

Polychlorinated biphenyls (PCB) consist of a mixture of compounds only slightly soluble in water; highly soluble in fats, oils, and nonpolar liquids; and highly resistant to both heat and biological degradation. PCB have a wide variety of industrial uses, primarily as insulating fluid in electrical and heat transfer equipment (Interdepartmental Task Force 1972).

Exposure to PCB is known to cause skin lesions (Schwartz and Pack (1943) and to increase liver enzyme activity that may have a secondary effect on reproductive processes (Risebrough et al. 1968, Street et al. 1969, Wassermann et al. 1970). It is not clear at this time whether the effects are due to PCB or its contaminants, the chlorinated dibenzofurans that are highly toxic (Bauer et al. 1961, Schulz 1968, Verratt 1970). It is also not known whether the chlorinated dibenzofurans are produced by degradation of PCB as well as during its manufacture.

The occurrence of PCB in our waters has been documented repeatedly (New Scientist 1966, Holmes et al. 1967, Risebrough et al. 1968, Jensen et al. 1969, Koeman et al. 1969, Schmidt et al. 1971, Veith and Lee 1971). They have been associated with sewage effluents (Holden 1970, Schmidt et al. 1971) and rainwater (Tarrant and Tatton 1968), as well as releases and leakage. Failures of closed systems using PCB have caused some of the more well known releases (Kuratsune et al. 1969, Duke et al. 1970). It has been reported that the defined treatment process does little or nothing to remove PCB (Ahling and Jensen 1970).

An epidemiological study on severe poisoning by rice oil contaminated with polychlorinated biphenyls in 1968 indicated that about 0.5 grams ingested over a period of approximately one month was sufficient to cause the Yusho disease. Many of those affected showed no signs of relief after about three years (Kuratsune et al. 1969).²¹⁴ Price and Welch (1971) have estimated on the basis of 194 samples that 41-45 per cent of the general population of the U.S. may have PCB levels of 1.0 mg/kg or higher (wet weight) in adipose tissue. Therefore, it appears that PCB may accumulate in the body. On this basis it can be calculated that a daily intake of 0.02 mg would require about 70 years to be toxic. Applying a factor of safety of 10 would permit a daily intake of 0.002 mg, and assuming a two liter per day intake, suggests a permissible concentration in water to be 0.001 mg/l.³¹⁶

However, evaluation of the retention and accumulation of PCB from water instead of oil in humans is highly desirable. A study on rats with a single oral dose of 170 mg/kg showed urinary excretion (of PCB) to be limited, while 70 per cent of the dose was found in the feces during an eight week period (Yoshimura et al. 1971).²²⁴⁻²²⁵ Information on PCB in the diet would also be helpful.

Recommendation

Because too little is known about the levels in waters, the retention and accumulation in humans, and the effects of very low rates of ingestion, no defensible recommendation can be made at this time.

SECTION IV
MARINE LIFE

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organisms also apply to wildlife in order to provide protection in local areas.

POLYCHLORINATED BIPHENYLS (PCB) ^(Cap. here inserted)

Evidence is accumulating that PCB does not contribute to the shell thinning that has been a major symptom of the reproductive failures and population declines of raptorial and fish-eating birds. Dietary PCB produced no shell thinning of eggs of Mallard Ducks (Anas platyrhynchos) (Heath et al. in press 1972), nor did PCB have any effects on eggs of Ring Doves (Streptopelia risoria) (Peakall 1971). A PCB effect could not be associated with the thinning of Brown Pelican (Pelecanus occidentalis) eggshells (Risebrough in press 1972). PCB may increase susceptibility to infectious agents such as virus diseases (Friend and Trainer 1970). Like other chlorinated hydrocarbons, PCB increases the activity of liver enzymes that degrade steroids, including the sex hormones (Risebrough et al. 1968; Street et al. 1968). The ecological significance of this phenomenon is not clear.

Because laboratory studies have indicated that PCB, with its derivatives or metabolites, causes embryonic death of birds (Vos et al. 1970; Vos and Kocman 1970; Vos in press 1972; Peakall et al. in press 1972) and because exceptionally high concentrations are occasionally found in fish-eating and raptorial species (Risebrough et al. 1968; Jensen et al. 1969), it is highly probable that PCB has had an adverse effect on the reproductive capacity of some species of birds that have shown population declines.

Median PCB concentrations in whole fish of eight species from Long Island Sound, obtained in 1970, were in the order of one ⁴⁷ (mg/kg) (Hays and Risebrough 1972), and comparable concentrations have been reported from southern California (Risebrough 1969).⁶¹ On the basis of the high probability that PCB in the environment has contributed to the reproductive failures of fish-eating birds, it is desirable to decrease these levels by at least a factor of two (See Section III on Freshwater Aquatic Life and Wildlife pp.000).²⁰³⁻²¹³

Recommendation

It is recommended that PCB concentrations in any sample consisting of a homogenate of 25 or more whole fish of any species that is consumed by fish-eating birds and mammals, within the same size range as the fish consumed by any bird or mammal, be no greater than 0.5 mg/kg of the wet weight.

In the absence of a standardized methodology for the determination of PCB in environmental samples, it is recommended that estimates of PCB concentrations be based on the commercial Aroclor[®] preparation which it most closely resembles in chlorine composition. If the PCB composition should resemble a mixture of more than one Aroclor[®], it should be considered a mixture for the basis of quantitation, and the PCB concentration reported should be the sum of the component Aroclor[®] equivalents.

DDT COMPOUNDS

DDT compounds have become widespread and locally abundant pollutants in coastal and marine environments of North America.

CHLORINATED BIPHENYLS

Polychlorinated biphenyls (PCB) have been found in fish and wild-

life in many parts of the world and at levels that may adversely affect aquatic organisms (Jensen et al. 1969; Holmes, Simmons, and Tatton 1967; Koener et al. 1969; Risebrough et al. 1968). The environmental occurrence,

uses, and present toxicological aspects of PCB were recently reviewed by Peakall and Lincer (1970), Gustafson (1970), Risebrough (1970), and Reynolds (1971).

Biphenyls may have one to 10 attached chlorine atoms, making possible over 200 compounds (Gustafson 1970). PCB occur as residues in fish, and presumably also in water, as mixtures of chlorinated biphenyl isomers as shown in Table III-15 (Stalling and Johnson, unpublished data 1970, Stalling in press).

Analysis of PCB has been accomplished by gas chromatography after separation of PCB from pesticides. A separation method has been described by Amour and Burke (1970) and modified by Stalling and Huckins (1971). A method using separation on a charcoal column has shown good reproducibility (R. Frank and J. Rees, personal communication). No standardized gas-liquid chromatography method has been proposed for the analysis of mixtures of

PCB in environmental samples. The solubility of these formulations in water has not been precisely determined, but is in the range of 100 to 1,000 $\mu\text{g/l}$ (Pope and George 1970). Since PCB have gas chromatographic characteristics similar to many organochlorine pesticides, they can cause serious interference in the gas chromatographic determination of chlorinated insecticides (Risebrough et al. 1968).

TABLE III-15

Composition of PCB residues in selected fish samples from the 1970 National Pesticide Residue Monitoring Program

River	Location	Species	PCB Residue as Aroclor® type (ug/g whole body)				
			1232	1248	1254	1260	Total
Ohio	Cincinnati, O.	Carp <u>Cyprinus carpio</u>	10	75	42	6.0	133
Ohio	Cincinnati, O.	White crappie <u>Pomoxis annularis</u>	16	17	27	5.6	66
Ohio	Marietta, O.	Channel catfish <u>Ictalurus punctatus</u>	38	23	11	4.9	77
Ohio	Marietta, O.	Channel catfish	16	5.2	13	4.6	38
Yazoo	Redwood, Miss.	Smallmouth buffalo <u>Ictiobus bubalus</u>	72	-	1.4	-	73
Hudson	Poughkeepsie, N.Y.	Goldfish <u>Carassius auratus</u>	9	173	32	-	213
Allegheny	Matrons, Pa.	Walleye <u>Stezostedion vitreum</u> V.	-	5.2	25	4.6	35
Delaware	Camden, N.J.	White perch <u>Roccus americanus</u>	-	8.0	6.8	3.9	19
Cape Fear	Elizabeth Town, N.C.	Gizzard shad <u>Porosoma cepedianum</u>	19	-	2.6	1.1	23
Lake Ontario	Port Ontario, N.Y.	White perch	13	-	4.6	1.2	19
Mississippi	Memphis, Tenn.	Drum <u>Aplodinotus grunniens</u>	11	-	4.5	3.4	19
Merrimac	Lowell, Mass.	Drum	14	75	6.1	3.2	98

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The Monsanto Company, the sole manufacturer of PCB in the

United States (Gustafson 1970), markets eight formulations of chlorinated biphenyls under the trademarks Aroclor[®] 1221, 1232, 1242, 1248, 1254, 1260, 1263, and 1268. The last two digits of each formulation designate the per cent chlorine. Aroclor[®] 1248 and 1254 are produced in greatest quantities. They

are used as dielectric fluids in capacitors and in closed-system heat exchangers

(Papageorge 1970). Aroclor[®] 1242 is used as a hydraulic fluid, and Aroclor[®] 1260 as a plasticizer. Chlorinated terphenyls are marketed under the

trademark Aroclor[®] 5442 and 5460, and a mixture of bi- and terphenyls is designated Aroclor[®] 4465. The isomer composition and chromatographic

characteristics of each formulation have been described by Stalling and Huckins (1971) and Bagley, Reichel, and Cromartie (1970). A contaminant

of some PCB, especially those manufactured in Europe, are chlorinated dibenzofurans (Brungs, personal communication 1972). Although

products it would appear to be extremely toxic, no data are available on their toxicity to aquatic life.

Direct Lethal Toxicity

Studies of toxicity of PCB to aquatic organisms are limited. They

show considerable variation of toxicity to different species, as well as variation with the chlorine content of the PCB. Nevertheless, some trends in the toxic characteristics have become apparent, principally from the work of Mayer as described below:

- the higher the per cent chlorine, the lower is the apparent toxicity of PCB to fish (Mayer, in press). This was found in 15-day intermittent-flow bioassays using bluegills (Lepomis macrochirus) and channel catfish (Ictalurus punctatus) with Aroclor® 1242, 1248, 1254. All LC50 values were in the range 10 to 300 µg/l.
- The bluegill/channel catfish experiments also illustrated that all LC50 values decreased significantly when exposures continued from 15 to 20 days. The 96-hour LC50 of a PCB to fish cannot adequately measure its lethal toxicity.
- The same tests showed that the toxicity of Aroclor® 1248 doubled when the temperature was raised from 20 C to 27 C.

To invertebrates, Aroclor® 1242 has about the same acute toxicity ^{that} it has to fish. In 4- and 7-day tests (Saunders, in press), it killed Gammarus at 42 µg/l, and crayfish (Cambarus) at 30 µg/l, with values that were similar to the 15-day LC50 reported for bluegills. However, there is an extreme range in the reported short-term lethal levels of Aroclor® 1254 for invertebrates. Saunders (in press) reported a 96-hour LC50 as 80 µg/l for crayfish and only 3 µg/l for glass shrimp (Palaemonetes) in 7-day tests; and Duke, Lower, and Wilson, Jr. (1970) reported that as little as 0.94 µg/l killed immature pink shrimp (Panaeus duorarum). Part of this variation is related to exposure periods in the tests; part is no doubt the variation in species response. Again this emphasizes the point that short-term tests of acute toxicities of PCB have serious limitations.

Marine animals may be more easily killed by PCB than freshwater

^{see} (see the Panel report on Marine Aquatic Life and Wildlife, Section IV).
Where estuarine fishes (Lagodon rhomboides and Lefostomus xanthurus) were
exposed for 14 to 45 days to Aroclor ³⁴⁵⁻¹² 1254, mortalities were observed at
5 ^{mg/l} (Hanson, et al. 1971). This indicated a toxicity about
five times greater than summarized above for freshwater fish, but
about the same as the toxicity for the marine crustaceans mentioned above.

Feeding and Studies

Dietary exposure to PCB seems to be less of a direct hazard
to fish than exposure in water. Coho salmon (Oncorhynchus kisutch) fed
Aroclor ¹²⁵⁴ 1254 in varying amounts up to 14,500 $\mu\text{g/kg}$ body weight per day
accumulated
whole body residues which were only 0.9 to 0.5 of the level in
the food after 240 days of dietary exposure. Growth rates were not affected.
However, all fish exposed to the highest treatment died after 240 days exposure;
and thyroid activity was stimulated in all except the group treated at the
lowest concentration (Mehrlie and Grant, ³⁹⁹³⁹⁴ unpublished data 1971). //

9.
At present, evaluation of data from laboratory experiments indicates that exposures to PCB in water represent a greater hazard to fish than dietary exposures. However, in the environment, residue accumulation from dietary sources could be more important, ^{because} since PCB have a high affinity for sediments, and therefore, they readily enter food chains ^{at al.} (Duke, ³⁷¹ Lowe, and Wilson 1970; Nimmo, et al. ³⁸² 1971).

Res in Tissue

It is clear that widespread pollution of major waterways has occurred, and that appreciable PCB residues exist in fish. When analyses of 40 fish from the 1970 National Pesticide Monitoring Program were made, only one of the fish was found to contain less than 1 $\mu\text{g/g}$ PCB (Stalling and Mayer 1972). ³⁷⁰ The 10 highest residue levels in the 40 selected fish ranged from 19 $\mu\text{g/g}$ to 213 $\mu\text{g/g}$ whole body weight.

By contrast, residues measured in ocean fish have been generally ^{below} 1 $\mu\text{g/g}$ (Risebrough ³⁸⁷ 1970; Jensen, et al. ³⁷⁴ 1969). Between the ranges in freshwater fish and those in marine fish are the levels of PCB found in seals (Jensen, et al. ³⁷⁶ 1969; Holden ³⁷⁴ 1970), and in the eggs of fish-eating birds in North America (Anderson et al. ³⁶⁸ 1969; Reynolds ³⁵⁵ 1971; Mulhern et al. ³¹⁶ 1971).

In laboratory experiments, crustacea exposed to varying levels of Aroclor ² 1254 in the water ^{can} concentrate the PCB within their bodies more than 20,000 times. The tissue residues may sometimes reach an equilibrium, and in Gammarus fasciatus PCB did not concentrate beyond 27,000 times despite an

Additional 3-week exposure to 1.6 $\mu\text{g/l}$ Aroclor ³³⁶® (Saunders 1972).
In contrast, PCB residues in crayfish did not reach equilibrium after a
28-day exposure. PCB concentration factors by two estuarine fishes,
Lagodon rhomboides and Leiostomus xanthurus, were similar to that described
for crustaceans, ^{A.C.} about 10,000 to 50,000 times the exposure levels
in water (Hansen et al. 1971). It is important to note that these accumu-
lations occurred at water concentrations of PCB that killed the fish in
15 to 45 days.

Also similar were the accumulation ratios of 26,000 to 56,000 for
bluegills (Lepomis macrochirus) chronically exposed to 2.15 $\mu\text{g/l}$ of Aroclor ¹²⁴⁸®
1248 and 1254. Fathead minnows (Pimephales promelas) chronically exposed to
Aroclor ¹²⁴⁸® 1242 and 1254 for 8 weeks concentrated PCB's 100,000 and 200,000
times the exposure levels, respectively. Residues of 50 $\mu\text{g/l}$ (whole body)
resulted from exposure for 8 weeks to 0.3 $\mu\text{g/l}$ Aroclor ¹²⁵⁴® 1254. These
experiments with bluegills also indicated that the maximum levels of PCB
were generally related to the concentration of PCB in the water (50,000 -
200,000 times higher) to which they were exposed (Stalling and Huckins, ³⁹²
unpublished data, 1971).

Effects on Reproduction

PCB residues in salmon eggs are apparently related to mortality
of eggs. In preliminary investigations in Sweden, Jensen and his associates
(1970) reported that when residues in groups of eggs ranged from 0.4
to 1.9 $\mu\text{g/g}$ on a whole weight basis (7.7 to 34 $\mu\text{g/g}$ on a fat basis), related
mortalities ranged from 16 per cent up to 100 per cent.

PCB concentrations in the range of 5 to 10 $\mu\text{g/l}$ in water interfered
with reproduction of several aquatic animals according to recent work of Nebeker,
Pagnan, and DeFog ³⁸¹ (1971). About 5 $\mu\text{g/l}$ of Aroclor ¹²⁴⁸® 1248 was the highest

Concentration that did not affect reproduction of Daphnia magna and Gammarus pseudolimnaeus. In tests of reproduction by fathead minnows (Pimephales promelas) all died when exposed chronically to greater than 8.3 $\mu\text{g/l}$ of either Aroclor $\text{\textcircled{R}}$ 1242 or Aroclor $\text{\textcircled{R}}$ 1254. Reproduction occurred at and below this same concentration of Aroclor $\text{\textcircled{R}}$ 1242, and at and below 2.8 $\mu\text{g/l}$ of Aroclor $\text{\textcircled{R}}$ 1254.

The association between residue levels and biological effects in aquatic animals is scarcely known, but the work of Jensen, ^{et al.} Johansson, Olsson (1970) suggested that about 0.5 $\mu\text{g/g}$ of PCB in whole salmon ^{377/} might be the threshold for egg mortality. Such a level in eggs would be associated with levels in general body tissue (e.g., muscle) of 2.5 to 5.0 $\mu\text{g/g}$. The residue in muscle corresponded to the present Food and Drug Administration level for allowable levels of PCB in fish used as human food. Residues measured in the survey by the 1970 National Pesticide Monitoring Program were generally above 5 $\mu\text{g/g}$.

Applying a minimal safety factor of 10 for protection of the affected population, and for protection of other species higher in the food chain, would yield a maximum permissible tissue concentration of 0.5 $\mu\text{g/g}$ in any aquatic organism in any habitat affected by PCB.

General Considerations and Further Needs

Another means of control would be justified in view of the toxicity of PCB, the lack of knowledge about how it first enters natural ecosystems as a pollutant, and its apparent distribution in high concentrations in freshwater fish in the United States. This method would be to regulate the manufacture of PCB and maintain close control of its uses to avoid situations where PCB is lost to the environment. The Monsanto Company recently restricted the sale of PCB for uses in which disposal of the end products could not be controlled, as with plasticizers (Gustafson,

BA 514 for Recommendations

For PCB levels in water, the most sensitive reaction shown by

aquatic organisms is to the lethal effects of low concentrations con-
tinually present in water for long periods (weeks or months). Concen-
trations in the range of 1 to 8 $\mu\text{g/l}$ have been shown to be lethal to several

animals.
 (1) The work of Hansen, et al. (1971) and
 Illing and Huckings (unpublished data, 1971) indicates that concentrations

of 0.01 $\mu\text{g/l}$ of PCB in water over periods of up to 36 weeks could lead to
dangerous levels of PCB in the tissues of aquatic organisms. Accumulation
 by factors of 75,000 to 200,000 times is indicated by their work. If the
higher ratio is taken, 0.01 $\mu\text{g/l}$ in water might result in 2.0 $\mu\text{g/g}$ in
fish on whole fish basis. This is comparable to the residue level in
eggs associated with complete mortality of embryos. Therefore, a
 concentration is recommended that is reduced by a factor of 5, or 0.002 $\mu\text{g/l}$.

In addition, a control based on residue levels is required, as well as one
 based on PCB in the water.

Recommendations

Aquatic life should be protected where the maximum concentration

of total PCB in unfiltered water does not exceed 0.002 $\mu\text{g/l}$ at any time or
place, and the residues in general body tissues of any aquatic organism do
not exceed 0.5 $\mu\text{g/g}$.