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ENVIRONMENTAL

AND

HEALTH CRITERIA

FOR

POLYCHLORINATED BIPHENYLS

MONS 059168

I. INTRODUCTION

Polychlorinated biphenyls (PCB's) were long used without regard to their potential long-range effect on the environment. Although used for a period of time as extenders in pesticide formulations, PCB's were not generally considered to be toxic materials. On the basis of mammalian testing data showing acute oral LD_{50} values of 4,000-10,000 mg/kg, such an assumption appears to be valid (1,2,3). It was not until 1966, when PCB's were identified in fish from the Baltic Sea, that these particular chlorinated hydrocarbons became suspect as another potential long-lived environmental contaminant (4,5). Since that time numerous field studies have documented the widespread distribution of these materials in many environmental samples including bottom sediments, soils, and all segments of the biota including man. Subsequent laboratory studies on PCB's have further established their persistent nature, high acute toxicity to some forms of aquatic life, bioaccumulative potential, and chronic toxicity to mammalian, aquatic, and avian species (6). The data generated by both field and laboratory studies indicated that subtle environmental damages were resulting from the uncontrolled use and discharge of PCB's. Initial action to control further environmental contamination came with the cancellation of PCB use as an extender in pesticide formulations. In order to reduce environmental contamination, the sole U.S. manufacturer voluntarily restricted its sales, selling only for use in closed system such as transformers. Thus, there is now general agreement that PCB's are a hazard to the environment.

This document presents data from both field and laboratory studies and integrates the available information into an overall assessment and of the potential hazards presented by PCB effluent discharges into the aquatic environment.

II. Chemical and Physical Properties of PCB's

Since commercial PCB's are chemically undefined mixtures of various isomers and homologs, their chemical and physical properties can best be discussed in terms of the characteristics of these commercial mixtures. PCB's are marketed by the average weight percentage of chlorine in the mixture and this percentage dictates the product's applicability to the desired use. Commercial PCB mixtures which contain 20% to 60% chlorine, by weight, have been the most commonly used products.

PCB's are produced by the chlorination of biphenyls with the crude products purified by distillation. Distillate fractions from various temperature ranges are blended to obtain the individual product mixtures of the desired chlorine content (7). Typically, the specific gravity, boiling point, and melting point of PCB's increase with increasing chlorine content. The following table of vaporization rates exemplifies the changes in physical properties as the chlorine content increases (8).

% Chlorine	Vaporization Rate at 100°C ($g/cm^2/hr$)
21	0.00174
32	0.000874
42	0.000338
48	0.000152
54	0.000053
60	0.000013
62	0.000009

Because of the non-polar chemical characteristics of PCB's, they are generally referred to as "insoluble" (8). Quantitative data on the water solubility of PCB's contain variances which reflect the non-homogeneous nature of the material. A PCB product containing 21% chlorine was found to have solubilities of 5 mg/l and 3.8 mg/l in fresh and salt water respectively, while two PCB products both labeled as containing 54% chlorine were found to have different solubilities. One product was soluble in fresh water at 2-3 mg/l and in salt water at 1-1.5 mg/l, while the other product had a solubility of 0.3-0.5 mg/l in both fresh and salt water. A gas chromatographic examination of the two products revealed that the one displaying higher solubility contained greater amounts of lower chlorinated homologs than did the other (9). Thus, the observed physical characteristics of PCB's are in part dependent on the isomer distribution which may vary from batch to batch. It therefore follows that PCB products should not be characterized soley on the basis of average chlorine content.

The low water solubility reported for PCB's cannot be used to predict the behavior of these materials in the aquatic environment. Non-ionic surface active agents have been found to affect the behavior of PCB's by greatly increasing their water solubility (10,11). The potential presence of surface active agents from municipal and industrial discharges as well as naturally occurring organic constituents of water may increase the solubility of PCB's present in water and bottom sediments.

The flammability of PCB's is related to the degree of chlorination. Increased chlorine content reduces the flammability, and increases the flashpoint. Products with an average chlorine content of 54% are virtually non-flammable and are widely used in explosion-proof transformers. When the sale of PCB's was unrestricted their flammability properties led to commercial use in a variety of fireproof coatings, and as hydraulic and heat-transfer fluids (7).

The low volatility, chemical inertness, and compatibility with plastic resins, paints, and varnishes gave rise to the wide use of PCB's as plasticizers and hardeners (12).

III. Analytical Method

The analysis of PCB's was hampered by two problems. First, PCB's could not be easily detected because of their chromatographic similarity to DDT and its metabolites. Second, commercial PCB mixtures contain a multitude of chemical compounds, making quantification difficult.

In PCB analysis the interference of organo-chlorine hydrocarbons is overcome by sequential column chromatography on Florsil and silica gel (13,14). Gas-liquid chromatography with highly sensitive and selective detectors such as electron capture, microcoulometric titration, and electrolytic conductivity have been employed successfully in the sub-microgram range (15,13). In general, the method compares retention times and peak areas with those of known standards. An outline of the generally accepted analytical method follows:

1. Scope and Application

   The method covers the determination of the following PCB mixtures: Aroclor 1221, 1232, 1242, 1254, 1260, and 1016. It is an extension of the organochlorine pesticide residue method for industrial effluents. The procedure is designed so that determination of both the PCB's and the organo-chlorine pesticides may be made on the same sample. The limit of detection is approximately 1 ug/l for each Aroclor.

2. Summary

   The PCB's and organochlorine pesticides are co-extracted by liquid-liquid extraction, and the two classes of compounds are then separated prior to gas chromatographic determination. A combination of the standard Florisil clean-up procedure and a silica gel microcolumn separation procedure is employed. The measurement of PCB's is accomplished by using an electron capture, microcoulometric or electrolytic conductivity detector. Identification is made from the unique gas chromatographic patterns obtained by the use of two or more dissimilar columns. Several techniques for confirming qualitative identification are available.

3. Interferences

The interferences in industrial effluents may pose great difficulty in obtaining accurate and precise measurement of PCB's and organochlorine pesticides. Separation and clean-up procedures are generally required to eliminate these interferences.

Organochlorine pesticides and other halogenated compounds constitute interferences in the determination of PCB's. Most of these are separated from PCB's by the silica gel microcolumn.

Phthalate esters, certain organophosphorus pesticides, and elemental sulfur will interfere when using electron capture for detection. These materials do not interfere when the microcoulometric or electrolytic conductivity detectors are used in the halogen mode. Sulfur interference can also be minimized by the addition of mercury.

IV. Production, Use and Consumption of PCB's

PCB's have been widely used for both consumer goods and industrial applications for more than 4 decades. PCB's are manufactured in France, Great Britain, Italy, Japan, and Russia, as well as in the United States. At the sales peak in 1970, the sole U.S. manufacturer of PCB's produced 85,054,000 pounds, 73,061,000 pounds of which were sold in the United States. No data are available on foreign production, sales, or imports of PCB's. In 1971, the U.S. producer voluntarily restricted the distribution of PCB's to various industries. Consequently, quantities of PCB's produced and sold decreased to less than one-half those of the previous year.

Prior to 1970, the major uses of PCB's in order of their importance were: dielectric fluids for capacitors, plasticizers, dielectric fluids for transformers, hydraulic fluids and lubricants, and heat transfer fluids. The consumer uses which release PCB's directly to the environment made up about 40% of the total domestic sales prior to 1970 and included such uses as plasticizers, hydraulic fluids and lubricants, surface coatings, inks, adhesives, pesticide extenders, and dye components for carbonless duplicating paper. Transformers and capacitors accounted for most of the remaining 60%. Presently, the PCB's marketed domestically for electrical application account for about 90% of the total quantity manufactured. Again, there is no accounting for the quantity of foreign manufactured PCB's which may be imported into the United States.

With the exception of certain electrical uses, suitable substitutes exist for most PCB applications and in many cases have been utilized since 1971. PCB's are used in explosion-proof transformers where fire protection is particularly important, such as in or near buildings. Transformers containing the PCB fluids are free of the fire and explosion hazards presented by failure of standard petroleum oil-type transformers. Approximately 16.8 million pounds of PCB's were used in transformer dielectric fluids in 1968.

Primarily for safety reasons, PCB's are used in more than 90% of the industrial and utility power capacitors. Fire and explosion hazards from capacitor failure are minimized by the use of PCB's as the dielectric fluid. During 1968, approximately 28.8 million pounds of PCB's were used in capacitors.

Even with voluntary controls placed upon the market by the U.S. producer, the presence of PCB's may continue in some industrial effluents due to stockpiling of materials or equipment with PCB-containing components, imports of PCB's, or the recycling and reprocessing of PCB-containing materials.

V. Sources of PCB's in the Environment

PCB's were first marketed in 1929 and virtually no environmental controls were exercised over the handling or disposal of PCB's until 1971. Although sales data before 1957 are unavailable, it has been estimated that about 500,000 tons of PCB's were marketed in the United States prior to 1971.

Because of the many and diverse uses of PCB's, it is not possible to quantify the various pathways into the environment. It can be shown, however, that PCB's have reached the environment by the following routes: (1) accidental spills; (2) migration into foods from packaging material; (3) open burning or incomplete incineration; (4) direct application as pesticide components; (5) dumping of municipal and industrial wastewater sludges at sea; (6) land disposal of wastewater sludges; (7) vaporization of paints and plastics; (8) municipal and industrial wastewater effluents.

Table I and Table II (6) show the quantities of PCB's found in various municipal and industrial wastewater. From these data it has been estimated that approximately 12 million pounds of PCB's reached the environment each year as wastewater components.

As components of solid waste, PCB's end up in dumps and landfills. Little is known about their concentration, degradation, or leaching from landfills. Although data are sparse, no PCB's were detected in stagnant water close to Swedish landfills, suggesting that little leaching occurs.

TABLE I

PCB'S IN MUNICIPAL WASTEWATER PLANT EFFLUENTS

MUNICIPAL PLANT	AROCLOR COMPOUND DETECTED	CONCENTRATION (ug/l)	FLOW (Mgd)	EST. PCB DISCHARGED lbs/day
Ohio-Miami River				
Dayton	1254	17	48	6.2
Hamilton	1248	10	8	0.6
Middleton	ND*	-	-	-
Wisconsin-Milwaukee River				
West Bend	1254	0.25	1.4	0.002
Fredonia	1254	0.12	0.1	0.002
Saukville	1260	0.13	0.1	0.002
Grafton	1254	0.04	0.8	0.002
California				
East Bay (San Fran.)	1254	3.1-3.8	155	4.2
San Francisco	1254	3.8-5.8	31.5	1.2
Terminal Island	1254	5.8-12.8	9.3	0.7
Orange County	1254	0.21-0.64	130	0.4
Hyperion	1254	0.16-0.37	340	0.7

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TABLE I CONTINUED

MUNICIPAL PLANT	AROCLOR COMPOUND DETECTED	CONCENTRATION (ug/l)	FLOW (Mgd)	EST. PCB DISCHARGED (lbs/day)
White Point	1254	76	350	213
Richmond	ND*	-	-	-
San Diego	ND*	-	-	-
Oxnard	ND*	-	-	-

ND* - Not Detected

1 Mgd - Million gallons per day = (8,000,000 lbs/day)

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TABLE II
PCB'S IN INDUSTRIAL EFFLUENTS

INDUSTRY	AROCLOR COMPOUND DETECTED	EFFLUENT CONCENTRATION (ppb)
Chemical Plant	1242	2.50
Paper Coating	1242 & 1248	27
Paper Treatment	1242	430, 470*
Appliance	1254	5
Appliance	1254	18
Chemical Plant	1254	2.5-275

*Sample from treatment lagoon

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VI. Toxicological

Microflora

Although the bacterial components of certain water bodies may be important in energy production through detritus breakdown, their rapid regeneration and adaptive capabilities make them of little consequence in the overall environmental hazard presented by any toxic chemical. The impact of PCB's on the microflora of a treatment plant is an altogether different aspect and one of possible importance for discharges of PCB's to municipal waste treatment plants. No information is avaivable on the inhibitory action of PCB's on treatment plant seed cultures nor on their degradability under treatment plant conditions. Evidence that PCB's pass through such facilities comes directly from monitored effluents, extractions and analyses of sewage sludge (16) and indirectly from the persistent and bioaccumulative nature of PCB's. In one set of experiments, bacterial cultures were found to significantly reduce the concentration of soluble material in the supernatant, but the mechanism of removal is postulated to be that of physical sorbtion on bacterial cell walls and cellular debris (16, 17).

Thus, some removal may occur under municipal treatment plant conditions, but such physical removal can only be considered as temporary. Sludge disposal by landfill, incineration, or ocean dumping can again release PCB's to the environment. Contact exposure of E. coli agar cultures to PCB's impregnated in filter paper at levels up to 1000 mg/l did not inhibit E. coli growth and 10 and 100 ug/l of PCB's in the agar medium were found to stimulate growth. No significant differences were noted between control and exposed E. coli cultures in terms of RNA and DNA levels or uridine uptake (18).

Primary Producers

Some data are available on the toxic effects of PCB's to the primary producers. The primary producers are those photosynthetic organisms generally referred to as phytoplankton, diatoms, or algae. The data are limited to three species of marine diatoms. Although the experimental conditions dictate bioassays only for a period of days, the lifetime of single-celled organisms such as diatoms and the speed with which they reproduce permit the data to be considered as long-term effects on generations of the plants.

Cylindrotheca closterium cultures exposed to 100 ug/l of a PCB with 42% weight average chlorine content were found to differ significantly in all test parameters (19). The parameters were biomass, cell count, RNA content, and chlorophyl content. Cultures exposed at 10 ug/l were found to vary only in RNA content. In another experiment, the growth rates of two other marine diatoms were measured as a function of PCB concentration. Significant reduction in the growth rates occurred at 25 ug/l for T. psuedonana and at 10 ug/l for S. costatum in 5 to 7 days.

Invertebrates

Bioassays with the marine benthic invertebrate Gammarus oceanicus have been conducted using a PCB with 54% chlorine content solubilized by a non-ionic surface active agent. Controls exposed to the surfactant only were used in all cases and several parameters were measured. In a 30-day chronic exposure under static conditions, 10 to 100 ug/l of the PCB was found to be lethal to the

organisms. Toxicity was evidenced by severe necrosis of certain tissues (20). An interesting observation was that the invertebrate is especially sensitive during periods of moult. In another experiment with the same organism, the time of exposure and concentration of PCB's were varied. Following exposure, the test population was transferred to clean water and observed for up to 30 days for mortalities. The median survival time in days was then calculated and compared to a control population. Significant decreases in median survival time accompanied exposure times greater than one hour at concentrations higher than 1 mg/l. When parents were exposed for 10 hours at 10 mg/l, only 25% of the embryos survived. One hundred percent survival of embryos was observed with exposure at 1 mg/l for the same period of time (10). The data indicate that long-term exposure in the 10-100 ug/l range and short-term exposure in the 1-10 mg/l range can have detrimental effects on this important fish food organism.

Of the estuarine-marine invertebrates directly used by man as foods, toxicity data are available on pink shrimp (Penoceus duorarum), blue crab (Callinectes sapidus), and oyster (Crassostrea virginicus). A PCB mixture containing about 54% chlorine by weight was lethal to 100% of a shrimp test population in 48 hours at a concentration of 100 ug/l. All of the same species survived 48-hour exposure to 10 ug/l (21). Measurements of oyster shell growth when the mollusk was exposed to the same PCB mixture at 1.0, 10, and 100 ug/l showed 19%, 41%, and 100% retardation of shell growth respectively after 96 hours of exposure. In a longer exposure study lasting 20 days, juvenile pink shrimp maintained in sea water at a concentration of 5 ug/l of a 54% chlorine concentration PCB suffered 72% mortality. Blue crabs exposed at the same concentration for 20 days showed no significant increase in mortality over controls (21).

More extensive data are available on the effects of PCB's to freshwater invertebrates. Acute toxicity studies using a variety of benthic macroinvertebrates, with assay times of 4 to 10 days using both flow-through and static bioassay conditions and PCB's of both 42% and 54% chlorine content, are presented in Table III (22).

Flow-through chronic bioassays observing both survival and reproductive success with 3 species of freshwater benthic macroinvertebrates have been reported (15). A 48% chlorine content PCB at a concentration of 5 ug/l was found to be unsafe for the water flea, Daphnia magna. The same PCB mixture at 5 ug/l was tolerated by the scud (Gammarus pseudolimnaeus), while 10 ug/l caused toxic effects. The midge fly larvae, Tanytarsus dissimilis, when exposed to both 48% and 54% chlorine PCB's at 10 ug/l survived and completed emergence. Total inhibition of growth of the same organism was observed following exposure to 30 ug/l of the same PCB mixture.

Fish

Two species of juvenile estuarine fish, the spot (Leiostomus xanthurus) and pinfish (Lagodon rhomboides) were bioassayed under flow-through conditions for time periods up to 56 days with a 54% chlorine content PCB (23). The results were reported as the percent mortality at the end of the test period and are summarized in Table IV. At the termination of chronic exposure, survivors were placed in clean water and held for residue testing which is discussed later. Significantly, the survivors often showed symptoms of chronic intoxication and delayed mortality. In addition, some survivors became diseased

and died during the clean water post-exposure period, thus indicating a predisposition to disease as a result of PCB exposure.

Acute toxicity data for estuarine and marine fish are rather limited. In one experiment with pinfish, concentrations of a 54% chlorine content PCB as high as 100 ug/l had no effect on the mortality rate in 48 hours (21). Exposure to PCB's of 54% and 21% average chlorine content at 2 mg/l for an unspecified period of time proved fatal to Atlantic salmon in the parr stage (19).

Extensive acute and chronic toxicity data for various PCB's are available on a variety of freshwater fish. The acute bioassay data presented in Table V (22) show an interesting trend in acute toxicity levels as a function of PCB average chlorine content. The trend is consistent with all 3 test species and is a strong indication that the lighter or lower chlorinated homologs are more acutely toxic to freshwater fish. The data do not provide any indication as to whether the observed effect is related to intrinsic toxicity differences expressed at the molecular level or to differences in gill cellular transport rates or efficiencies. The same trend is again apparent in the long-term exposure data summarized in Table VI, (22). Where species comparison between Table V and VI can be made, at least one order of magnitude increase in toxicity is observed when the exposure is increased from 4 to 10-15 days.

In a chronic exposure bioassay under flow-through conditions with the fathead minnow as a test organism, at 25°C 8.3 ug/l of a 42% chlorine content PCB was found to be the maximum concentration for survival and reproduction. Identical assay conditions using a 54% chlorine PCB produced a maximum acceptable concentration of 2.8 ug/l (15). These results are the reverse of the trend noted above in Tables V and VI, and indicate that when exposure time is increased to include a reproductive cycle, the higher chlorine content material is the more toxic.

Birds

Most of the data concerning the effects of PCB's on avian species deal with long-term feeding studies and resulting chronic and reproductive effects. In a chronic feeding study lasting 56 days and measuring lethatity, Bengalese finches were fed a PCB diet with the average daily dosage ranging between 6 and 171 mg/kg of body weight. Ninety percent survival was observed at the 6 mg/kg/day level and 60% survival at 101 mg/kg/day (24). Only 2 test subjects were exposed to the maximum dosage of 171 mg/kg/day and the results are not to be considered as significant.

A chronic feeding study with pigeons and bob-white quail at 500 and 250 ppm respectively, noted significant increases in liver weights as well as increased liver microsomal protein and cytochrome levels with both 42% and 54% chlorine content PCB's (25). The observed hepatic changes are similar to those reported for marmalian species (1,2, and 26), and typical of those associated with chronic chlorinated hydrocarbon intoxication (27, 28).

Consideration of the subject of chlorinated hydrocarbon toxicity to avian species immediately raises the much debated point of eggshell thinning and other reproductive effects on predator birds and the eventual impact on entire populations of certain species (27, 28). Much of the information related to those effects is the result of field studies which have attempted to correlate PCB residue levels in parents, eggs, and fledglings with reproductive failure. Such studies are difficult to interpret in a definitive manner since DDT and its metabolites as well as other chlorinated hydrocarbons such as dieldrin are often found in significant quantities (29). Laboratory studies aimed at elucidating the "cause and effect" relationship between PCB residues and reproductive failure have not been conclusive. Pheasants dosed at a rate of 50 mg/week with a 54% chlorine PCB for 17 weeks produced fewer eggs and had higher chick mortality than did the control group. However, no variation in eggshell thickness was observed. Mallard ducks fed a 25 ppm diet of 54% chlorine PCB through 2 breeding seasons or for 5 weeks at dietary levels up to 500 ppm produced eggs with shell thickness equivalent to control groups. Equally negative results were obtained with quail and ring doves at dietary levels of 50 to 10 ppm. PCB's of 42% chlorine content at dietary concentrations of 10 and 100 ppm caused chickens to produce fewer eggs with thinner shells and less hatching frequency than controls. When the dietary level was decreased to 1 ppm, no such effects were observed. The same effects were noted at 100 ppm of a 54% chlorine PCB, while 10 ppm was a "no effect" dosage. Surprisingly, dietary levels up to 100 ppm of a 60% chlorine content PCB produced none of the reproductive effects observed with the lighter homologs (6). Ring doves

exposed to a 10 ppm diet of a 54% chlorine PCB for 3 months showed no significant differences in the number of eggs produced or hatched, or in the number of fledglings. Upon maturation, the offspring of those exposed parents displayed a marked decrease in the number of eggs hatched and young fledged. Cytogenic examination of embryos produced by exposed parents showed an increase in the frequency of chromosome aberrations which may explain the delayed reproductive effects (30).

Two possible mechanisms by which PCB's and other chlorinated hydrocarbons affect eggshell thickness have been advanced and are reviewed in detail elsewhere (19, 27, 28, 29). The one most generally subscribed to is that of induction of hepatic oxidase enzyme systems which alter sex hormone metabolism, thus resulting in rapid hormone degradation and excretion. In the case of avian species, hormone level and excretion apparently exert a direct depleting influence on the calcium reserves which are required for eggshell formation. The second possible mechanism is that of direct inhibitory action by the chlorinated hydrocarbons on the enzyme carbonic anhydrase which is critical to carbonate ion metabolism and the subsequent formation of calcium carbonate, the principal constituent of eggshells (19).

In vitro experiments have demonstrated that PCB's exert approximately 5 times the estradiol hormone degradation potential as does DDT or its metabolite DDE (29, 31). Additional evidence of hormone effects has been demonstrated in males by decreased testicular development and comb growth in cockerels (31).

Mammals

Single oral dose LD_{50}s for various PCB's have been reported in the range of 4,000 - 10,000 mg/kg of body weight for the rat (6). In a controlled study, only the ranging of LD_{50} values was possible since individual variation was extreme and sex-dependent. The data are sufficient to classify PCB's as very low in acute toxicity. The persistent and bioaccumulative nature of PCB's emphasizes the need to examine chronic toxicity and sub-acute effect data. An extensive chronic study on rats, using PCB's of 54% and 60% chlorine content with dietary levels ranging between 20 and 1,000 ppm, documented a variety of histopathological effects after 8 months of exposure (1). Degenerative liver changes observed with both male and female test animals at all dosages of both PCB's included hypertrophy, hyperchromatic nuclei, pleomorphism, cytoplasm vacuoles, and porphyria, all characteristic of chlorinated hydrocarbon intoxication. In some cases, adenofibrosis was observed. Sexual variation was noted in two gross parameters, mortality and liver weight changes. Of the mortalities that occurred during exposure to the 60% chlorine content material, all were female. In nearly all gross pathological examinations, males demonstrated enlarged livers while females did not.

Similar increased liver weight and increased liver lipid content were observed in another study employing male rats (32). Experimental intoxication of a primate, the rhesus monkey, showed bone marrow hypoplasia, increased hepatic microsomal enzyme activity, liver hypertrophy, cytoplasmic lipid droplets, and a proliferation of endoplasmic reticulum. Infants exposed to 35 mg/kg/day gained less weight than did controls, but in general displayed a greater tolerance to PCB's than did adults (26).

Experiments with boars showed hepatic mixed oxidase enzyme induction upon dosages of either 100 mg/kg or 10 mg/kg/day for 10 days, as expressed by altered sex hormone excretions. No effects in hormone metabolism were observed with a single oral dose of 10 mg/kg or multiple doses of 1 mg/kg/day for 10 days (31).

A study of the effects of PCB's containing 21% and 54% chlorine on female rabbits and their reproductive capabilities showed no effect in mating, pregnancy, or weight gain of adults at levels of 1 and 10 mg/kg/day. Vitamin A content and 3 enzyme levels in the liver were assayed in both adults and fetuses. Vitamin A content was found to vary in mothers, but only the 21% chlorine PCB affected fetal livers by increasing Vitamin A content. Of the 3 enzymes, only carboxyesterase showed significant change by increasing in the mothers. No alterations in enzyme activity were detected in either fetuses or placental tissue (33). Although placental transport of PCB's does occur, no changes in fetal livers were detected by the measured parameters.

In a preliminary report, some embryotoxicity was observed with oral PCB dosages of 50 mg/kg administered to pregnant female rabbits (34).

Man

Unfortunately, sub-acute toxic effects of PCB's on humans are known. In October of 1968, an epidemic of a skin disease affecting over 1,000 people was reported in Japan. The cause was soon traced to ingestion of rice oil contaminated with 2-3 g/l of a 48% chlorine content PCB. The estimated average dose was 2 grams with a minimum of 0.5 grams. The most notable symptoms of intoxication were increased pigmentation of skin and nails, acne-like eruptions, increased eye discharge, increased perspiration in the palms, and weakness.

Other symptoms reported included swelling of the limbs, gastrointestinal disturbances, and signs of neurological disorders (35). No fatalities directly attributable to PCB ingestion were reported. An epidemiological study found no significant difference in the severity of intoxication between the sexes. The same study showed a significant difference in clinical severity by age, with the 13-29 year group being the most sensitive. Of the 11 babies born to affected mothers, 2 were stillborn, 9 had dark-brown stained skin, and increased eye discharge was noticeable in most. Growth rates of affected children, as measured by both height and weight gains, were monitored and compared with unaffected classmates. A significant decrease in growth rate was detected in the males after poisoning, while no definite change was found in the females.

More than 3 years after the first symptoms of intoxication appeared, most of the patients were still affected by chronic symptoms of chlorinated hydrocarbon poisoning. Of the 159 patients examined approximately one year after initial symptoms appeared, only 50% showed clinical evidence of improvement, approximately 38% maintained stationary levels, while 12% had, in fact, worsened. The obvious implication is that human detoxification from chronic PCB intoxication is an extremely slow process (35).

The apparent human health threat from chronic ingestion of PCB's prompted the United States Food and Drug Administration to issue proposed limitations on the levels of PCB's in foods, animal feeds, and food packaging material (36).

On July 6, 1973, final regulations were issued (37), which established temporary tolerances for PCB residues arising from unavoidable contamination. These tolerances are:

1. 2.5 parts per million in milk (fat basis).

2. 2.5 parts per million in manufactured dairy products (fat basis).

3. 5 parts per million in poultry (fat basis).

4. 0.5 parts per million in eggs.

5. 0.2 parts per million in finished animal feed for food-producing animals (except the following finished animal feeds: feed concentrates, feed supplements, and feed premixes).

6. 2 parts per million in animal feed components of animal origin, including fishmeal and other by-products of marine origin and in finished animal feed concentrates, supplements, and premixes intended for food-producing animals.

7. 5 parts per million in fish and shellfish (edible portion). The edible portion of fish excludes head, scales, viscera, and inedible bones.

8. 0.2 parts per million in infant and junior foods.

9. 10 parts per million in paper food-packaging material intended for or used with human food, finished animal feed and any components intended for animal feeds. The tolerance shall not apply to paper food-packaging material separated from the food therein by a functional barrier which is impermeable to migration of PCB's.

TABLE III

Test Organism	Type Assay	Duration	Test Parameter	Concentration (µg/l)	% Chlorine by weig
crayfish	static	7 days	LC_{50}	30	42
crayfish	static	7 days	LC_{50}	100	54
crayfish	flow	7 days	LC_{50}	80	54
scud (G. pseudolimnaeus)	flow	4 days	LC_{50}	10	42
scud (G. pseudolimnaeus)	flow	10 days	LC_{50}	5	42
scud (G. pseudolimnaeus)	static	4 days	LC_{50}	52	48
scud (G. pseudolimnaeus)	static	4 days	LC_{50}	2,400	54
glass shrimp	flow	7 days	LC_{50}	3.0	54
dragon fly larvae	static	7 days	LC_{50}	800	42
dragon fly larvae	static	7 days	LC_{50}	1,000	54
damsel fly larvae	flow	4 days	LC_{50}	400	42
damsel fly larvae	flow	4 days	LC_{50}	200	54

Water Conditions: 15.6°C, Alkalinity = 35 ppm, pH = 7.1

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TABLE IV

Test Species	Duration	% Mortality	Concentration (ug/l)	Notes
spot	56 days	0	1 ppb	appeared unaffected
spot	33 days	17	1 ppb	150 fish, 14-16°C, 16-32% salinity
spot	56 days	1	1 ppb	150 fish, 23-32°C, 10-34% salinity
spot	20 days	51	5 ppb	50 fish, S.L.* = 24 mm, 11-18°C, 16-32% salinity
spot	26 days	53	5 ppb	300 fish, S.L. = 24 mm, 8-10°C, 10-32% salinity
spot	45 days	62	5 ppb	50 fish, S.L. = 74 mm, 28-33°C, 23-34% salinity
pinfish	14 days	66	5 ppb	50 fish, S.L. = 30 mm, 16-22°C, 20-32% salinity
pinfish	35 days	41	5 ppb	54 fish, S.L. = 27 mm, 22-32°C, 14-34% salinity

* S.L. = Standard Length

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TABLE V

Test Species	Type Assay	Duration (hrs)	LC_{50}*(mg/l)	% Chlorine	Comments
cutthroat trout	static	96	1.170	21	8.9°C, alk = 139 ppm, pH = 7.6
"	"	"	2.5	32	" " "
"	"	"	5.43	42	" " "
"	"	"	5.75	48	" " "
"	"	"	42.5	54	" " "
"	"	"	60.9	60	" " "
"	"	"	50.0	62	" " "
"	"	"	50.0	68	" " "
channel catfish	"	"	6.00	48	18.3°C, alk = 35 ppm, pH = 7.1
"	"	"	12.00	54	" " "
					" " "
bluegill	"	"	0.278	48	" " "
"	"	"	2.74	54	" " "

* LC_{50} = Median Lethal Concentration

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TABLE VI

Test Species	Type Assay	Duration (Days)	LC_{50}*(ug/l)	% Chlorine	Comments
Rainbow trout	intermittent flow	5	156	54	20°C, alk = 260, pH = 7.4
"	"	10	8.0	54	" " "
bluegill	"	5	154	42	" " "
"	"	10	72	42	" " "
"	"	15	54	42	" " "
"	"	5	307	48	" " "
"	"	10	160	48	" " "
"	"	15	76	48	" " "
"	"	20	10	48	" " "
"	"	10	443	54	" " "
"	"	15	204	54	" " "
"	"	20	135	54	" " "
"	"	25	54	54	" " "
"	"	20	245	60	" " "
"	"	25	212	60	" " "

* LC_{50} = Median Lethal Concentration

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TABLE VI , (Cont'd)

Test Species	Type Assay	Duration (days)	LC50*(mg/l)	% Chlorine	Comments
Bluegill	Intermittent flow	30	151	60	20°C, alk = 260, pH = 7.4
channel catfish	"	10	174	42	" " "
"	"	15	107	42	" " "
"	"	10	275	48	" " "
"	"	15	127	48	" " "
"	"	15	741	54	" " "
"	"	20	300	54	" " "
"	"	25	113	54	" " "
"	"	20	296	60	" " "
"	"	25	166	60	" " "
"	"	30	137	60	" " "
bluegill	"	5	137	48	27°C " "
"	"	10	76	48	27°C " "
channel catfish	"	10	94	48	27°C " "
"	"	15	57	48	27°C " "

* LC50 = Median Lethal Concentration

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VII. Environmental Transport of PCBs

Introduction

When discussing a persistent environmental contaminant such as PCBs, its transport in the environment is of critical concern. Physical transport processes such as vaporization, solubilization, and particulate sorbtion often dictate which of the media will be affected by the material. Chemical transformations such as oxidation, photo-oxidation, and hydrolysis can significantly impact the lifetime and form of the material in the individual media. For these bioaccumulative materials, biological transport and transformation processes are of paramount importance, often dictating which species or trophic level will be the ultimate receptor of the material in toxic amounts.

Physical Transport

Virtually no data exist concerning the rate of physical transport processes. Some data indicative of the extent to which these processes occur are available, but these are usually gathered under controlled laboratory conditions. When extrapolating these data to actual environmental conditions, where a multitude of uncontrolled variables exist, they can only be used in a qualitative manner. Where the effects of environmental conditions on a physical parameter are known, they can be applied, again in a qualitative manner, to generally describe the process.

Transport to the air medium occurs either by direct vaporization or by co-vaporization with water (38,39). Sorbtion of PCBs on suspended solids and soil clays and organic matter can act to significantly slow the vaporization process. PCBs entering the atmosphere as vapor will in all probability maintain their affinity for particulates, and atmospheric precipitation can again

bring air-borne PCB's back to the aquatic and terrestrial environments. A PCB containing 54% chlorine has a vapor pressure nearly equivalent to DDE, and by analogy would be expected to have a soil half-life of approximately 5 years due to vaporization processes (38).

Depending on the chlorine content and the distribution of homologs within each batch, PCB solubilities have been reported as ranging from 0.025 mg/l to 5 mg/l (9, 38). When acute and chronic toxicity data are considered along with bioaccumulation, these solubilities are sufficient to produce damage to the aquatic environment. Environmental factors such as temperature, dissolved solids, turbidity and naturally occurring organic matter all act to alter observed laboratory solubilities. Sorbtion on suspended clay particles can remove PCB's from solution, while organic matter, such as the humic acids, can have the opposite effect of solubilization (10, 38, 40). Under bioassay conditions, a PCB of 54% chlorine content was found to have very little sorbtion affinity and remained in solution, while a 21% chlorine PCB sorbed rapidly, showing a decrease in concentration of from 10 to 6 mg/l in about 5 hours. The primary site of sorbtion was presumed to be the glass of the bioassay test chambers. PCB vapors were detected in the environmental chamber atmosphere. Presumably, co-vaporization was facilitated by aeration within the bioassay tanks (9).

Chemical Transport

One of the properties of PCB's which led to their wide variety of uses was their chemical inertness. Although generally regarded as quite stable, limited photochemical breakdown has been observed with some PCB homologs (29,41). In one observation, ultraviolet irradiation resulted in dechlorination to hydroxyl derivatives, which are presumably further degraded by microbial action (42).

Biological Transport

Biological transport of PCBs was initially indicated by numerous findings of residues in nearly all segments of the biota (6). Laboratory studies have demonstrated bioaccumulation of PCBs by a variety of aquatic plants and animals directly from water and sediments. Feeding studies with mammals and birds have verified the bioaccumulative nature of these chlorinated hydrocarbons in higher animals through oral ingestion. Recognition of the accumulative nature and chronic toxicity of PCBs caused the Food and Drug Administration to issue the tolerances on PCB residues in food sources listed in Part VI of this document.

Bioaccumulation of PCBs has been found in various species representing most levels in the aquatic food web. In an experiment with the marine diatom Cylindotheca closterium exposure to 0.01 and 0.1 mg/l of a 42% chlorine PCB resulted in residue levels of 900 to 1,000 times the level of exposure (19). A variety of fresh water benthic macroinvertebrates exposed to radiolabeled PCBs of 54% chlorine content at concentrations of 1 to 2 ug/l showed magnification factors of 3,000 to 47,000 times depending on species and duration of exposure. The data are tabulated and presented in Table VII(43). Although bioaccumulation by fish food organisms is apparent, the significance of PCB transport via food organisms to fish is in doubt. Direct feeding of a 54% chlorine PCB in dietary levels of 14.5-14,500 ug/kg body weight resulted in coho salmon body residues of only 5 times the exposure level (22). This concentration factor can be compared with the much larger values obtained from whole body exposures presented in Table VIII.

Evidence of direct transport from sediments to the biota was obtained when two filter feeding organisms, the fiddler crab and pink shrimp were exposed

to PCB contaminated sediments under clean flowing water for 30 days. Both species bioaccumulated PCB's directly from the sediments and in proportion to concentration in the substrate (44). Maximum body concentrations were 80 and 240 ppm (wet weight) for the crab and shrimp respectively. These occurred with sediments contaminated to a level of 61 ppm (dry weight basis).

The rate of uptake of PCB's by aquatic organisms is apparently rapid and is maximal in 14 to 28 days for those species tested (6,23,44). Detoxification and excretion of body residue is rather slow as would be expected of bioaccumulative materials. Spot with whole body residues of 27 ppm decreased to 7.2 ppm after 84 days in clean water (23). Oysters at a body burden of 425 ppm still maintained 37 ppm of PCB residue after 16 weeks of exposure to clean water (15). After 4 weeks in clean water, the blue crab, Callinectes sapidus, still retained 50% of accumulated PCB's (6).

Indications of metabolic breakdown by fish are evident in observed shifts of certain PCB peaks between residue and standard chromatograms (15,23). The benthic macroinvertebrate, scud, showed an accelerated selective uptake of the lower chlorinated homologs of a PCB mixture. However, no evidence of metabolic dechlorination was found upon gas chromatographic analysis of residues (43).

Bird and mammal residue studies indicate that ingestion results in rapid accumulation and slow excretion (6,22,25,31,32). Metabolic breakdown has been implied in some avian species by observed shifts in residue gas chromatogram peaks (6,25). Excretion in the eggs and feces of birds (6) and in the feces of mammals has been reported (31).

Although PCB's are generally distributed in all body tissue and organs of fish, birds, and mammals, their high lipid solubility is expressed by a preferential accumulation in body fat tissue and the liver (23,31,32). Rapid accumulation and slow detoxification of orally ingested PCB's are evident in the

Yusho disease epidemic in which, 3 years after intoxication, nearly all patients still suffered chronic symptoms of chlorinated hydrocarbon intoxication (35).

TABLE: VII

Biological magnification of ^{36}Cl-labeled Aroclor® 1254 by aquatic invertebrates

Organism	Organisms per sample[1]	Water concentration ppb $\bar{x}$ ± SE[2]	Organism concentration (4 day exposure) ppm $\bar{x}$ ± SE	Magnification factor[3] 1-day	4-day	7-day	14-day	21-day
Daphnid *Daphnia magna*	60	1.1 ± 0.2	52 ± 2.0	24,700	47,000	-	-	-
Phantom midge *Chaoborus punctipennis*	5	1.3 ± 0.1	30 ± 1.6	22,000	23,000	23,800	24,800	-
Scud *Gammarus pseudolimnaeus*	6	1.6 ± 0.1	39 ± 3.0	17,000	24,000	26,000	27,500	27,000
Mosquito larvae *Culex tarsalis*	10	1.5 ± 0.3	27 ± 2.0	12,600	18,000	20,000	-	-
Grass shrimp *Palaemonetes kadiakensis*	3	1.3 ± 0.1	16 ± 2.6	10,300	12,300	13,700	14,200	16,600
Stonefly *Pteronarcys dorsata*	3	2.8 ± 0.8	7.0 ± 0.30	2,100	2,500	2,800	2,900	2,800
Dobsonfly *Corydalus cornutus*	3	1.1 ± 0.1	5.1 ± 0.23	1,400	4,600	5,700	6,600	6,800
Crayfish *Orconectes nais*	2	1.2 ± 0.1	0.2 ± 0.20	570	1,700	3,400	4,500	5,100

[1] Samples were taken in triplicate.

[2] Samples were taken in triplicate and expressed as mean value ± standard error (P=.05).

[3] Concentration in organism/concentration in water.

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TABLE VIII

Test Species	Exposure Time	Exposure Concentration (ug/l)	Concentration Factor	% Chlorine	Comments	Reference
Eastern Oyster	8 weeks	0.01	160,000	54	Whole body residues(meats)	45
" "	24 weeks	5	85,000	?	Whole body residues(meats)	15
Blue Crab	4 weeks	5	4,600	54	Whole body residues(meats)	6
Pinfish *Lagodon Rhomboides*	2-15 weeks	5	2,800-21,800	54	Whole body residues	45
Spot *Leiostomus Xanthurus*	4-8 weeks	1	17,000-27,000	54	Whole body residues	45
Spot *Leiostomus Xanthurus*	3-6 weeks	5	9,200-30,400	54	Whole body residues	45
Bluegill Sunfish	11 weeks	14.1	52,000	48	Whole body residues	45
Channel Catfish	11 weeks	6.2	47,000	48	Whole body residues	15
Bluegill	11 weeks	7.3	26,300	54	Whole body residues	15
Channel Catfish	11 weeks	6.7	49,500	54	Whole body residues	15

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TABLE VIII (Continued)

Test Species	Exposure Time	Exposure Concentration (ug/l)	Concentration Factor	% Chlorine	Comments	Reference
Grass Shrimp Palaemonetes Pugio	1 week	0.62	2,069	54	Whole body residues	45
" "	2 weeks	0.62	11,920	54	" " "	45
" "	3 weeks	0.62	10,903	54	" " "	45
" "	4 weeks	0.62	17,425	54	" " "	45
" "	5 weeks	0.62	26,580	54	" " "	45
" "	1 week	0.09	3,611	54	" " "	45
" "	2 weeks	0.09	4,800	54	" " "	45
" "	3 weeks	0.09	5,000	54	" " "	45
" "	4 weeks	0.09	17,400	54	" " "	45
" "	5 weeks	0.09	8,355	54	" " "	45
" "	1 week	0.037	1,594	54	" " "	45
" "	2 weeks	0.037	3,405	54	" " "	45
" "	3 weeks	0.037	3,918	54	" " "	45
" "	4 weeks	0.037	4,567	54	" " "	45
" "	5 weeks	0.037	5,729	54	" " "	45

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TABLE VIII (Continued)

Test Species	Exposure Time	Exposure Concentration (ug/l)	Concentration Factor	% Chlorine	Comments	Reference
Pink Shrimp Penaeus Duorarum	2 days	2.5	1,800	54	Whole body residues	45
" "	4 days	2.5	2,760	54	" " "	45
" "	6 days	2.5	6,800	54	" " "	45
" "	9 days	2.5	7,600	54	" " "	45
" "	12 days	2.5	9,600	54	" " "	45
" "	15 days	2.5	15,600	54	" " "	45
" "	22 days	2.5	12,400	54	" " "	45

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VIII. Criteria Formulation

To determine the most sensitive critical receptor which will suffer a detrimental effect from PCB discharges in the aquatic environment, it is necessary to evaluate the potential impact and transport path to each receptor. Drawing on the toxicity and transport data presented above, the impact of PCBs on various life forms in the environment will be assessed.

Microflora

No apparent detrimental effects of PCBs on bacteria have been observed. To the contrary, the data show that concentrations of PCBs up to 100 ug/l are stimulatory to the common intestinal bacteria, E. coli. No disruptions of sewage treatment plants would be expected from discharges to municipal sewerage systems but the non-degradable nature of PCBs assures that such discharges ultimately represent sources to the environment.

Primary Producers

PCB concentrations of 10, 25, and 100 ug/l are inhibitory to growth and productivity of 3 species of marine phytoplankton. Since these organisms are building blocks of the aquatic food web, the transport to phytoplanktonic receptors is direct via water solubilized material. On the basis of the limited data, it can only be concluded that PCB concentrations of 10-100 ug/l can inhibit phytoplankton productivity. The impact of decreased productivity is difficult to assess since the affected organisms are of little direct value to man. The overall environmental consequences can only be generally described as an inhibition of the link between the solar energy source and the aquatic

food web. Some accumulation of PCB's by phytoplankton has been noted but its significance is not clear. The mechanism of uptake is probably sorbtion onto external cutinous material rather than internal, where metabolic disruptions would be expected to occur.

Invertebrates

The aquatic invertebrates can be divided into two groups: those which are food organisms for fish, and those valued by man as food sources. Data on one species of marine benthic macroinvertebrate indicate that this fish food organism can be affected upon long-term exposure to PCB's at 10-100 ng/l. Short-term exposures at substantially higher levels of 1-10 ng/l also demonstrate detrimental effects. Observed effects included acute, chronic, and embryo toxicity. Acute toxicity (4-10 days), as measured by the LC_{50} (50% lethal concentration) on crayfish, scud, grass shrimp, dragon fly larvae and damsel fly larvae, shows that PCB's are impactive on freshwater invertebrates important as fish food organisms in the range of 3-2,400 ug/l, depending on chlorine content, species, and length of exposure. The insect larvae were generally the more tolerant of the test organisms. All tested invertebrates were also efficient concentrators of PCB's.

Of the invertebrates valued by man as food sources, data on the toxicity and bioaccumulation of PCB's exist for 3 organisms, the pink shrimp, blue crab, and oyster. Short-term exposure to PCB's resulted in toxicity to shrimp between 10 and 100 ug/l and inhibition of oyster growth at concentrations as low as 10 ug/l. Long-term exposures at 5 ug/l for 20 days were fatal to juvenile shrimp, but had no effect on crabs. Of perhaps greater significance to man are the bioaccumulation data for these organisms presented in Section VII. Oysters

have shown concentration factors of 85,000 and 160,000 while the pink shrimp showed a maximum concentration factor 15,600 times above exposure level. In addition to being the least sensitive to toxic action, the blue crab is also the least efficient accumulator of PCBs with a reported concentration factor of only 4,600. Although no evidence of accumulation through food chain organisms is available, certain filter feeders have shown bioconcentration from contaminated sediments. Thus, the transport path to invertebrate organisms is direct from either media to the animal and toxic action to critical estuarine receptors can be expected at PCB concentrations of 1 to 10 ug/l. No evidence of direct adverse effects on invertebrates due to bioconcentration of PCBs is available and the fish feeding data minimize oral ingestion as a critical transport mechanism to fish. Mammals (including man) are, however, quite susceptible to PCB intoxication and accumulation via oral ingestion and this aspect of invertebrate bioaccumulation will be discussed later.

Fish

Acute toxicity data presented in Table V in Section VI show a wide range of values depending on the test species and percent chlorine content of the PCB. LC_{50} values for 3 freshwater test species range from 0.28 to 60 mg/l. Chronic exposure data on 2 species of estuarine fish (Table IV) indicate that as little as 5 ug/l produces significant mortality. Long-term exposure data for fresh water fish summarized in Table VI in Section VI show LC_{50} values ranging between 8 and 740 ug/l again depending on the test species, time of exposure, and chlorine content of the PCB. Fish, like the aquatic invertebrates are efficient accumulators of PCBs. Table VIII in Section VII shows concentration factors of approximately 30,000 for estuarine fish and up to 50,000 for fresh water species.

As is the case with the invertebrates, no detrimental effects on fish life have been associated with bioaccumulation. The impact of such bioaccumulation may well center on the documented biological transport of PCBs from fish and aquatic invertebrates to birds and mammals.

Birds

Feeding studies with birds have generated data that largely document sub-acute effects. Dosages of 6-100 mg/kg/day for 2 months resulted in mortalities ranging from 10-40%. Although the sub-acute effects are well documented, feeding or dosage levels required for expression of the effect are quite large. The data pertaining to reproductive impairment are conflicting and consequently cannot, at present, be qualitatively interpreted.

Mammals

As with avian species, much of the mammalian data are directed towards sub-acute effects. Direct application of the data to potential effects on wildlife through an aquatic transport mechanism is not possible.

Man

In depth evaluation of the mammalian toxicity data and its extrapolation to man is outside the scope of this document. Suffice it to say that such in depth studies have been conducted by the Food and Drug Administration in order to establish PCB residue tolerances. Those tolerances for fish and shellfish are 5 mg/kg (ppm) in the edible portion. Using the concentration factors presented in Section VII and the maximum permissible residue levels in fish and

shellfish (5 ppm), it is possible to calculate the water concentration of PCB's that will just achieve the tolerance level. In estuarine water, the oyster is the more efficient concentrator and is a valuable food source. Average concentration factors yield a receiving water value of 0.045 ug/l that could be accumulated in excess of FDA limits. In fresh water, using fish concentration factors, the calculated value is 0.1 ug/l.

Chronic Critical Limit

The calculated values that may exceed FDA residue tolerances for PCB in fish and shellfish destined for human consumption are below those of other receptors. Therefore, the critical receptor of PCB discharges in the environment is man and the critical transport path is bioaccumulation. For the protection of both health and the economic resource represented by commercial fisheries, the chronic critical limits established are 0.045 ug/l in estuarine and marine water and 0.1 ug/l in fresh water.

Acute Critical Limit

For the protection of aquatic life in the vicinity of the discharge from short-term exposure effects, the acute critical limit established is based on 96 hour LC_{50} values. The comparatively low acute toxicity to fish and invertebrates indicates an acute critical limit of 100 ug/l as a threshold level.

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