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Science Support for Evaluating Natural Recovery of Polychlorinated Biphenyl Concentrations in Fish from Crab Orchard Lake, Crab Orchard National Wildlife Refuge, Illinois

Open-File Report 2018–1006

U.S. Department of the Interior
U.S. Geological Survey

Cover. Inset photographs: Kids Fishing Derby at Crab Orchard Lake, Crab Orchard National Wildlife Refuge (CONWR), Illinois. Photographs by Jim Osborn (CONWR volunteer). Background photograph: Sailboat on Crab Orchard Lake. Photograph by Dana Malave-Miller (U.S. Fish and Wildlife Service).

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Abbreviations

BAF	bioaccumulation factor
BCF	bioconcentration factor
BMF	biomagnification factor
BSAF	biota-sediment accumulation factor
PCB	polychlorinated biphenyl
P450	cytochrome P450-dependent monooxygenase system

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Introduction

Crab Orchard Lake in southern Illinois is one of the largest and most popular recreational lakes in the state. Construction of the nearly 7,000-acre reservoir in the late 1930s created employment opportunities through the Works Progress Administration, and the lake itself was intended to supply water, control flooding, and provide recreational opportunities for local communities (Stall, 1954). In 1942, the Department of War appropriated or purchased more than 20,000 acres of land around Crab Orchard Lake and constructed the Illinois Ordnance Plant, which manufactured bombs and anti-tank mines during World War II. After the war, an Act of Congress transferred the property to the U.S. Department of the Interior. Crab Orchard National Wildlife Refuge was established on August 5, 1947, for the joint purposes of wildlife conservation, agriculture, recreation, and industry. Production of explosives continued, but new industries also moved onsite. More than 200 tenants have held leases with Crab Orchard National Wildlife Refuge and have operated a variety of manufacturing plants (electrical components, plated metal parts, ink, machined parts, painted products, and boats) on-site. Soils, water, and sediments in several areas of the refuge were contaminated with hazardous substances from handling and disposal methods that are no longer acceptable environmental practice (for example, direct discharge to surface water, use of unlined landfills).

Polychlorinated biphenyl (PCB) contamination at the refuge was identified in the 1970s, and a PCB-based fish-consumption advisory has been in effect since 1988 for Crab Orchard Lake. The present advisory covers common carp (*Cyprinus carpio*) and channel catfish (*Ictalurus punctatus*); see Illinois Department of Public Health (2017). Some of the most contaminated areas of the refuge were actively remediated, and natural ecosystem recovery processes are expected to further reduce residual PCB concentrations in the lake. The U.S. Fish and Wildlife Service sought technical support to understand environmental drivers of current (2017) PCB residues in fish tissue and patterns in PCB residues through time to inform the fish-consumption advisory for Crab Orchard Lake. This project is planned in two phases (Tasks 1 and 2); the first phase is included in this report.

- Task 1, reported here, includes a review of existing literature and a brief overview focused on environmental and biochemical/physiological processes that drive PCB residues in fish tissue. This review specifically targets processes that are relevant for freshwater lacustrine environments such as those at Crab Orchard Lake. In addition to discussions of environmental fate, metabolism, and accumulation of PCBs, this review includes a brief scientifically based explanation of approaches used to establish fish-consumption advisories.
- A planned second task (Task 2) will include a compilation and summary of existing data on PCB residues in fish tissue samples from Crab Orchard Lake. This summary will also place Crab Orchard Lake data in a broader geographic context through a comparison with fish data from other Midwestern lakes.

When Task 1 and Task 2 are complete, resource managers will have

(a) a synthesis of existing literature that characterizes the processes influencing the fate of residual PCBs remaining in systems such as Crab Orchard Lake,

(b) a summary of natural PCB attenuation processes for use in risk communication with the public, and

(c) a summary of existing data on PCBs in fish tissues from Crab Orchard Lake, including exploratory plots of tissue residues through time.

Overall, this project will provide data to help resource managers better understand the ecological and public health consequences of residual PCBs in Crab Orchard Lake.

Report structure.—In this review (Task 1) we will consider (1) background material on PCB ecotoxicology and PCB-impacted resources at Crab Orchard Lake;

(2) descriptions of environmental fate processes—biodegradation, volatilization and photolysis—that effectively reduce PCB concentrations in sediments and consequently affect exposure to PCBs in field settings at Crab Orchard Lake; (3) a summary of "whole animal" accumulation processes—bioconcentration, bioaccumulation, and biomagnification—that link fish to PCB sources and directly relate to derivation of fish-consumption advisories; (4) a summary of PCB bioaccumulation models applied to fishes; (5) an overview of fish-consumption advisories based on measured or estimated values for PCBs in fish tissue; and (6) an overview of potential natural recovery processes for Crab Orchard Lake.

Background

Polychlorinated biphenyls are a group of more than 200 human-made organic chemicals in which chlorine atoms replace hydrogen molecules in the biphenyl structure. These chemicals were widely used in industrial applications, particularly electrical equipment, due to their thermal stability and electrical resistivity. Individual PCBs (that is, congeners) are distinguished by the number and position of chlorine substitutions. Most PCBs, however, were produced and used as commercial congener mixtures (for example, Aroclor 1242 and Aroclor 1254). In the United States, Monsanto Chemical Company manufactured Aroclors from the late 1920s through 1977 (Voogt and Brinkman, 1989). Despite a ban on PCB manufacture in 1979, the load of PCBs in the global environment has been estimated at more than 370,000 metric tons (Tanabe, 1988), and contamination is widespread in sediments, soils, water, and air. Once released into the environment, PCBs are remarkably persistent; some congeners have half-life times of more than 38 years (Sinkkonen and Paasivirta, 2000).

At Crab Orchard Lake, PCB contamination was primarily associated with an industrial complex known as Area 9 adjacent to Sangamo Bay of the lake. During World War II, the property housed the Illinois Ordnance Plant (fig. 1), which was used to manufacture munitions. The land was later leased to private industrial tenants, and it continues to be used for industry today. Notably, from 1946 through 1962, the Sangamo Electric Company Capacitor Division manufactured PCB-containing electrical equipment at Area 9. A landfill 100 yards south of Sangamo Bay was used in the manufacturing process, and PCBs are assumed to have entered the lake through several intermittent creeks (McKee, 1992). The proximity of the Area 9 building complex and landfill to Sangamo Bay is displayed in figure 2. Early sampling efforts in Crab Orchard Lake detected higher PCB concentrations in sediment, benthic organisms, and fish from Sangamo Bay than from other areas of the lake. The highest concentrations were detected in common carp and channel catfish (Kohler and others, 1990). As part of remedial actions related to the Comprehensive Environmental Response, Compensation, and Liability Act, PCB-contaminated sediments were dredged from Sangamo Bay in 1996. After that action, active remediation in the lake was considered complete, although some PCB residues remained.

Figure 1. Aerial photograph of Illinois Ordnance Plant, looking east (Marion Illinois History Preservation, 2013).

PCBs and Their Metabolism in Fishes and Other Vertebrates

Polychlorinated biphenyls exert a variety of toxicological effects in vertebrates such as fishes (Safe, 1994). Mechanistic toxicity depends on PCB structure; most attention has focused on congeners that activate the aryl hydrocarbon receptor pathway, but others act through a more general narcosis pathway. In general, PCBs are poorly metabolized and slow to excrete. The primary pathway of PCB metabolism is transformation to hydroxylated metabolites through the cytochrome P450-dependent monooxygenase system (P450) pathway; the rate of this transformation determines the biological half-lives of PCBs (Sipes and Schnellmann, 1987). The extent of P450 induction varies with the animal species, as well as the number and position of chlorine atoms on the biphenyl molecule (Skaare and others, 1991). Species-specific differences in the rate of PCB metabolism appear to be driven by differences in the basal level of different P450 isozymes in the liver. Rate differences among individual PCB congeners are dependent on PCB structure. Hydroxylation rates decrease as the number of chlorines increases and as the number of substituted adjacent carbon atoms increases. Highly reactive metabolites in PCB biotransformation subsequently bind covalently to tissue macromolecules or conjugate with glutathione (Sipes and Schnellmann, 1987). Importantly, not all biotransformations are beneficial to the organism because metabolites can be more toxic or biologically active than the parent compound (Parkinson and Safe, 1987; Purkey and others, 2004). Grimm and others (2015) provide an extensive review of PCB metabolism and reactive and stable PCB metabolites.

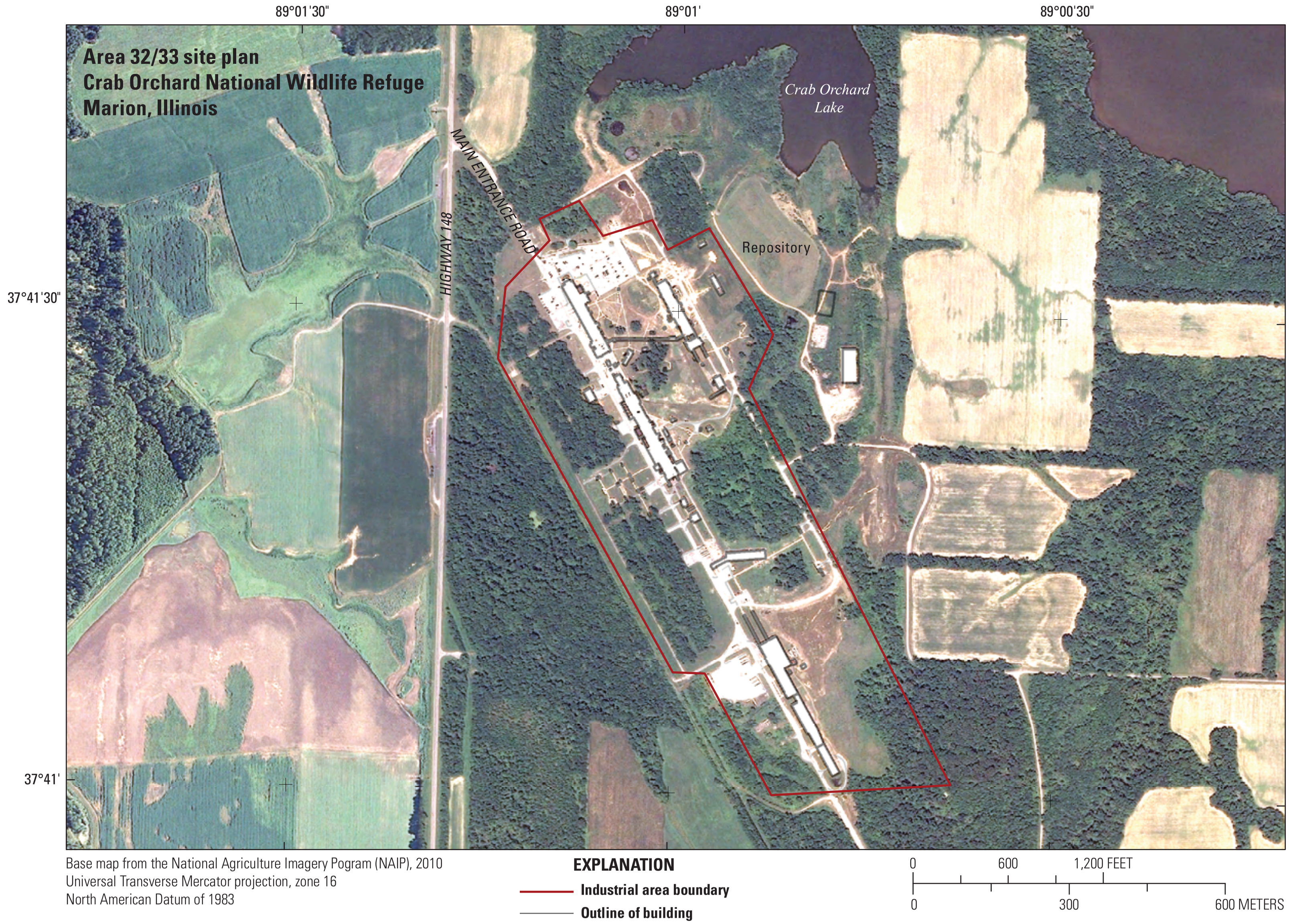

Figure 2. Aerial photograph of the Area 9 building complex and landfill (labeled “Repository”) adjacent to Sangamo Bay of Crab Orchard Lake. Figure courtesy of L. Moore, Crab Orchard National Wildlife Refuge.

In fish, the capacity for PCB biotransformation is considered limited (Matthews and Dedrick, 1984; Boon and others, 1989), particularly when compared to birds and mammals (Buckman and others, 2006). Although fish appear to have some capacity to biotransform and hydroxylate PCBs (White and others, 1997; Wong and others, 2002; Campbell and others, 2003; Li and others, 2003; Wong and others, 2004; Buckman and others, 2006), PCB metabolite concentrations in wild fish tend to be extremely low (for example, Campbell and others, 2003). Therefore, limited rates of biotransformation likely contribute to PCB retention by fish, subsequently increasing PCB exposure in wildlife and humans consuming fish tissue.

Risks of Exposure to PCBs

Residual PCBs in Crab Orchard Lake present toxicity risks to many ecological receptors, including humans. Contaminated sediments are likely the primary source of PCB exposure in Crab Orchard Lake and many other aquatic systems (for example, Gewurtz and others, 2009). Therefore, sediment-dwelling organisms may suffer direct adverse effects of exposure. Sediment-dwelling organisms also consume and concentrate PCBs, thereby transferring sediment-borne PCBs into aquatic food chains, and ultimately terrestrial food chains. These PCBs may bioaccumulate in other invertebrates, amphibians and fish, and predators at higher trophic levels, including humans (Hanrahan and others, 1999; Hinck and others, 2009). Previous reports from the refuge have specifically documented PCB contamination in crayfish (*Diogenes* sp. [presumably *Cambarus diogenes*]; McKee, 1992), frogs (*Rana pipiens sphenocephala* [now *Lithobates sphenocephalus*]; McKee, 1992), several species of fish (Kohler and others, 1990; Straub and others, 2007), birds (*Ardea herodias* and *Tachycineta bicolor*; Maul and others, 2006; Straub and others, 2007), and mammals (*Mustela vison* [now *Neovison vison*]; Hope, 1999).

The trophic accumulation of PCBs and potential subsequent effects on human health are the motivation for fish-consumption advisories at Crab Orchard Lake and other PCB-contaminated sites across the country. Human consumption of PCB residues in fish tissues can result in many potential adverse outcomes, which were reviewed by the Agency for Toxic Substances and Disease Registry (2000). Although most attention on human health issues linked to recreational fishing focuses on sensitive groups of individuals (females of childbearing age, infants, and preadolescent children), consumption of PCB-contaminated fish likely increases risk for cancer in other demographics (for example, Cogliano, 1998). Ingestion of PCB-contaminated fish by adults also can be associated with increased incidence of liver disease and diabetes, and epidemiological studies indicate that exposure to PCBs may impair the immune, reproductive, and nervous systems (ATSDR 2000).

Crab Orchard Lake Fishes and the Fish-Consumption Advisory

In figure 3, we present a conceptual model of PCB exposure pathways in Crab Orchard Lake. This model illustrates transformation and partitioning processes influencing PCB concentrations in sediments and surface water and highlights pathways resulting in wildlife or human consumption of PCB-contaminated fish. Crab Orchard Lake has a very active recreational fishery, in which anglers target largemouth bass (*Micropterus salmoides*), bluegill (*Lepomis macrochirus*), crappie (*Pomoxis annularis* and *Pomoxis nigromaculatus*), channel catfish and white bass (*Morone chrysops*). This fishery is supported by active management, including stocking of largemouth bass and threadfin shad (*Dorosoma petenense*). On the basis of biomass, Crab Orchard Lake fish communities are dominated by gizzard shad (*Dorosoma cepedianum*), but large numbers of common carp also inhabit the lake.

Currently (2017), two fish species are covered by a State of Illinois fish-consumption advisory for Crab Orchard Lake (table 1; Illinois Department of Public Health, 2017)—common carp and channel catfish. Both species are benthivorous and incidentally ingest sediments, which tend to be the primary source of PCB exposure in systems such as Crab Orchard Lake. Because habitat associations and feeding preferences strongly affect exposure to PCBs, we briefly review characteristics of each fish species below. Large numbers of nonnative common carp inhabit reservoirs and other built impoundments in the United States, including Crab Orchard Lake, and display preference for turbid waters (Pflieger, 1975; Trautman, 1981; Pflieger, 1997). Whereas larval common carp primarily feed on zooplankton, juveniles and adults feed on benthic organisms (for example, gastropods, chironomids, and other larval insects), vegetation, detritus, and zooplankton (Summerfelt and others, 1970; Eder and Carlson, 1977; Panek, 1987). Common carp are very active when feeding, disturbing submerged sediments and increasing turbidity. Notably, sediment disturbances may increase the bioavailability of contaminants such as PCBs (Josefsson and others, 2010).

Along with common carp, channel catfish are popular sport fish included in the fish-consumption advisory at Crab Orchard Lake. Although channel catfish are taxonomically distant from common carp, both species feed predominately on prey items tightly linked to sediments; hence, they share routes of PCB exposure that are relevant for the Crab Orchard Lake fish-consumption advisory. Channel catfish generally are considered stream fish associated with fast-flowing, sand- and gravel-bottomed rivers, but they also inhabit a wide range of habitats (including nearly all waters in Illinois). Channel catfish typically feed by touch or taste; the species is much more reliant on olfactory sense (Hara, 1986) than visual cues to stimulate feeding behaviors. Channel catfish are considered bottom-feeding omnivores that consume a highly varied diet of invertebrates (for example, larval and adult insects, mollusks) and plant material. Smaller fish also may be part of their diet. Throughout their range at high-water periods in the

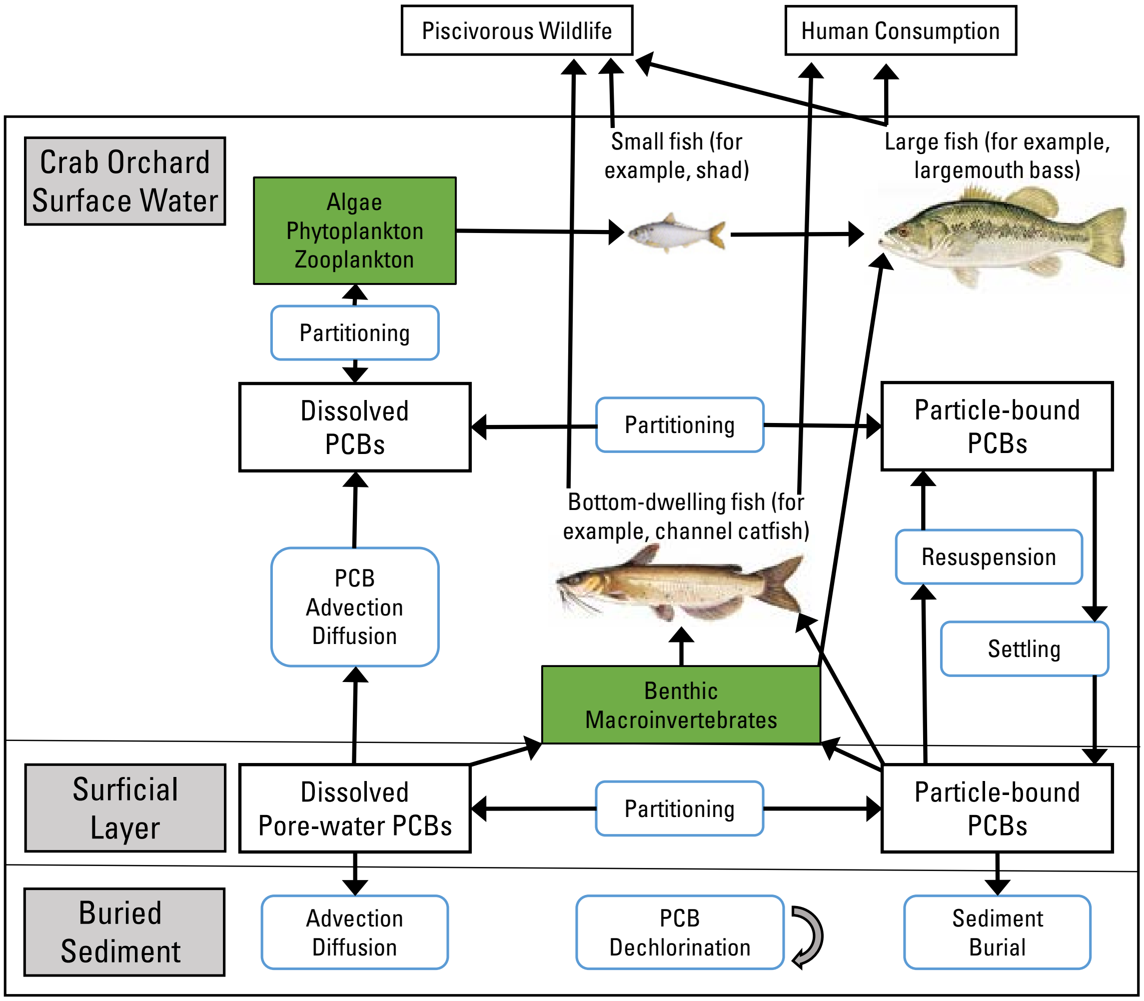

Figure 3. Conceptual model of PCB exposure pathways at Crab Orchard Lake (modified from Magar and others, 2009).

Table 1. Fish-consumption advisory, Illinois Department of Public Health.

[PCB, Polychlorinated biphenyl]

Area under advisory	Crab Orchard Lake, Williamson County		
	Species	**One meal per**	**Contaminant**
East of Wolf Creek Road	Common carp, channel catfish	Week	PCB
West of Wolf Creek Road	Channel catfish	Week	PCB

early spring (March–April), channel catfish may move into the mouths of small tributary streams where they will forage for food.

Until 2016, the Crab Orchard Lake fish consumption advisory also covered one fish not traditionally associated with sediments. Largemouth bass generally inhabit pools and back-water areas in rivers and streams (Trautman, 1981; Wydoski and Whitney, 2003), but also may act as habitat generalists. In contrast to common carp or channel catfish, largemouth bass are piscivores (Heidinger, 1975). However, the species displays a widely ranging diet that potentially includes many species of aquatic organisms, from larval insects to adult frogs and salamanders (Jenkins and Burkhead, 1994). Largemouth bass generally feed in the early morning or late in the day but may feed at other times if prey is available. Largemouth bass were removed from the Crab Orchard Lake fish consumption advisory in 2016, based on apparent declines in PCB tissue residues sampled by the Illinois Environmental Protection Agency (A. Martin, IL Department of Public Health, pers. comm. July 2017).

Environmental Fate and Transport

Abiotic and biotic environmental processes critically affect the fate and transport of chemicals in air, water, or soil; therefore, these processes help determine the environmental concentrations of chemicals to which biological receptors may potentially be exposed.

Chemical stability and environmental persistence are primary characteristics of PCBs. This chemical stability coupled with poor water solubility results in the tendency of these chlorinated organic compounds to accumulate through food chains and become incorporated into tissues of consumers ranging from aquatic invertebrates and fishes to terrestrial vertebrates, including humans. Offsetting these accumulations are environmental degradation processes—primarily microbial transformations—that contribute to dehalogenation of PCBs in freshwater systems, and particularly in freshwater sediments (Brown and others, 1987; Brown and Wagner, 1990; Quensen and others, 1990; Abramowicz, 1994; Fish and Principe, 1994; Bedard and others, 1996; Berkaw and others, 1996). Physical processes also contribute to the degradation of PCBs in the environment or a reduction in their bioavailability. For example, volatilization and photolysis contribute to loss of the parent chemical under some environmental conditions, and adsorption of PCBs to organic matter reduces their bioavailability.

Biodegradation and Transformation

Biodegradation by aerobic or anaerobic microbial activity is the primary process that degrades PCBs in sediments. Complete biodegradation results in mineralization of PCBs to carbon dioxide, water, and chlorine—a process that requires (1) removal of chlorine from the biphenyl ring of the parent PCB structure (for example, through dechlorination), and (2) cleavage and oxidation of resulting intermediate compounds (Boyle and others, 1992). Although the chemical processes vary from species to species, degradation is generally achieved through aerobic oxidative dechlorination, hydrolytic dehalogenation and anaerobic reductive dechlorination (Beyer and Biziuk, 2009). Because different members of microbial communities have different capabilities to degrade PCBs, degradation is most effective in multiple-species communities (for example, in biofilms; Abraham and others, 2002; Macedo and others, 2005). Most relevant microbial activity is bacterial in origin. Several different metabolic processes have been characterized for a wide range of bacteria that are capable of PCB dechlorination (see Brown and Wagner, 1990; Sokol and others, 1994; Bedard and Quensen, 1995; Bedard and May, 1996).

Although PCB biodegradation is a complex process and rates are highly variable, several general patterns should be noted. Aerobic conditions favor the degradation of less chlorinated congeners. As degree of chlorination increases, aerobic biodegradability decreases (Furukawa, 2000). In contrast, anaerobic processes act on the more highly chlorinated PCB congeners. In anaerobic conditions, reductive chlorination transforms highly chlorinated PCBs to less chlorinated congeners. This transformation can yield metabolites that are less toxic than parent molecules and amenable to subsequent degradation by aerobic microbes (Quensen and others, 1990). Importantly, anaerobic conditions are most common in subsurface sediments, which means that reductive dechlorination likely provides more PCB risk reduction in buried sediments than at the sediment-water interface (Magar and others, 2005a; Magar and others, 2005b). Field and Sierra-Alvarez (2008) provide a detailed review of microbial processes involved in the environmental transformation of PCBs.

Volatilization and Photolysis

Volatilization is the dominant pathway of PCB loss from many large lakes (Mackay and others, 1986; Swackhamer and others, 1988; Hornbuckle and others, 1993) and is likely to be a critical process influencing PCB concentrations in Crab Orchard Lake. Volatilization is typically studied at the air-water interface, but volatilization can also deplete PCBs from exposed sediments. Losses of PCBs through volatilization from sediment appear tightly linked with evaporative processes. Early work by Haque and others (1974) indicated minimal volatilization from thin PCB films or dry sediment. In wet sediments, however, Chiarenzelli and others (1996) reported PCB losses of up to 60 percent after just four hours of drying. Therefore, in systems such as Crab Orchard Lake, PCB volatilization is likely to be most pronounced when conditions favor high rates of evaporation—for example, at warmer temperatures; in moist coarse-grained sediments; and in shallow water, particularly in areas subject to repeated cycles of wetting and drying (Chiarenzelli and others, 1996). Volatilization preferentially depletes some congeners (as described by Henry's law constants) and therefore changes the congener profile of remaining PCBs.

Although volatilization may result in loss of PCBs from water and sediment, the net effect of volatilization on PCB loads in aquatic systems can be difficult to predict. Exchange rates between sediments and water and between water and air are critical to this environmental fate process, and atmospheres can be enriched by PCB volatilization from sediment-water sources. Such processes may yield small gains in PCB loads because subsequent deposition from the atmosphere may return PCBs to the aquatic habitat (see U.S. Environmental Protection Agency, 2007). At Crab Orchard Lake, volatilization from the Great Lakes and upwind urban areas may serve as sources of additional PCB deposition.

Although early work identified photolysis as an additional key component in the environmental fate of PCBs, most of these studies evaluated PCB decomposition in controlled laboratory conditions with solvent additions. In natural aquatic habitats characterized by submerged sediments, photolysis will likely have only a seasonal and modest effect on PCB concentrations. In water, photochemistry of PCBs is dominated by hydrolysis in which chlorine is replaced by hydroxide. Polychlorinated biphenyls may also photoisomerize (Pagni and Sigman, 1999), depending on ambient conditions and water quality.

Adsorption to Organic Matter

In addition to biological and physicochemical processes serving in degradation of PCBs, mechanisms such as adsorption affect the bioavailability of parent compounds. In surface waters such as Crab Orchard Lake, the environmental fates of PCBs are driven by partitioning among dissolved and particle-bound phases of the chemical (fig. 3). These phases differ in their bioavailability to aquatic organisms. Because partitioning processes depend on chemical properties such as water solubility and octanol-water partition coefficients (K_{ow}), these properties are helpful in predicting likely environmental fates of PCBs. However, partitioning under field conditions may differ significantly from theoretical predictions (Butcher and others, 1998).

For example, the chemical properties of a PCB congener affect its level of adsorption to organic matter. This adsorption is important, because PCBs adsorbed to dissolved organic matter become less bioavailable (for example, for uptake by gill). Adsorption to particulate organic matter also decreases direct uptake of PCBs, but does not eliminate PCB entry into detrital food webs or ingestion by bottom-feeding fishes (Baker and others, 1991). Geometry of parent compounds also affects PCB fate; for example, planar PCBs are less bioavailable and less likely assimilated by detrital feeders because of their strength of binding with particulate organic matter (Boese and others, 1995; vanBavel and others, 1996).

Quantitative predictions of partitioning, preferably derived from empirically validated models, may yield initial estimates regarding dissolved-sorbed fractions of PCBs, their long-term dispersion in the environment, and their bioavailability to ecological receptors. Such predictions are critical for predicting fish tissue residues and for assessing ecosystem recovery from PCB contamination (Gdaniec-Pietryka and others, 2007).

Processes Involved in Accumulation of PCB Tissue Residues in Fish

Because the development of fish-consumption advisories is based on PCB residues in fish tissue, processes governing the accumulation of PCBs in tissue are especially critical. Scherer and others (2008) provide a technical analysis of advisory development as undertaken by state regulatory agencies. Accumulation of tissue residues of any chemical by fishes can be generalized as a discrete, stepwise process captured by the component processes of (1) bioconcentration, (2) bioaccumulation, and (3) biomagnification. Each of these processes affects fish tissue residues of PCBs in Crab Orchard Lake.

Bioconcentration, Bioaccumulation, and Biomagnification

Aquatic organisms such as fishes, amphibians, or aquatic invertebrates can accumulate high concentrations of PCBs in their tissues relative to PCB concentrations in abiotic compartments of their physical habitat. Because bioconcentration, bioaccumulation, and biomagnification in aquatic organisms have been comprehensively reviewed for organic contaminants (Arnot and Gobas, 2006) and PCBs specifically (for example, Beyer and Biziuk, 2009; Sparling, 2016) we provide a brief overview here.

Bioconcentration

Bioconcentration results from uptake of chemicals directly from water through respiratory surfaces and other epithelial tissues (for example, skin, lining of buccal cavity) and yields a chemical concentration in an organism that is greater than that in the surrounding medium. A bioconcentration factor (BCF) is defined as the ratio of the chemical concentration in biota (*Cb*) to a measure of chemical concentration in water (*Cw*). Both *Cb* and *Cw* may be expressed in different forms—for example, *Cb* as concentration in blood or whole organism and *Cw* as dissolved or total concentration in water. Chemical residue concentrations in fish may be calculated from a simple, one-compartment model, where k_1 is the uptake rate constant and k_2 is the elimination rate constant (Spacie and Hamelink, 1982; appendix 1, eq. 1.1). Ideally, BCF captures steady-state conditions for C_b and C_w—that is, $dC_b/dt=0$ (Neely and others, 1974; Veith and others, 1979)—which yields:

$$BCF=C_b/C_w=k_1/k_2 \quad (1)$$

Although the water-only exposures modeled by BCF calculations are conceptually straightforward, they represent an oversimplification of most aquatic systems (in which significant fish PCB exposures are mediated through the diet) and thus have limited utility in informing fish-consumption advisories. Protective fish-consumption advisories rely on realistic fish tissue residues, which may be underestimated if water-only exposures are modeled in isolation.

Bioaccumulation

Bioaccumulation offers a more complete picture of chemical uptake, including diet and all other exposure routes, and therefore may provide a more realistic characterization of PCB residues in Crab Orchard Lake fishes. Concentrations of total PCBs in fishes are assumed to increase with higher dietary and gill uptake rate constants (k_D and k_1, respectively), and decrease with higher rates of gill elimination, metabolic transformation, fecal egestion, and growth dilution (represented by k_2, k_M, k_E, and k_G; fig. 4). Mathematically similar to BCF, bioaccumulation factor (BAF) represents another ratio estimator of concentration of chemical in biota, C_b, to that in water, C_w. Individual fish are conceptualized as a system of solvents—water, lipid, and nonlipid organic matter—that have corresponding chemical concentrations. Whole body chemical concentrations can then be calculated (appendix 1, eq. 1.2).

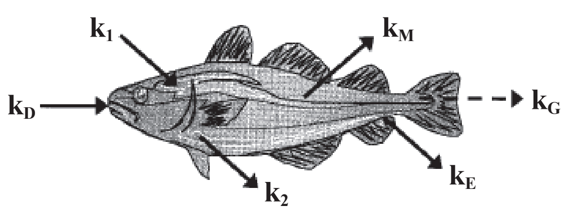

Figure 4. A conceptual diagram representing the primary routes of chemical uptake and elimination in an aquatic organism (from Arnot and Gobas, 2004) © 2004 SETAC.

Provided depuration rates of chemicals from different fish tissues do not differ significantly (Grzenda and others, 1970; Van Veld and others, 1984; Branson and others, 1985; Norheim and Roald, 1985; Kleeman and others, 1986b, a); equilibration among these three phases of a fish is assumed to be rapid in comparison to external exchanges. Hence, for organic chemicals such as PCBs, the calculation can be simplified using partition coefficients between lipid and water and between organic carbon and water (appendix 1, eq. 1.3).

Individual-based models can be scaled up to inform community-based bioaccumulation models, such as the Bioaccumulation and Aquatic System Simulator (BASS; Barber, 2006). The BASS model solves a series of differential equations for each age class of fish in an age-structured community (appendix 1, eq. 1.4), and produces outcomes that are potentially useful as inputs for deriving fish-consumption advisories.

Such models, however, are incompletely validated, particularly for aquatic systems such as Crab Orchard Lake, in which sediments are the primary sources of PCB contamination. An extension of the BAF concept for habitats involving sediment-water systems is the biota-sediment accumulation factor (BSAF; appendix 1, eq. 1.5), that takes into account contaminant concentrations in sediment, as well as the sediment organic carbon fraction. A BSAF approach may be useful in evaluating fish tissue concentrations in light of residual PCBs in Crab Orchard Lake sediments, if field-based data are adequate. These approaches have been used in other freshwater systems contaminated with PCBs (for example, Ankley and others, 1992; Babut and others, 2012; Burkhard and others, 2013), although use has been limited when compared to more traditional bioconcentration or bioaccumulation approaches.

Biomagnification

Biomagnification describes the process by which concentrations of environmental chemicals such as PCBs increase through trophic transfer; that is, from lower levels of a food chain to higher levels. An example in Crab Orchard Lake might take the form of transfer of PCBs from phytoplankton to shad to largemouth bass, and ultimately to piscivorous wildlife or humans. Animals (including humans) feeding at the top of aquatic food chains are most susceptible to chemical exposures mediated through biomagnification. Biomagnification across trophic levels is most pronounced for highly lipophilic and poorly metabolized chemicals. A biomagnification factor (BMF) is expressed as a simple ratio estimator of concentration of chemical in the consumer organism (C_{bc}) to that in their prey (C_{bp}):

$$BMF = C_{bc}/C_{bp} \qquad (2)$$

Given the importance of lipid content in determining body burdens of PCBs, BMF is often viewed relative to octanol-water partition coefficient (K_{ow}) as a lipid-normalized BMF, which represents a multiplication factor above an equilibrium concentration. In general, lipid-normalized BMF values greater than 1.0 indicate biomagnification, whereas values equal to or less than 1.0 do not.

As pointed out by Sparling (2016), we do not have a complete understanding of the relevance of biomagnification factors for deriving fish-consumption advisories. Fish trophic level appears to be an important determinant of PCB tissue residues in some aquatic systems (for example, Rasmussen and others, 1990; Kiriluk and others, 1995). In some fish species, however, feeding location and individual foraging strategies appear to be more important (for example, Lopes and others, 2011). Feeding location may be especially important in systems such as Crab Orchard Lake, where PCB contamination is spatially heterogeneous. For example, PCB concentrations in sediments are higher in the eastern part of Crab Orchard Lake than in the western part, and these differences are reflected in the present fish-consumption advisory (Illinois Department of Public Health, 2017).

Sources of Variability in PCB Tissue Residues in Fish

Concentrations of hydrophobic chemicals such as PCBs in fish tend to be highly variable when measured in tissue residues, even among individuals of the same species. This pattern is noted by Lopes and others (2012) and is also apparent in fish samples collected from Crab Orchard Lake for the past 20 years since lake sediments were remediated. Among-individuals variability is further complicated by the potential for seasonal (Laender and others, 2009) or species-specific (Kiriluk and others, 1995) differences in PCB residues in fish tissues. Such variability may be linked to environmental factors, such as initial quantities and types of PCBs released to the environment, the co-occurrence of other chemicals in the releases, and the initial conditions of the system involved in the release (for example, amount of organic material in the system). Biological factors can also confound interpretation of empirical data on fish tissue residues in Crab Orchard Lake (for example, lipid content and age structure of fish). One additional important source of variability in Crab Orchard Lake fish tissues is the stocking history of fish in the lake, which has typically included tens of thousands of largemouth bass stocked each year at varying size classes, as well as several thousand threadfin shad (IL Department of Natural Resources, pers. comm. February 2015). Differences in size at stocking and the unknown PCB exposure history of these fish may complicate interpretation of tissue residue patterns detected by fish monitoring.

Predictions of PCB effects can also be confounded by differences in fate and effects of individual PCB congeners, particularly when considering organisms at higher trophic levels. As secondary or tertiary consumers, humans are vulnerable to chemicals such as PCBs that biomagnify and their associated potential adverse health effects (for example, Bierman, Jr., 1990). However, not all PCB congeners that bioaccumulate also biomagnify. Degree of chlorination is the primary predictor of biomagnification potential among congeners, but the pattern of chlorine substitution also plays a role. For example, PCB congeners with less orthosubstitution are more metabolically stable in invertebrate and vertebrate tissues (Bright and others, 1995) and are accumulated through food-chain exposures at a greater rate than other congeners with the same degree of chlorination (Campfens and Mackay, 1997). Given that fish-consumption advisories are based on tissue residues of all PCBs combined (that is, total PCBs), these differences among individual congeners may be an important source of "hidden" variability to consider.

Overview of Relevant Bioaccumulation Models

As Arnot and Gobas (2004, p. 2343) observed in their seminal review, "bioaccumulation is a fundamental process in environmental toxicology and risk assessment, because it controls the internal dose of potential toxicants." Not surprisingly then, many useful models of bioaccumulation have been developed, from a water-only exposure system (bioconcentration; see Hamelink and others, 1971) to simple equilibrium partitioning models (Neely and others, 1974; Veith and others, 1979) and two-compartment (that is, organism–water) models reliant on kinetic processes (see Branson and others, 1975; Linder and others, 1985; Gobas and others, 1986; Gobas and others, 1989). Other fugacity-based models (Mackay, 1982) and physiologically based models (Barber and others, 1988; Barber and others, 1991) have been applied in a regulatory setting (see BASS, Barber, 2006 and references cited therein). For additional detail, reference to Arnot and Gobas (2004) is encouraged.

Subsequent models attempted to track chemicals through aquatic food webs and estimate BAFs and BSAFs in realistic environmental conditions. Thomann (1989) and Gobas (1993) have advanced kinetic food web models, and Campfens and Mackay (1997) have focused on a fugacity-based food-web bioaccumulation model. Similarly, early work by Norstrom and others (1976) among others (see Madenjian and others, 1993; Eby and others, 1997; Luk and Brockway, 1997) incorporated bioenergetics into bioaccumulation algorithms. In the past decade, Lopes and others (2012) implemented a food-web modeling approach in support of management of PCB-contaminated sediments.

Fish-Consumption Advisory Implementation

Our focus in Task One has been on the environmental, biochemical, and physiological mechanisms ultimately determining PCB residues in fish tissues. In Task Two, we plan to evaluate if and how fish tissue residues in Crab Orchard Lake have changed during the process of "natural recovery," and how residues compare to trigger values for fish-consumption advisories. Although the derivation and evaluation of fish-consumption advisories is beyond the scope of this document, the following summary is provided as background.

In general, derivation of fish-consumption advisories is based on models of human consumption of fish at given fish tissue residue concentrations. The original model for Crab Orchard Lake was based on fish tissue residues of total PCBs measured from channel catfish, common carp, and largemouth bass. The fish-consumption model generally followed that of Czub and McLachlan (2004), wherein individual exposure assessments for selected sensitive categories within human populations (for example, females of child-bearing age, infants, and preadolescent children) were evaluated with respect to their consumption of fish meals and modelled using standard model conditions. A first-tier evaluation would simplify to a typical food-chain model, including variables for chemical concentration in the edible part of fish, daily consumption rate of fish, and body weight of an individual consumer (appendix 1, eq. 1.6).

Individual exposure assessments rely on empirical data of chemical residues in fish and on human consumption patterns to estimate exposure for hypothetical individuals within given populations. This estimated exposure often takes the form of an estimated chronic daily intake, which expands the basic food-chain model above to include exposure frequency and exposure duration (appendix 1, eq. 1.7). Commonly, default values may be applied for selected human receptors such as females of child-bearing age, infants, and preadolescent children or a default receptor (see appendix 1). A statistically defined value, for example, 95 percent lognormal upper confidence limits will generally be applied for estimates of PCB concentrations in the regularly consumed fish for evaluating different model scenarios. Within a regulatory context (see for example U.S. Environmental Protection Agency, 2000), table 2 summarizes a range of guidance values for total PCB residues in fish. These values determine the number fish meals (one meal=8 oz) recommended as upper limits to assure health safety to a human consumer. Guidance values are provided for noncancer endpoints (chronic, systemic effects) and cancer endpoints (at a 1 in 100,000 risk level).

The U.S. Environmental Protection Agency also provides national recommended water quality criteria for total PCBs in surface water to protect aquatic life (0.014 ug/L) and human health (0.000064 ug/L). Although not a regulatory agency, the U.S. Food and Drug Administration has also identified “action levels” for total PCBs in edible fish tissue at 2.0 ppm (21 CFR 109.30, 2000). Differences between the U.S. Environmental Protection Agency and the U.S. Food and Drug Administration guidance stem, in part, from model differences and the target population driving the analysis.

Because most human exposure to persistent organic pollutants is through the ingestion of contaminated food (Binnington and others, 2014) and particularly fish (Turyk and others, 2012), fish-consumption advisories based on PCBs have been widely implemented around the Great Lakes and in the European Union (European Commission, 2006; Kiljunen and others, 2007). By encouraging reduced consumption of lipid-rich fish, these advisories are intended to limit human exposure to PCBs that can cause neurocognitive (Jacobson and others, 1990; Walkowiak and others, 2001; Stewart and others, 2008) and reproductive impairments (Guo and others, 2000; Hsu and others, 2003; Toft and others, 2004), as well as probable increases in cancer risk (IARC, 2009).

Table 2. Guidance values for fish-tissue residues of total polychlorinated biphenyls (PCBs) in fish to be consumed as a meal (after U.S. Environmental Protection Agency, 2000).

[PCBs, polychlorinated biphenyls; ppm, parts per million; > greater than; <, less than]

Fish meals per month	Noncancer health endpoints	Cancer health endpoints
	Fish tissue concentration, in ppm, wet weight	Fish tissue concentration, in ppm, wet weight
16	>0.006–0.012	>0.0015–0.003
12	>0.012–0.016	>0.003–0.004
8	>0.016–0.023	>0.004–0.006
4	>0.023–0.047	>0.006–0.012
3	>0.047–0.063	>0.012–0.016
2	>0.063–0.094	>0.016–0.023
1	>0.094–0.19	>0.023–0.047
0.5	>0.19–0.38	>0.047–0.094
None (<0.5)[1]	>0.38	>0.094

[1]No consumption of suspect fish recommended.

Natural Recovery as a Risk Management Tool for Crab Orchard Lake

In natural recovery remediation approaches, contaminated sediments are left in place and ecosystem processes are allowed to “contain, destroy, or otherwise reduce the bioavailability of the contaminants” (National Research Council, 1997). These ecosystem processes are critical in the strategy of Monitored Natural Recovery (U.S. Environmental Protection Agency, 2005; Magar and Wenning, 2006; Magar and others, 2009; U.S. Environmental Protection Agency, 2014), but may also act as a complement to other remediation approaches. At Crab Orchard Lake, natural recovery processes are expected to gradually reduce residual PCBs that remained in sediments after dredging. Natural processes that can reduce risk associated with chemical residuals in sediment include:

- biological or chemical transformation of chemical contaminants to less toxic forms;
- sorption or binding of contaminants to the sediment; and
- burial of contaminated materials through deposition of clean sediments.

Of these, biological and chemical transformation are preferred processes because of their potential to completely eliminate the contaminant. For PCBs, however, transformation rates are generally too slow to reduce risk at target time scales (Magar and others, 2009); therefore, sorption and especially deposition of clean sediment are likely to be more important recovery processes in Crab Orchard Lake.

Natural recovery by sedimentation is most successful in net depositional environments, in which contaminated sediments are gradually buried by cleaner sediments. This process results in a decrease in contaminant concentrations in surface sediments and, presumably, reduced exposures of benthic and aquatic organisms to the contaminant. For example, natural burial processes have been effective in reducing PCB concentrations in surface sediments at a Superfund site at Lake Hartwell, South Carolina (Brenner and others, 2004). However, because contaminants are left in place with no engineered containment, they may be resuspended by natural events or human activities that disturb sediments (Eggleton and Thomas, 2004). Also, as was the case at Lake Hartwell, reductions in contaminant concentrations in surface sediments may not result in corresponding reductions in fish tissue concentrations (Brenner and others, 2004), particularly if contaminant sources have not been completely controlled.

A key limitation of natural recovery is that the recovery process is likely slow in reducing risks compared to active engineering remedies. Therefore, natural recovery is most appropriate at sites at which human exposure is low or can be reasonably controlled by institutional controls such as fish consumption advisories during the recovery period (U.S. Environmental Protection Agency, 2005). Institutional controls in general are difficult to implement and manage (Magar and Wenning, 2006), and consumption advisories in particular may be misunderstood or disregarded by the public (for example, Reinert and others, 1991). Despite these weaknesses, fish-consumption advisories have a role to play in protecting public health at Crab Orchard Lake while natural recovery is ongoing.

Summary

In our technical review, we have presented a brief overview of selected PCB topics. Our interest focused, in part, on environmental fate of PCBs, especially a characterization of sediment-water systems in which we considered biodegradation, volatilization, and photolysis. We also reviewed physiological characteristics of fishes potentially exposed to PCBs in sediment and water at Crab Orchard Lake. With these environmental fate and biological processes as background, we summarized available bioconcentration, bioaccumulation, and dietary exposure models that underlie derivation of fish-consumption advisories such as those developed for Crab Orchard Lake.

Given their environmental persistence and potential for bioaccumulation in organisms such as fish, PCBs will remain contaminants of concern for the foreseeable future. In freshwater systems such as Crab Orchard Lake, processes such as volatilization may cause modest decreases in sediment PCB concentrations, although gradual burial through the deposition of cleaner sediments will likely play a more important role in reducing the bioavailability of PCBs. Microbial biodegradation processes can also break down PCBs in sediments under both aerobic conditions (at the sediment-water interface) and anaerobic conditions (in deeper sediment layers). Processes such as sediment resuspension, however, may increase the bioavailability of PCBs.

Aquatic organisms exposed to PCBs may suffer direct adverse effects, but more attention has focused on the consequences of PCB exposures mediated through the food chain. Some PCB congeners (particularly the more heavily chlorinated) are likely to biomagnify, resulting in higher tissue concentrations in organisms feeding at the top of aquatic food webs. This biomagnification is of particular concern for humans consuming contaminated fish. Regulatory controls such as the Crab Orchard Lake fish-consumption advisory are intended to limit the amount of contaminated fish consumed, and thus limit dietary exposure to PCBs. As natural recovery processes decrease overall PCB loads in Crab Orchard Lake, PCB residues in fish tissue are expected to eventually reach a level at which the fish-consumption advisory could be lifted.

Substantial progress has been made in understanding the environmental dynamics of PCBs and their human health and ecological effects. However, the relationship between residual

PCBs in Crab Orchard Lake sediments and concentrations in fish tissue is confounded by many sources of variability. A better understanding of biotic and abiotic factors determining fish tissue residues of PCBs in Crab Orchard Lake will facilitate prediction of tissue residues through time as natural recovery processes are ongoing, and allow better risk communication with the public regarding Crab Orchard Lake fish-consumption advisories.

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Appendix 1. Equations Describing Bioconcentration, Bioaccumulation, and Fish-Consumption Advisory Development

Bioconcentration Factor (BCF)

A bioconcentration factor (BCF) is defined as the ratio of the chemical concentration in biota to a measure of chemical concentration in water. A one-compartment model may be used to calculate the residue concentration in fish, where dC_b is the change in chemical concentration in biota, dt is change in time, k_1 is the uptake rate constant, C_w is the chemical concentration in water, k_2 is the elimination rate constant, and C_b is the chemical concentration in biota (Spacie and Hamelink, 1982):

$$\frac{dC_b}{dt} = k_1 C_w - k_2 C_b \qquad (1.1)$$

Bioaccumulation Factor (BAF)

A bioaccumulation factor (BAF) represents another ratio estimator of concentration of chemical in biota, C_b, to that in water, C_w. To model these exchanges, we must consider how chemicals distribute within the bodies of fish. The whole body chemical concentration of individual fish (C_f) can be expressed as

$$C_f = \frac{B}{W_f} = P_a C_a + P_l C_l + P_o C_o = \left(P_a + P_l \frac{C_l}{C_a} + P_o \frac{C_o}{C_a} \right) C_a \qquad (1.2)$$

where

- B is the body burden of chemical in a fish;
- W_f is live fish weight;
- P_a, P_l, and P_o are fractions of whole fish that are water, lipid, and nonlipid organic material, respectively; and
- C_a, C_l, and C_o are corresponding chemical concentrations.

With the assumption of equilibrium among water, lipid, and nonlipid phases of a fish, this equation simplifies to

$$C_f = \left(P_a + P_l k_l + P_o k_o \right) C_a \qquad (1.3)$$

where

- k_l and k_o are partition coefficients between lipid and water and between organic carbon and water, respectively.

Scaling up the process of bioaccumulation to age-structured fish communities, the Bioaccumulation and Aquatic System Simulator (BASS; Barber, 2006) solves a system of differential equations for each age class of fish, beginning with

$$\frac{dB}{dt} = J_g + J_i - M \qquad (1.4)$$

where

- dB denotes change in chemical body burden, such as that based on tissue-residues;
- dt denotes change in time;
- J_g denotes net chemical exchange across the gills of a fish from the water;
- J_i denotes the same exchange across the intestine from food; and
- M denotes the biotransformation or metabolism of the chemical.

Biota-Sediment Accumulation Factor (BSAF)

The biota-sediment accumulation factor (BSAF) extends the BAF concept for habitats involving sediment-water systems:

$$\mathrm{BSAF} = \frac{\left(C_b / f_{lipid} \right)}{\left(C_{sed} / f_{soc} \right)} \qquad (1.5)$$

where

- C_b is the contaminant concentration in biota,
- f_{lipid} is the lipid fraction of the biota,
- C_{sed} is the contaminant concentration in sediment, and
- f_{soc} is the sediment organic carbon fraction.

Fish Consumption Models

A typical food-chain model can be summarized (U.S. Environmental Protection Agency, 2000) as

$$E_m = \frac{C_m \times IR}{BW} \tag{1.6}$$

where

- E_m is the individual exposure to chemical contaminant m from ingesting fish (in milligrams per kilogram-day; mg/kg-d),
- C_m is the concentration of chemical contaminant m in the edible part of fish (mg/kg),
- IR is the mean daily consumption rate of fish (kg/d), and
- BW is the body weight of an individual consumer (kg).

Derivation of estimates of chronic daily intake (CDI) expands equation 1.6 to include exposure duration and exposure frequency:

$$\text{CDI (mg/kg-d)} = \frac{C \times IR \times FI \times ED \times EF}{BW \times AT} \tag{1.7}$$

where

- C is the concentration of, for example, PCB in tissue (mg/kg),
- IR is the ingestion rate (kg/d or kg/meal),
- FI is the fraction ingested from the contaminated source,
- ED is exposure duration (year; yr),
- EF is exposure frequency (d/yr or meals/yr),
- BW is body weight (kg), and
- AT is averaging time (d).

Fish Consumption Model Default Values

Model values for a default receptor are often applied in the derivation of fish-consumption advisories (for example, *BW*, 70 kg-adult; *IR*, 0.054 kg/d; *FI*, 1.0 [all fish eaten were those from Crab Orchard Lake]; AT_{cancer}, 25,550 d [70 yr at 365 d/yr] or $AT_{noncancer}$, 10,950 d [30 yr at 365 d/yr]; *ED*, 30 yr; and *EF*, 350 d/yr).

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