaluating levels of bioaccumulation is to compare the levels retained by the organism, with levels in the contaminated medium in which they live (Kucklick et al. 1996).

3.4.2 Bioconcentration

Bioconcentration results from uptake of chemicals from water (usually under laboratory conditions). Uptake occurs via the respiratory surface and/or skin, and results in the chemical concentration in an organism being greater than that in the surrounding medium.

The BCF is defined as the ratio of the chemical concentration (C_B) in an organism to the total chemical concentration (C_{WT}) in the water, or to the freely dissolved chemical concentration in water C_{WD} (it only takes into account the fraction of the chemical in the water that is biologically available for uptake). The BCF is expressed as follows:

$$\text{BCF} = C_B/C_{\text{WT}} \text{ or } C_B/C_{\text{WD}}$$

Although sometimes applied to other aquatic species, the principal target organism for BCF assessment tends to be fish, primarily because of their importance as food for many species, including humans. Moreover, there are standardized testing protocols to determine BCFs for fish (European Centre for Ecotoxicology and Toxicology of Chemicals 1996). However, experimental determination of BCF values is expensive and time-consuming. Therefore, methods are used to correlate physical or structural properties of chemicals to estimate probable BCF values. For example, quantitative structure–activity relationships (QSARs) that draw on results of experimental BCF testing are used to predict bioconcentration potential of chemicals in fish (Papa et al. 2007).

3.4.3 Biomagnification

Biomagnification is the bioaccumulation of a substance up the food chain when residues are transferred from consumption of smaller organisms by larger ones in the chain. It generally refers to the sequence of processes that produces higher concentrations in organisms at higher levels in the food chain (at higher trophic levels). These processes always result in an organism having higher concentrations of a substance than is present in the organism's food. Biomagnification also results in higher concentrations of the substance than would be expected if water was the only exposure mechanism (IUPAC 1993). A biomagnification factor (BMF) can be defined as the ratio of the concentration of chemical in the organism (CB) to that in the organism's diet (CA), and it can be expressed as follows:

$$\text{BMF} = \text{CB}/\text{CA}.$$

This is the simplest definition of a BMF. It can also be described as the ratio of the observed lipid-normalized BCF to K_{ow} , which is the theoretical lipid-normalized BCF. This is equivalent to the multiplication factor above the equilibrium concentration. If this ratio is equal to or less than 1, then the compound has not been biomagnified. If the ratio exceeds 1, then the chemical is biomagnified by that factor.

The mechanism of biomagnification is not completely understood. To achieve a concentration of a chemical greater than its equilibrium value indicates that the elimination rate is slower than for chemicals that reach equilibrium. Transfer efficiencies of the chemical would affect the relative ratio of uptake and elimination. There are many factors that control the uptake and elimination of a chemical after contaminated food is consumed; these include factors specific to the chemical (solubility, K_{ow} , molecular weight and volume, and diffusion rates between organism gut, blood, and lipid pools), as well as factors specific to the organism (the feeding rate, diet preferences, assimilation rate into the gut, rate of chemical's metabolism, rate of egestion, and rate of organism growth). Because humans occupy a very high trophic level, we are particularly vulnerable to adverse health effects from exposure to chemicals that biomagnify (Bierman 1990).

A problem arises when an organism has several food sources with different concentrations. Chemicals that bioaccumulate do not necessarily biomagnify, although many papers report that PCB congeners do in fact biomagnify (Burreau et al. 2006; Nfon et al. 2008). Some early bioaccumulation models used the concept of a food-chain multiplier, which is now considered excessively simplistic (Campfens and Mackay 1997). Exposure of PCBs solely from one source only occurs in laboratory experiments (Pelka 1998). In nature, organisms are always exposed to different sources of contaminants, and therefore what happens in the field is more complex than what is reflected in laboratory studies, and it cannot easily be emulated by laboratory studies. Mass balance models are simple tools that allow evaluation of various uptake and loss processes (Mackay and Fraser 2000). A variety of mass balance models have been developed to address water quality issues in lakes, estuaries, and slow-flowing water bodies (Chapra and Reckhow 1983). The simpler models only consider advection and an overall loss due to the combined processes of volatilization,

net transfer to sediment, and degradation. The rate constant for the overall loss is derived from fugacity calculations for a single segment system. The more rigorous models perform fugacity calculations for each segment and explicitly include the processes of advection, evaporation, water–sediment exchange, and degradation in both water and sediment. In this way, chemical exposure in all compartments (including equilibrium concentrations in biota) can be estimated (Warren et al. 2005, 2007). In general, these models consider the organism to be a single “box” (Fig. 8; Arnot and Gobas 2004; Mackay and Fraser 2000). These models require information about the chemicals, the organism, and associated environmental parameters (Arnot and Gobas 2004). Mackay and Fraser (2000) suggest using a tier 1 approach to screen chemicals for tendency to bioaccumulate. For those substances that are suspected of being bioaccumulative, because of their partitioning properties, a more detailed tier 2 evaluation is suggested; tier 2 uses a fish mechanistic model, which can be expressed in rate constant or fugacity formats. Such a model reveals the relative significance of gill ventilation, food uptake, egestion, and metabolism. The most detailed tier 3 evaluation is used to predict the potential for biomagnification in a food chain that involves both fish and air-breathing animals (Mackay and Fraser 2000).

PCB congeners with less *ortho*-substitution are accumulated up the food chain at a greater rate than other congeners in their homolog group (Campfens and Mackay 1997). Non-*ortho*-substituted congeners, especially those that lack adjacent unsubstituted *meta* and *para* sites and unsubstituted *ortho* and *meta* sites, are undoubtedly metabolically recalcitrant in invertebrate and vertebrate tissues (Bright et al. 1995). Changes in distributions of congeners are mainly caused by transfers among biotic compartments. There is no enrichment in higher trophic levels of mono- and non-*ortho*-substituted congeners. However, many coplanar congeners, especially very toxic PCB 77, are depleted with increasing trophic levels; PCB 77 is, therefore, almost certainly metabolized (Campfens and Mackay 1997).

Information on PCB bioaccumulation in *algae*, *invertebrates*, and fish is presented in Table 2. PCB bioaccumulation in sea gulls was described as a relatively simple two-compartment model representing partitioning between fat and blood plasma (Clark et al. 1998). But piscivorous mammals are exposed to high levels of pollutants because they feed at the top of aquatic food webs (Bremle et al. 1997).

Exposure of PCBs solely from one source only occurs in laboratorial experiments. In nature, there are always multiple sources of contaminants, and therefore field results must be studied carefully. Moreover, the properties of individual PCB

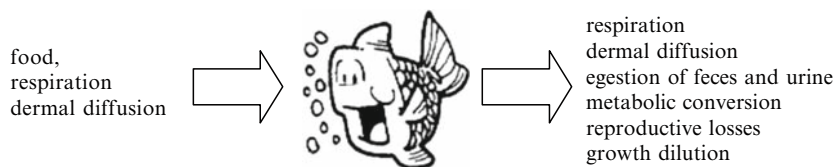


Fig. 8 Mechanisms of PCB uptake and elimination in organisms

Table 2 Factors that affect PCB bioaccumulation

Organism	Dependent factors	Lipid profile	Bioaccumulation mechanism	Uptake/sorption and desorption
<i>Algae (Periphyton and Phytoplankton)</i>	-Solubility	-PCBs partition to lipids in algae	-Probably a two-step mechanism, with rapid surface sorption of 40–90% within 24 hr and then a small, steady increase with transfer to interior lipids for the exposure duration (Swackhamer and Skoglund 1991)	-Desorption is significantly slower than sorption (Swackhamer and Skoglund 1991)
	-Hydrophobicity	-PCBs preferentially partition to internal neutral lipids, but those are usually a minor fraction of the total lipids, and they vary depending on growth conditions and species (Stange and Swackhamer 1994)	-Highly chlorinated congeners are associated with cell membranes (phospholipids) (Stange and Swackhamer 1994)	-Depuration from algae is very slow (Zaranko et al. 1997)
	-Molecular configuration of the congener	-Diatoms have a sticky mucus layer consisting of polysaccharides (which are polar molecules and may repel hydrophobic compounds) covering the cell surface, whereas flagellates generally lack such a layer; this may explain the significantly lower BAFs for PCBs in diatoms (Kiorboe and Hansen 1993)	-Higher BAF values for PCBs were found in the size fraction defined as phytoplankton, than in the size fraction defined as zooplankton (Magnusson et al. 2007)	-Lack enzymes for dechlorinating PCBs and no metabolism of PCBs (Hill and Napolitano 1997)
	-Growth rate ^a			
<i>Invertebrates (zoobenthos and zooplankton)^b</i>	-Surface area and type			
	-Content and type of lipid in the algae (Stange and Swackhamer 1994)			
	-Pore water (Forbes et al. 1998)		-Zooplankton may accumulate PCBs both across their gut epithelium, via contaminated food, and over their body surface from contact with contaminated water (Magnusson et al. 2007)	-Uptake of PCBs is rapid (Zaranko et al. 1997)
	-Physiological and ecological factors such as selective grazing and predation, different assimilation efficiencies of the contaminated food matrix, and the structure of the pelagic food web (Magnusson et al. 2007)		-The partitioning between sediment carbon and lipids in zoobenthos is referred to as the biota sediment accumulation factor (BSAF ^c) (Magnusson et al. 2006)	-Ingestion rates in deposit feeders can be greater than 100 times body w/d (Forbes et al. 1998), and they feed selectively on fine organic matter, which tends to have higher PCB contamination
			-Uptake of very hydrophobic compounds from sediment was observed to be one to five times greater than that predicted by equilibrium partitioning from pore water (Loonen et al. 1997)	
				-Clearly the important pathway of exposure through the ingested detritus

(continued)

Table 2 (continued)

Organism	Dependent factors	Lipid profile	Bioaccumulation mechanism	Uptake/sorption and desorption
Fish	Fish age, size, and position in the food web (Stow and Carpenter 1994)	The lipid fraction is quite variable among species and even in the same species over the course of a year or lifetime Lipid concentrations in fish have been documented to range from 2 to 27.6% (Gerstenberger et al. 1997; Sijm and van der Linde 1995; Zaranko et al. 1997)	Bioconcentration has been modeled to mean diffusion through aqueous and lipid layer (Sijm and van der Linde 1995)	<ul style="list-style-type: none">–There are two pathways for direct uptake: through the gills and through the skin–Gill exposure is a function of respiration rate, decreasing in larger fish–Dermal uptake can be significant for benthic feeders–For the species at intermediate trophic positions, the most significant loss process was loss by excretion while at higher trophic levels, growth dilution was the dominant loss process (Nfon and Cousins 2007)

^aPhytoplankton biomass may double or triple in 1 d and periphyton turnover may be so rapid that some PCBs will not reach equilibrium, therefore, the term “bioaccumulation factor” (BAF) should be used for plants as well as animals, rather than “bioconcentration factor,” which implies equilibrium (Hill and Napolitano 1997; Stange and Swackhamer 1994).

^bInvertebrates are a critical link in both detrital and phytoplankton food webs. Higher molecular weight and more hydrophobic compounds are incorporated into sediments and are recycled by the zoobenthos (Baker et al. 1991). Lower weight, lower hydrophobicity compounds tend to be dissolved, and their uptake is enhanced in filter feeders through sorption to phytoplankton (Gilek et al. 1996).

$$c_{BSAF} = \frac{tissue\ conc / tissue\ lipid}{sed\ conc / sed\ TOC}$$

where tissue conc is total PCB concentration in tissue; tissue lipid is lipid content; sed conc is total PCB concentration in sediment, and sed TOC is total organic content in sediment

congeners substantially affect accumulation or degradation pathways. Empirical models only reflect one of several possible mechanisms (Antunes et al. 2007).

4 Summary

In recent decades, regulators, academia, and industry have all paid increasing attention to the crucial task of determining how xenobiotic exposures affect biota populations, communities, or entire ecosystems. For decades, PCBs have been recognized as important and potentially harmful environmental contaminants. The intrinsic properties of PCBs, such as high environmental persistence, resistance to metabolism in organisms, and tendency to accumulate in lipids have contributed to their ubiquity in environmental media and have induced concern for their toxic effects after prolonged exposure.

PCBs are bioaccumulated mainly by aquatic and terrestrial organisms and thus enter the food web. Humans and wildlife that consume contaminated organisms can also accumulate PCBs in their tissues. Such accumulation is of concern, because it may lead to body burdens of PCBs that could have adverse health effects in humans and wildlife. PCBs may affect not only individual organisms but ultimately whole ecosystems.

Moreover, PCBs are slower to biodegrade in the environment than are many other organic chemicals. The low water solubility and the low vapor pressure of PCBs, coupled with air, water, and sediment transport processes, means that they are readily transported from local or regional sites of contamination to remote areas.

PCBs are transformed mainly through microbial degradation and particularly reductive dechlorination via organisms that take them up. Metabolism by microorganisms and other animals can cause relative proportions of some congeners to increase while others decrease. Because the susceptibility of PCBs to degradation and bioaccumulation is congener-specific, the composition of PCB congener mixtures that occur in the environment differs substantially from that of the original industrial mixtures released into the environment. Generally, the less-chlorinated congeners are more water soluble, more volatile, and more likely to biodegrade. On the other hand, high-chlorinated PCBs are often more resistant to degradation and volatilization and sorb more strongly to particulate matter. Some more-chlorinated PCBs tend to bioaccumulate to greater concentrations in tissues of animals than do low-molecular-weight ones. The more-heavily chlorinated PCBs can also biomagnify in food webs. Other high-molecular-weight congeners have specific structures that render them susceptible to metabolism by such species as fish, crustacea, birds, and mammals.

In recent years, there has been substantial progress made in understanding the human health and ecological effects of PCBs and their environmental dynamics. However, risk assessments based only on the original PCB mixture that entered the environment are not sufficient to determine either (1) the persistence or toxicity of the weathered PCB mixture actually present in the environment, or (2) the risks to humans and the ecosystem posed by the weathered mixture.

In this paper, we have reviewed the status of current knowledge on PCBs with regard to environmental inputs, global distribution, and environmental fate. We conclude that to know and understand the critical environmental fate pathways for PCBs, both a combination of field studies in real ecosystems and more controlled laboratory investigations are needed. For the future, both revised and new models on how PCBs behave in the environment are needed. Finally, more information on how PCBs affect relevant physiological and behavioral characteristics of organisms that are susceptible to contamination is needed.

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