

Gas Chromatographic Characteristics of Authentic Chlorinated Dibenzofurans; Identification of Two Isomers in American and Japanese Polychlorinated Biphenyls

Retention times of 15 authentic standards of chlorinated dibenzofurans containing from three to six chlorine atoms were determined relative to dieldrin on gas chromatograph columns of four different polarities (3% OV-1; 5% SP-2401; 1.5% OV-17/1.95% QF-1; and 3% OV-1/5% OV-210/8% OV-225). On the basis of coinjections of the standards

with extracts from American (Aroclor) and Japanese (Kanechlor) polychlorinated biphenyls (PCB), two of the compounds in those extracts have been identified as 2,3,7,8-tetra- and 2,3,4,7,8-pentachlorodibenzofuran. The concentrations of the compounds were estimated to range from 0.08 to 0.83 µg/g of PCB.

Vos et al. (1970) have identified toxic components of European polychlorinated biphenyls (PCB) as two chlorinated dibenzofurans. Subsequently, several additional chlorinated dibenzofurans were found in the German preparation examined by Vos et al. (Bowes et al., 1973). Further investigations have led to the detection by high-resolution mass spectrometry of those contaminants in American (Aroclor) PCB (Bowes et al., 1975). A recent study reports unidentified tri-, tetra-, and possibly pentachlorodibenzofuran components in Kanechlor 400 (Roach and Pomerantz, 1974). Kuratsune et al. (1972) have identified Kanechlor 400 as the agent responsible for Yusho, a toxic syndrome in humans who have ingested contaminated rice oil.

The reported high toxicity of the chlorinated dibenzofurans (Vos et al., 1970) and their presence in PCB have prompted synthesis of authentic standards and determination of their effects on induction of the enzyme aryl hydrocarbon hydroxylase (Kende et al., 1974). The present paper reports gas chromatographic characteristics of those standards, and the identification of two isomers in American and Japanese PCB based on co-injections of the standards with extracts from the PCB.

EXPERIMENTAL SECTION

Synthesis of Chlorinated Dibenzofurans. Authentic dibenzofuran derivatives (Table I) were synthesized by regioselective methods and characterized by proton magnetic resonance and mass spectrometry (Kende et al., 1974).

Extraction of Chlorinated Dibenzofurans from PCB, and Quantification. Aroclor 1248 and 1254 preparations (obtained in 1969 from Monsanto) were placed on Florisil columns, eluted in solvents of increasing polarity, and partitioned over alumina as described elsewhere (Bowes et al., 1975). The latter separation yielded mixtures of chlorinated dibenzofurans essentially free of PCB interference. Chromatograms of extract fractions resulting from use of these techniques are illustrated in Bowes et al. (1975) for Aroclor 1254 and in Bowes et al. (1973) for Clophen A-80, a German PCB. Coinjections of authentic standards were made with one or more fractions recovered from the Florisil column (and passed over alumina) for each PCB. All chlorinated dibenzofurans usually were found in the first three fractions. Once coincidence of authentic dibenzofuran peak with extract peak was established on the four GC columns (described below) for one or more fractions from each PCB, isomer concentrations were estimated for each fraction by assuming a peak having the same retention time as the dibenzofuran isomer is that compound, and summing the concentrations for all fractions if the peak occurred on the chromatogram (the OV-1 column was used for this basis of comparison). Compounds were quantified with the use of standard curves based on injections of different amounts of the appropriate dibenzofuran isomer.

For the present study, the same extraction and partitioning techniques were applied to Kanechlor 200 and 500.

Compounds having the same gas chromatographic retention times on the OV-1 column as compounds identified previously by mass spectrometry as chlorinated dibenzofurans in the Aroclor PCB were isolated as mixtures also from the Japanese PCB. The gas chromatographic techniques described for the Aroclor extracts (co-injection and quantification) were used for the Kanechlor extracts.

Gas Chromatography. Concurrent with the foregoing analyses, 15 chlorinated dibenzofurans containing from three to six chlorine atoms were injected into gas chromatographs having electron-capture detectors. The retention times relative to dieldrin were carefully determined ($\pm 1\%$) on four columns of different polarities (Thompson et al., 1969; Canada, Department of National Health and Welfare, 1973).

Instruments and chromatographic conditions are listed below: (1, Micro Tek 220): (a) column: 3% OV-1 on 100-120 mesh Supelcoport, 6 ft $\times$ 4 mm i.d., glass; (b) detector: ^{63}Ni ; (c) detector, inlet, and oven temperatures, 273, 225, and 195°, respectively; (d) column flow: 50 cm³ of N₂/min; (e) purge flow: 55 cm³ of N₂/min; (2, Micro Tek 220): (a) column: 5% SP-2401 (= QF-1, Supelco, Inc., Bellefonte, Pa.) on 100-120 mesh Supelcoport, 6 ft $\times$ 4 mm i.d., glass; (b) detector: ^{63}Ni ; (c) detector, inlet, and oven temperatures: 273, 225, and 200°, respectively; (d) column flow: 55 cm³ of N₂/min; (e) purge flow: 55 cm³ of N₂/min; (3, Varian 1440): (a) column: 1.5% OV-17/1.95% QF-1 on 100-120 mesh Chromosorb W, acid washed, DMCS treated, 6 ft $\times$ 2 mm i.d., glass; (b) detector: ^3H ; (c) detector, inlet, and oven temperatures: 225, 230, and 192°, respectively; (d) column flow 35 cm³ of N₂/min; (4, Varian 1440): (a) column: 3% OV-1/5% OV-210, 5 ft $\times$ 3% OV-1/8% OV-225, 0.5 ft on 60-80 mesh Chromosorb W, acid washed, 4 mm i.d., glass; (b) detector: ^3H ; (c) detector, inlet, and oven temperatures: 232, 220, and 213°, respectively; (d) column flow: 40 cm³ of N₂/min.

RESULTS AND DISCUSSION

Retention times of the 15 authentic dibenzofurans relative to dieldrin ranged from 0.68 to 5.37 (Table I); these are within the range of retention times of the common chlorinated pesticides and PCB. The gas chromatographic parameters are the same as those used for the pesticides and PCB.

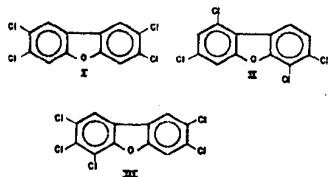
Individual coinjections of 2,3,7,8-tetra- and 2,3,4,7,8-pentachlorodibenzofuran with fractions isolated from Aroclor 1248 and 1254 and Kanechlor 200 and 500 showed coincidence of isomer retention time with peaks in the fractions on all four columns, with the exception that on the OV-17/QF-1 the 2,3,7,8 compound eluted in an unresolved region between two large peaks in the Aroclor 1254 fraction. The retention time of 1,3,6,7-tetrachlorodibenzofuran also coincided upon coinjection with that of a peak in the isolates from all four PCB on the OV-1 and SP-2401 columns, but definitive peak matches on the other two column sys-

Table I. Gas Chromatograph Retention Times of Authentic Chlorinated Dibenzofurans Relative to Dieldrin

Structure of compd	Column			
	OV-1	SP-2401	OV-17/QF-1	OV-1/210/225
2,3,8-Cl ₃	0.82	0.68	0.83	0.77
2,3,6-Cl ₃	0.85	0.72	0.88	0.80
2,3,4-Cl ₃	0.87	0.71	0.90	0.81
1,3,7,8-Cl ₄	1.26	0.89	1.21	1.02
1,3,6,7-Cl ₄	1.29	0.92	1.26	1.09
2,3,6,8-Cl ₄	1.42	1.10	1.74	1.28
2,4,6,7-Cl ₄	1.42	1.18	1.59	1.32
2,3,7,8-Cl ₄	1.57	1.28	1.70	1.30
2,3,6,7-Cl ₄	1.64	1.34	1.92	1.58
3,4,6,7-Cl ₄	1.68	1.47	1.96	1.64
1,2,3,7,8-Cl ₅	2.70	1.84	3.08	2.15
1,2,3,6,7-Cl ₅	2.74	1.87	3.18	2.20
2,3,4,7,8-Cl ₅	2.96	2.29	3.66	2.66
2,3,4,6,7-Cl ₅	3.04	2.39	3.80	3.01
1,2,3,6,7,8-Cl ₆	4.13	3.11	5.37	3.71

tema were not possible because of interference from overlapping peaks.

On the basis of the procedures described, 2,3,7,8-tetrachlorodibenzofuran (I) and 2,3,4,7,8-pentachlorodibenzofuran (III) appear to be present in Aroclor 1248 and 1254 and Kanechlor 200 and 500. The presence of the 1,3,6,7 isomer (II) in the PCB cannot be considered established to the same extent. The retention times relative to dieldrin on an OV-1 column for a tetra- and a pentachlorinated dibenzofuran identified in Clophen A-60 (Bowes et al., 1973; note Figure 1, peaks B and E) are the same as those reported here for the 2,3,7,8- and 2,3,4,7,8-dibenzofuran isomers (Table I).



Amounts of chlorinated dibenzofuran isomers calculated to be present in the PCB were estimated to range from 0.08 to 0.83 µg/g (Table II). Although there is no evidence that chlorinated dibenzofurans are present in wildlife, these data allow one to estimate their potential concentrations. If the dibenzofurans were present in tissue in the same amounts relative to the PCB, the range of concentrations of the dibenzofuran isomers identified here would be approximately 25 to 250 pg/g fresh weight in Great Lakes herring gull (*Larus argentatus*) livers found to contain 300 ppm of PCB (Bowes et al., 1973).

Chicks fed 5 µg/kg per day of 2,3,7,8-tetrachlorodibenzofuran died within 8–15 days (Goldstein et al., 1974). The toxicity to chicks is therefore similar to that reported by Schwetz et al. (1973) for 2,3,7,8-tetrachlorodibenzo-p-dioxin. The latter compound is known to be teratogenic (Schwetz et al., 1973); the teratogenic potential of the chlorinated dibenzofurans has not been established. Chick em-

Table II. Concentration of Chlorinated Dibenzofuran Isomers in PCB (µg/kg of PCB)

Structure of compd	PCB			
	Aroclor 1248	Aroclor 1254	Kanechlor 200	Kanechlor 500
2,3,7,8-Cl ₄ (I)	0.33	0.11	0.10	0.19
2,3,4,7,8-Cl ₅ (III)	0.83	0.12	0.10	0.08

bryo bioassays have demonstrated that the ability of 2,3,7,8-tetrachloro- and 2,3,4,7,8-pentachlorodibenzofuran to effect induction of aryl hydrocarbon hydroxylase is comparable to that of 2,3,7,8-tetrachlorodibenzo-p-dioxin (Kende et al., 1974); this property was characteristic of all polychlorodioxins tested (Poland and Glover, 1973). Our findings point to compounds I and III as possible sources of toxicity of commercial PCB.

ACKNOWLEDGMENT

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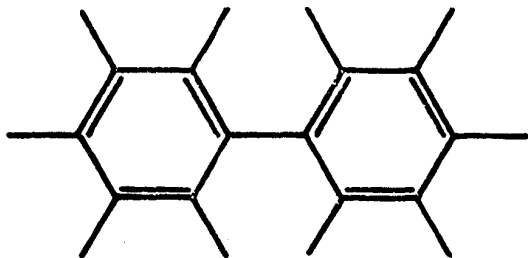
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The accumulation continues

POLYCHLORINATED BIPHENYLS (PCBs) were first introduced into commercial use more than 45 years ago, and for a long time the widely used industrial chemicals were considered to be relatively non-toxic. Their potential threat to the environment, however, was recognized in 1966, when the Swedish scientist, S. Jensen, observed the presence of the compounds in fish and wildlife samples while analyzing for chlorinated hydrocarbon pesticide residues.^{1,2} Since that initial discovery, PCBs have been detected in numerous aquatic and animal species and have been found in high concentrations in industrial waste discharges, river-bottom sediments, food packaging materials, and in foods such as poultry, fish, and dairy products. Detectable levels of the chemical substance have been found in marine life ranging in size from small (plankton and zooplankton)^{3,4,5} to large species (tuna).⁶ High levels of PCBs have been found in fish-eating and predatory birds,^{7,8,9,10} demonstrating the process of concentration of the substance along the food chain from lower to higher animals,¹¹ a process which unfortunately has become all too well known in the era of modern industrial and agricultural activity. Relatively large concentrations of PCBs also have been discovered in the eggs of wild birds.^{6,9,12}

During the past ten years, PCB residues have been analyzed in wildlife and fish samples in Holland, Germany, and the Baltic Sea.^{7,13} Off the coast of Scotland, PCB residues have been found in several marine species, the contamination presumably having arisen from sewage wastes dumped into the open sea.¹⁴ Nevertheless, despite global incidence of the chemical in the environment, residues are generally found mostly in regions of the world where there is intense industrial and urban activity. In the U.S., PCBs in increasing amounts have been detected in the eastern and western coastal regions, in highly urbanized areas (for instance, in and around the Chesapeake Bay, San Francisco Bay, and Puget Sound), and in regions where industrial discharges are known to occur, such as Escambia Bay, Florida, and the Hudson River.^{8,15,16,17}

The environmental accumulation of PCB residues continues despite attempts by both industry and the federal government to improve the situation. As will be explained, industry's countermeasures have been

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limited, and government action has been hesitant.

In the U.S., the impact of PCBS on the environment is best illustrated by ever increasing contamination of sport and commercial fisheries in the Great Lakes region and elsewhere. According to data from the Michigan Department of Natural Resources, levels of PCBS in lake trout and Coho salmon from Lake Michigan showed a sharply increasing trend since 1972. By 1974, the values ranged from more than 10 to nearly 25 parts per million (ppm), well above the present Food and Drug Administration's (FDA's) maximum allowable concentration of 5 ppm for fish and shellfish.¹⁸ In sharp contrast, DDT levels in the same species of fish showed a declining trend since 1970, when DDT consumption in the U.S. was curtailed.¹⁹ Not only are the ecological consequences of continued PCB contamination onerous, but the potential economic implications are serious, since the total annual value of sport fishing in Lake Michigan is as high as \$46.5 million.¹⁹ In addition, the commercial catch of fish is now worth about \$2.5 million each year.²⁰

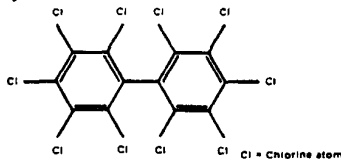
The Michigan studies are supported by studies made by the Wisconsin Department of Natural Resources. Fish samples collected in 1974 showed extensive contamination.²¹ Affected types included Chinook salmon, Coho salmon, brown trout, tiger trout, lake trout, whitefish, and carp. Generally, the residue levels ranged well above 5 ppm, with lake trout (up to 43.8 ppm) and carp (up to 51.6 ppm) showing the highest concentrations.

In the Upper Fox River, samples taken at one station by Wisconsin researchers in 1975 showed high concentrations of PCBS in white suckers (32.7 ppm), carp (average 35.9 ppm), and northern pike (average 15.4 ppm). Samples from several areas of the Mississippi River analyzed by the department in 1973 showed similar residues of PCBS in walleyed pike, largemouth bass, northern redear, and carp. An interagency task force on PCBS, formed of federal and several state agencies of Minnesota and Wisconsin, sampled fish in the Mississippi River during the early summer of 1975 and again found similar levels of PCBS in several fish species.²²

Elsewhere, the Division of Fish and Game of the Commonwealth of Massachusetts has in the past detected high PCB levels in fish in several rivers and streams in the state.²³ From other data collected by the agency, it is clear that in 1972 there was extensive contamination of a large variety of fish species in nearly all the rivers which were sampled in the state, with mean

STRUCTURE OF PCBS

Polychlorinated biphenyls (PCBS) is a generic term used for a number of synthetic organic compounds that possess the following chemical structure:



PCBS are a mixture of different chlorinated biphenyl homologs (very closely related chemical substances). Thus, different PCB formulations or products may contain mixtures of biphenyls which have different percentages of the many possible chlorinated homologs, each containing a different number of chlorine atoms per biphenyl molecule. Monsanto Company, the sole U.S. producer of PCBs, sells its product under the trade name Aroclor. This product is sold under different formulations: Aroclor 1268, 1262, 1260, 1254, 1248, 1242, 1232, 1221, 1016, and so on. The last two digits in this biphenyl series denote percentage of chlorine in the mixture. Aroclor 1016 is an exception to this nomenclature, since it contains 41 percent chlorine and is, in fact, very similar in composition to Aroclor 1242. Trade names used by foreign manufacturers include: Phenoclor and Pyreclor (Prodelec, France), Kanechlor and Santatherm (Kansaguchi Chemical Company and Mitsubushi-Monsanto, respectively, Japan), Clophen (I.G. Farben-Industrie A.G., Germany), Fencior (Caffaro, Italy), and Sorol (U.S.S.R.).

PCB residue levels ranging between 1.11 and 197.0 ppm.

To date, however, the most contaminated fish in the U.S. have been found in the upper reaches of the Hudson River, near and below two General Electric plants in the Fort Edward-Hudson Falls area in the upper Hudson River more than 40 miles north of Albany. The two plants manufacture electrical capacitors in which PCBs are used as insulating fluids. In 1974, sampling by the U.S. Environmental Protection Agency (EPA) showed yellow perch and shiner minnows to contain average PCB residues of 17 and 78 ppm, respectively, with one rock bass sample containing 350 ppm.²⁴ It appeared that the two General Electric plants had been discharging approximately 30 pounds of PCBs into the river each day; in recent months, this amount seems to have declined to less than 10 pounds per day. Fish samples collected in 1975 by the New York State Department of Environmental Conservation confirm the earlier findings. The composite PCB averages in fish caught near one plant show typical values well above 20 ppm, with one American eel sample yielding residue

values of 403.4 ppm (concentration measured as Aroclor 1016/1242).²⁵

As large amounts of PCBs were being discovered in the upper Hudson River, New York State Department officials also were finding high residues of PCBs in fish species of recreational and commercial importance farther south in the Hudson River.¹⁵ Striped bass caught near the Albany area showed total PCB residue values of between 11.08 and 89.76 ppm. Fish caught in the lower reaches of the river had a relatively higher concentration of an older PCB formulation (Aroclor 1254), indicating long-range transport of PCBs discharged by industrial activities in the past. Striped bass sampled near the West Point and Tappan Zew area of the Hudson River (50 miles north of New York City and several hundred miles south of the Fort Edward-Hudson Falls area where the General Electric plants are located) had total PCB residues of from 1.16 to 7.54 ppm. Similarly, a sample of American shad in the Poughkeepsie area showed PCB residue levels of 9.0 ppm.

Disruption of the striped bass commercial fishing industry on the Hudson



High levels of PCBs have been detected in aquatic birds.

River would have serious economic consequences, since the value of the annual commercial catch appears to be about \$1.2 million.^{16,27,28} In addition, the annual take of striped bass by sportsmen exceeds the annual commercial catch.²⁸ Furthermore, the overall problem is serious in its relation to reproduction in striped bass. The striped bass is a migratory fish which has stock that intermingles in various locations such as the Chesapeake Bay, the Delaware River, and the Hudson River regions. As a result, PCB contamination may affect a large segment of the entire North Atlantic stock of striped bass.

Bottom Buildup

Of equal concern is the accumulation of PCBs in river-bottom sediments. PCBs, like most chlorinated hydrocarbons, are highly insoluble in water and tend to settle and then be adsorbed on silt and fine particles on the bottom.¹⁵

Data collected by the U.S. Geological Survey (USGS) on the Hudson River during the years 1973 and 1974 show the presence of PCBs in the water and in the bottom sediments of the river.²⁹ Values ranging between 0.3 and 3.0 parts per billion (ppb) PCBs have been reported in water samples collected at several stations on the Hudson River. The sampling sites include locations near Poughkeepsie, Waterford, Chelsea, and Rhinebeck.

In contrast to water samples, PCB concentrations analyzed from the river-

bottom sediments showed considerably higher values characteristic of the insoluble nature of PCBs. The USGS data are summarized below.²⁹

Date	Station	PCB in Sediments (ppb)
7/74	Chelsea	3,200
7/74	Poughkeepsie	11,000
7/74	Waterford	13,000
7/74	Winebrook Hills	0.1
9/74	Chelsea	1,800
9/74	Poughkeepsie	3,600
9/74	Waterford	640
8/75	Roger's Island, south of Glenn Falls	18,000

More recently, the EPA conducted water and sediment analyses for PCBs in the vicinity of Ford Island, New York, near one of the General Electric plants. Water samples collected directly south of the plant's waste-water discharge pipes had PCB values of 2.2 to 3.1 ppb. Nearby sediments, in contrast to the water, contained 340,000 to 2,980,000 ppb (540 to 2,980 ppm), with 6,600 ppb being detected several miles downstream of the discharge pipes.²⁴ These concentrations obviously are much larger than those at the more distant sites checked by USGS personnel, even though the latter concentrations are themselves excessive. One immediate practical problem is that the largest concentration of PCBs is likely to exist in the deep navigation channel of the Hudson, and dredging in this channel will further disperse the material.^{15,20}

Toxic Effects of PCBs

Based on the high lethality of PCBs to marine and freshwater aquatic species, the National Academy of Sciences-National Academy of Engineering Committee made the following recommendation:¹¹

"Aquatic life should be protected [that is, safe from toxic effects] where

the maximum concentration of total PCB in unfiltered water does not exceed 0.002 microgram per liter [0.002 ppb] at any time or place and the residues in the general body tissues of any aquatic organism do not exceed 0.5 microgram per gram [0.5 ppm]."

These levels obviously are exceeded in areas of the Hudson River and elsewhere.

The danger to aquatic life is a warning of the danger to human life. In fact, the toxicity of PCBs to humans has been clearly and tragically demonstrated. In 1968, a severe form of skin disease was diagnosed in Japanese families who consumed rice oil contaminated with Kanechlor 400 (KCB with 48 percent chlorine content).^{31,32} The disease, known as the "Yusho syndrome," is characterized by swelling of the upper eyelids, visual impairment, acne-like formations, and heightened pigmentation of the skin,³¹ and a total of 1,057 poisoning cases have been recorded to date in Japan.³³ Patients with this disease also have exhibited neurological disorders and have shown signs of hearing loss. Babies born to several patients had many of the symptoms of the disease, indicating a placental transport of PCBs; most fetuses of the victims were smaller than the national average. Another disturbing feature of the disease was the observation that most patients recovered very slowly, which suggested that PCBs were retained in their bodies over a very long period.³³ PCBs have been detected in human adipose tissue (connective tissue in the body containing stored cellular fat) in such widespread occurrence that over 40 percent of the U.S. population contains one part per million or more.³⁴

A number of studies have conclusively shown the effect of PCBs on the reproduction processes of some birds and animals, including monkeys.^{31,35,36,37,38,39,40} It is important to note that PCBs with lower chlorine content may have a greater effect on reproduction than biphenyls with higher chlorine content, even though the former substances are now being suggested as substitutes for the latter. In a recent review article on the toxicology of PCBs, Renate Kimbrough of the Center for Disease Control in Atlanta, Georgia, stated:³¹

"The results obtained from the reproduction studies in birds particularly seem to indicate that the lower chlorinated biphenyls affect reproduction more than the higher chlorinated biphenyls, and it is very important to determine whether the lower chlorinated biphenyls that have been suggested as replacements for presently employed chlorinated biphenyls have an effect on

PCBs, like most chlorinated hydrocarbons, are highly insoluble in water and tend to settle and then be adsorbed on silt and fine particles on the bottom.

reproduction."

The recent PCB toxicity tests in monkeys are particularly relevant to human health considerations. In an extensive series of tests carried out by James Allen and his colleagues at the University of Wisconsin School of Medicine, PCBs in the diet of rhesus monkeys caused significant effects that mimicked both the symptoms of known human toxicity and reproduction failures in experimental animals. When female rhesus monkeys were given 25 ppm of Aroclor 1248 for a period of two months, severe symptoms of facial swelling and skin eruptions were manifested.³⁸ At lower dietary levels (5.0 ppm and 2.5 ppm) of the same substance, similar clinical symptoms were observed after a few months; more significantly, these low levels of PCBs had a marked effect on reproduction. After three matings with male monkeys, only 12.5 percent of the 5 ppm PCB dietary group and 37.5 percent of the 2.5 ppm group were pregnant as compared to 90 percent pregnancies in a comparable control group.³⁹ At the same time, offspring were significantly smaller in size; their health was further threatened since they continued to ingest high PCB levels from the nursed milk of their mothers.⁴⁰ One notable consideration related to the experiments in monkeys is that the present FDA tolerance limit of 5 ppm⁴¹ for PCB residue in fish and shellfish is based on the assumption that toxic effects occur at levels much higher than is actually the case. On this basis, then, the FDA tolerance limit is too high.

Also, of serious concern is a recent study conducted by a group of research investigators at the Center for Disease Control in Atlanta, the EPA, the National Cancer Institute, and the Johns Hopkins University School of Medicine.⁴² They observed that when experimental female rats were fed a diet containing 100 ppm of PCB (Aroclor 1260) for about 21 months, 26 of 184 of the experimental animals examined had malignant liver tumors (hepatocellular carcinomas), whereas only 1 of 173 control animals had the disease. At the same time, 146 of the experimental and none of the control animals had tumorous lesions (neo-plastic nodules) in the liver.

Other effects of PCBs that have been reported in the scientific literature include: (1) increased production of certain enzymes in the livers of human beings and laboratory rats, (2) induction of liver porphyria in several animal species, (3) interference with the immune defense mechanisms in rabbits, (4) neurological disorders in rats, (5) increased PCB residue in fatty

tissues, serum, and milk, and (6) toxic effects in chick embryos.^{43,44}

There has been some concern that reported PCB effects may be attributed in part to the presence of chlorinated dibenzofurans as a contaminant in PCB mixtures.⁴⁵ Chlorinated dibenzofurans have been shown to be acutely embryotoxic and have been detected in several PCB preparations (Clorphen A 60, Phenoclor DP-6, Aroclors 1248, 1254, 1260).⁴⁶ Recent reports by M. Kuratsune and his associates have indicated that the "Yusho" patients apparently had been exposed to PCBs in rice oil contaminated with chlorinated dibenzofurans and showed detectable levels of this compound in their body tissue.⁴⁷ However, it was not determined from this study whether the contamination occurred during manufacture of the PCBs or during the use of the PCB mixture as a heat exchange fluid in the rice oil plant, since high temperatures may have led to its formation through the hydroxylation and oxygen ring bridging of chlorinated biphenyl molecules. At present, the possible link of PCB toxicity to the presence of chlorinated dibenzofuran contaminants remains highly speculative, as there appears to be no direct experimental evidence that dibenzofuran derivatives have long-term chronic effects similar to those which have been amply demonstrated with PCBs.^{48,49}

Control of PCBs

The attack on PCB contamination has taken two forms, one a voluntary response by manufacturers of the substance and the other a response by various government bodies. Both responses have been inadequate, as will be shown in what follows.

In 1971, over 106 million pounds of PCBs was produced in the industrial countries of the world, with a third of this, nearly 40 million pounds, being manufactured in the U.S.⁵⁰ During the late 1950s and into the 1960s, U.S. domestic sales of PCBs increased nearly threefold, reaching a peak of approximately 72 million pounds in 1970.⁵¹ Prior to 1970, about 60 percent of PCB material sold domestically in the U.S. was used for closed electrical systems such as capacitors and transformers. Another 25 percent was used as plasticizers and in the production of carbonless duplicating paper.⁵² In September 1970, Monsanto Company, the sole manufacturer of PCBs in the U.S., restricted its sale of PCBs because of environmental contamination by the chemicals. This restriction consisted of limiting sales to those materials earmarked for closed electrical systems. Thereafter, U.S. consumption was re-

duced considerably. It is important to note, however, that these reductions occurred mainly in the use of PCBs as plasticizers and as hydraulic and lubricating fluids, since the bulk of PCBs (60 percent), as mentioned above, had been used in electrical systems. This chemical substance is still being used almost exclusively as the insulating fluid for large, high-voltage capacitors (as well as for certain small ones) and for electrical transformers. In addition, the EPA estimates that approximately one-half million pounds of PCBs is being imported into this country each year⁵³ and presumably is being used in products other than electrical equipment.

As to government response to the PCB problem, a federal interdepartmental Task Force was convened in 1972. The group concluded that because of the highly persistent nature of PCBs and their bioaccumulation in the food chain, these chemical compounds posed a serious threat to human health. The group recommended restriction of the use of PCBs to closed electrical systems such as electrical transformers and capacitors.⁵⁴ However, since Monsanto Company actually had already begun restricting domestic sales in this fashion, it is not clear whether the task force's recommendations, issued a year and a half after Monsanto's action, had any influence on the company's decision (although this has recently been suggested by a federal official).⁵⁵

In July 1973, the FDA established PCB tolerance limitations in a variety of food products, as follows:⁵⁶

FDA Tolerances for PCBs in Food and Food Packaging (ppm)	
Milk*	2.5
Dairy products*	2.5
Poultry*	5.0
Fish and shellfish**	5.0
Eggs	0.5
Infant and junior food	0.2
Complete and finished animal feed	0.2
Animal feed components	2.0
Paper food-packaging material	10.0

*On fat basis

**Edible portion

Toxicity data obtained during the past year showing severe reproductive failure with low concentrations of PCBs in primates (5.0 and 2.5 ppm) and the recent demonstration of carcinogenicity with rats^{57,58} indicate the need to substantially revise the present FDA tolerance limitations of PCBs in foods. Although the present tolerance standards are deficient, the FDA has in recent months taken severe action based on these standards

against several fish shipments earmarked for commercial markets in Lake Michigan, since PCB concentrations in the fish exceeded the present 5 ppm guideline.⁵¹ Similarly, the FDA seized a shipment of carp this summer in Lake Pepin on the Mississippi River.⁵² In view of a recent legal decision that apparently allows the FDA to define pesticide residues in foods as an indirect food additive, the FDA is expected to enforce its guidelines more forcefully in the future. FDA officials in the Great Lakes region have warned that shipment of chubs, Coho salmon, and lake trout may be disallowed in interstate commerce, since the fish continue to contain high residue levels of PCBs.⁵¹ It is apparent that with a more thorough monitoring and surveillance program, many species of fish in the Great Lakes region and in several rivers in the East Coast area will be essentially unavailable for human consumption for many years.

One of the great ironies of the present situation is that more than three years ago the Federal Water Pollution Control Act (FWPCA) directed the federal government (specifically the EPA) to take swift action to reduce the discharge of PCBs and other toxic water pollutants to safe levels. The fact that today there is no meaningful national program to regulate PCB discharges is a result of the federal government's failure to carry out the act as written.

The congressional authors of the FWPCA believed that the "hazards posed by toxic substances" made their effective regulation "especially urgent."⁵³ Accordingly, they established in the act a "national policy that the discharge of toxic pollutants in toxic amounts be prohibited"⁵⁴ and added to the act a far-reaching special provision, Section 307(a), to ensure that this goal was fully implemented. If the section's mandatory deadlines had been met, toxic pollutant effluent standards covering PCBs and other toxic pollutants would have been promulgated by the EPA roughly two years ago and complied with by dischargers about one year ago. Yet, to date, the EPA has failed to promulgate a single standard under this provision.

Despite shortcomings in federal enforcement, several states have taken administrative and legislative initiatives to control the discharge of PCBs into lakes and rivers. Earlier this year, the Michigan Natural Resources Commission endorsed the recommendation of the Lake Michigan Toxic Substances Committee, calling for a ban on PCB imports and their use in the U.S. and for a rapid replacement of PCBs in electrical capacitors and transformers.⁵⁵ At the same time, a bill has

been introduced in the state legislature to place strict control on the use and sale of PCBs in Michigan.⁵¹

In Wisconsin, the Division of Health of the State Department of Natural Resources has issued warnings on consumption of large lake trout and salmon from Lake Michigan and the upper Mississippi River. In August 1975, the department held a hearing with a view to developing a strategy to control the sale and distribution of PCBs within the state.⁵¹ In Minnesota, an Interagency Task Force was established, principally to broaden the monitoring of PCBs in fish in several rivers and streams in the state.⁵²

In terms of administrative remedies, the strongest action was taken this year in New York state, after it was discovered that the two General Electric plants on the Hudson River ap-

peared to be the major dischargers of PCBs in upper regions of the river. On September 23, 1975, the New York State Department of Environmental Conservation issued an abatement order requiring the General Electric plants to achieve zero discharge of PCBs by September 30, 1976. Administrative hearings were begun on October 6, 1975.⁵⁵ At the same time, Ogden Reid, commissioner of the department, publicly warned consumers against eating striped bass from the Hudson River and salmon caught in Lake Ontario.⁵⁶

On the international front, the Organization for Economic Cooperation and Development (OECD) a few years ago recommended that its member countries which are involved in PCB production (including the U.S., West Germany, France, the United Kingdom, Japan, Italy, and Spain) restrict the commercial use of PCBs to closed systems.⁵⁷ Only Japan has taken fur-

Large amounts of PCBs have been found in fish in the Hudson River, especially in those of recreational and commercial value.



Photo by Wendy Holmes

MONS 087389

ther strict measures to completely eliminate the use of PCBs in all commercial applications, including capacitors and transformers. Japan also prohibits the import and export of the compound, and, since September 1972, there has been a virtual elimination of almost all applications of PCBs in that country.¹⁰

Most recently, following the National Conference of PCBs which was held in Chicago on November 19 to 21, 1975, the EPA on December 22, 1975, announced a far-reaching federal policy of "totally eliminating the use of polychlorinated biphenyls (PCBs) as readily as possible" in the U.S.^{11,12} In a more specific action, the EPA has ordered the two General Electric plants on the Hudson River to achieve zero discharge by June 1977.¹³ It is clear from these actions that the federal government has recognized the gravity of the PCB issue and at last has taken definitive steps to address the problem.

The title of this article as originally submitted was "A Review of Federal and State Government Roles in Controlling Impacts of PCBs on the Environment." The Publisher and Editors of Environment are responsible for the published title and textual selection of photographs and lead-in excerpt, photo captions, and preparation of most graphs and illustrations which appear in Environment articles.

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BY JOSEPH HIGHLAND

THE FIRST EVIDENCE that food supplies had become contaminated with polychlorinated biphenyls (PCBs) was reported in 1966, when fish from various Swedish waters were shown to contain these chemicals.¹ Since then, PCBs have been identified as contaminants in food supplies around the world. They have been found in pike taken from a lake in Finland, in mollusks and marine fish from Scottish waters, in a wide variety of fish caught in the St. John River system, in wildlife in California, and in trout and salmon taken from Lake Michigan.^{2,3} More recent findings indicate that contamination by PCBs continues to be widespread. Within the last few months, striped bass caught in the Hudson River have been found to contain as much as 90 parts per million (ppm) PCBs, while pike caught in the Baltic Sea were shown to contain 0.31 ppm PCBs in their muscle and up to 190 ppm in their ovaries.^{4,5} Moreover, plankton from the Baltic Sea, upon which many fish feed, contained levels of 25 ppm PCBs in their fat.⁶

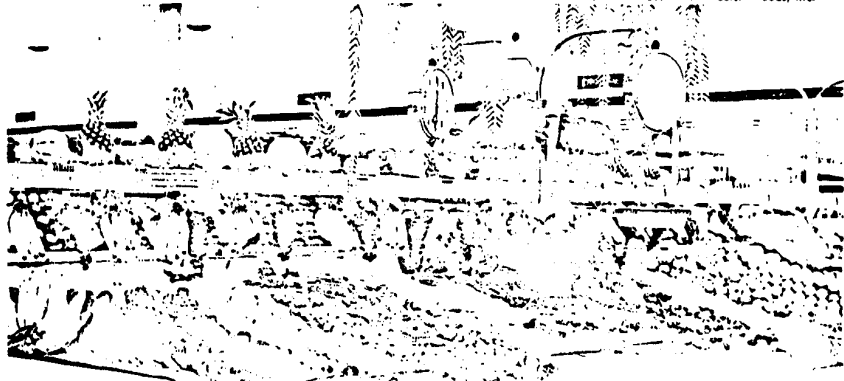
The federal government currently conducts three separate programs in which the contamination of foods by PCBs is measured: the FDA (Food and Drug Administration) Pesticide Surveillance Program, the FDA Total Diet Program, and the USDA (U.S. Department of Agriculture) Surveillance Program.

The FDA Pesticide Surveillance Program, which began in 1968, was developed to gather information on the levels of pesticides, as well as of PCBs, in foods and animal feeds based on a statistical sampling program designed to cover specific geographical regions of the U.S. The program is intended to assure the safety of the human food supply by screening and removing shipments of foods containing PCBs or pesticides in excess of established tolerances. Commodities covered under this testing program include dairy products, eggs, fish, animal feeds, fruits, vegetables, and processed foods.

In the FDA Total Diet Program, the levels of pesticides, PCBs, and trace heavy metals in sample diets are measured. Twice a month, a diet approximating that which would be consumed in a two-week period by a hypothetical fifteen- to twenty-year-old male is collected at retail stores in five regions around the U.S. The foods are prepared in the appropriate manner and then divided into twelve basic food composite classes, including meat, fish, poultry, and leafy vegetables. Each com-

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Photo from Bettendorf's Select Foods, Inc.



PCBs IN FOOD

What IS acceptable?

posite class is then analyzed for contaminants.

The USDA Surveillance Program consists of periodic sampling and analysis of meats and poultry for chemical contaminants, including PCBs. Samples are taken from federally inspected slaughterhouses.

Levels of PCBs in Food

A summary of the findings regarding PCBs in the three survey and surveillance programs are listed in Table 1.^{1,2,3} Although the data are incomplete, it is evident from this table that PCBs continue to contaminate our food. Results of the FDA Pesticide Surveillance Program appear to indicate an increase in the extent of contamination, while those of the USDA Surveillance Program indicate a decrease; this apparent conflict may be explained by the fact that different foods are sampled in these programs, and it is quite possible that contamination of some kinds of foods has decreased while it has increased in others.

The apparent decrease in the levels of PCBs reported in the FDA Total Diet Program Survey between fiscal year 1973 and the first half of fiscal year 1974 must be viewed with caution. During the first half of fiscal year 1974, the average PCB levels for milk and other dairy products, for eggs and egg products, and for animal feed ingredients were equal to or greater than those reported for fiscal year 1973.

Tests are being conducted by the FDA Pesticide Surveillance Program on food items such as fruits and vegetables to determine the amounts of PCBs in areas of suspected acceptable levels.

TABLE 1

COMPILATION OF DATA FROM FEDERAL SOURCES

FDA Pesticide Surveillance Program

Year	Number of Samples	Percent Contaminated	Levels of Contamination
1968-71	15,000	3.2	"detectable" (fish, specifically, 1-10 ppm)
Fiscal 1973	7,882	4.0	"detectable"
Fiscal 1974	1,723	5.4	"detectable"

FDA Total Diet Program Survey

Year	Number of Composite Food Samples	Percent Contaminated	Levels of Contamination
1968-71	900	6	trace — 0.36 ppm
Fiscal 1973	360	6	trace — 6.0 ppm
Fiscal 1974	360	4	trace — 0.05 ppm

USDA Surveillance Program

Year	Number of Samples	Percent Contaminated	Levels of Contamination
1971	4,175	4.0	trace — 15 ppm
Fiscal 1973 (Poultry only)	1,037	2.6	trace — 4 ppm
Fiscal 1974 (Poultry only)	574	1.0	1.25 ppm

Then why is there an apparent decrease between 1973 and 1974? In large part, this reflects an apparent decrease in the number of fish that were found to be contaminated with PCBs. In fiscal year 1973, 62.2 percent of the fish samples examined contained PCBs ranging from trace amounts to 123 ppm. In the first half of fiscal year 1974, only 35.3 percent of the samples checked were contaminated, and the levels found ranged from trace

amounts to 9.7 ppm. However, the fish data reported for fiscal year 1973 included mostly freshwater fish, in which PCB contamination has been shown to be greatest, while during the first half of fiscal year 1974 the samples consisted mainly of marine fish. Since the samples are not comparable, the apparent decrease is probably a reflection of the difference in levels of PCB contamination between freshwater and marine fish rather than an indication

of a general decline in PCBs in fish.

In addition to these observations, numerous specific incidents of PCB contamination of food have been reported by the FDA and the USDA. In 1970, the Campbell Soup Company detected high levels of PCBs (26.8 ppm on a fat basis) in chickens grown in New York state. As a result, 140,450 chickens were destroyed. In July 1971, the Monsanto Company informed the FDA that large amounts of fish meal might have been contaminated with PCBs during the pasteurization process, due to a leak in the heating system at the company's East Coast Terminal in Wilmington, North Carolina. An investigation by the FDA revealed that the leak had begun in April 1971 and had continued through July 1971. Contamination of the fish meal was verified, and, as a result, over 123,000 pounds of egg products and 88,000 chickens were destroyed. That same year, the FDA was notified that high levels of PCBs (20 ppm) had been found in Swift and Company turkeys. As a result, approximately 1,000,000 birds were kept from market. These incidents and the results of the federal surveillance programs demonstrate further that contamination of food by PCBs is not a new phenomenon; it is a continuing problem which has resulted in part from the continued indiscriminate discharge of PCBs into the environment, and it is one which must be eliminated because of the severe health hazards posed by these compounds.

"Acceptable Dietary Intake"

The FDA has calculated what is termed an "acceptable daily dietary intake" for PCBs in the human diet — or a level of PCBs which, if ingested for an extended period of time, would be toxicologically insignificant. Even if the validity of such a concept is assumed, the basis for establishing such a level must be evaluated. In this connection, the FDA's final environmental impact statement for rulemaking on PCBs indicated that both animal and human toxicological data were used to establish an allowable dietary intake for human beings.

According to data then available (1973), long-term studies of dogs and rats showed "no-effect levels" (the dose levels below which the effects looked for were not observed) of 10 ppm for three commercial types of PCBs (Aroclors 1242, 1254, and 1260) fed in the diet. Employing the standard 100-to-1 safety factor used in applying results of animal experiments to human health standards, a no-effect level for human beings, based on data derived from the dog experiments, was

It appears that the metabolism and excretion of PCBs, especially of Aroclors with high chlorine content, are even slower than for some of the most persistent pesticides, such as dieldrin and DDT.

calculated to be an intake of 0.0025 milligram per kilogram of body weight per day. From the data on rats, a figure of 0.003 milligram per kilogram of body weight was obtained. On the basis of the tests in dogs, the allowable level of PCB intake in a man weighing 70 kilograms (about 155 pounds) was figured to be 0.175 milligram per day (70 kilograms x 0.0025 milligram).

The human toxicological data used in determining the allowable daily dose were derived from investigations of human PCB poisonings in Japan. In 1968, over 1,000 Japanese persons experienced PCB poisoning after consuming rice oil contaminated with Kanechlor 400.^{9,10} In addition to causing headaches, swelling of eyelids, temporary loss of vision, and many other symptoms, PCBs stored in the adipose tissue (connective tissue containing stored cellular fat) of pregnant women were passed through the placental wall to the fetuses. As a result, nine of the ten live-born babies had unusually greyish, darkened skin, and most were born underweight.

The outbreak was interpreted by the FDA to have the following implications for PCB intake standards:¹

"Since 2,000 milligrams was reported to be the average total dose causing an effect in the Japanese, it is possible that 200 milligrams total dose PCBs (applying a safety factor of 10 to 1) may be tolerated over a much more protracted period of time without overt adverse effect if daily exposure is held to minimal levels. This would permit ingestion of 4 micrograms (0.004 milligram) per kilogram body weight per day in a 70-kilogram man. Since the lowest total dose producing an effect in man in the Japanese incident was 500 milligrams, a similar analysis leads to an allowable protracted ingestion of 1 microgram (0.001 milligram) per kilogram body

weight per day as derived from a 70-kilogram man."

On the basis of these data, the FDA arrived at an allowable dose of PCBs (for a 70-kilogram man) of 70 to 280 micrograms per day — or 0.07 to 0.28 milligram per day — compared to the 0.175 milligram-per-day permissible level established on the basis of the dog experiments. The validity of these calculations is very questionable, however. Particularly suspect are the estimates that had to be made of doses received by individual Japanese, as well as the arbitrary assumption that a safety factor of 10 to 1 is sufficient to determine a safe level of intake for a "protracted period of time." In addition, the calculations fail to consider the potential long-term effects caused by stored PCB residues.

Furthermore, according to H. Blumenthal (acting director, Division of Toxicology, Bureau of Foods, FDA) in the time since these calculations were made, there have been two important experimental observations concerning PCBs which make the previously calculated allowable daily intake level obsolete.² First, recent studies by J.R. Allen and his colleagues on the effects of low-level exposure of nonhuman primates to PCBs must be considered.^{11,12,13,14} The effects seen in these studies of monkeys are similar in appearance and persistence to the symptoms of the human poisoning in Japan. Moreover, these symptoms were documented at dietary intake levels as low as 2.5 ppm (equivalent to about 2.5 milligrams per kilogram of body weight). To date, a no-effect level for PCBs in monkeys has not been established. This would indicate that dogs and rats are not particularly reliable subjects for tests relating to primates, since they are not as sensitive as are primates to many of the toxic effects of PCBs.

Second, Allen's work indicates that the use of gross observations of toxicity are not reliable in the study of PCBs.¹ Allen observed that routine toxicological evaluations of rats which were fed 100 ppm Aroclor 1248 for one year revealed no abnormalities in growth, mortality, or hematology. However, more sophisticated tests showed that these rats experienced drastically altered fat and cholesterol metabolism as well as changes in liver size and structure. Therefore, even if an allowable human daily dosage for PCBs calculated from animal toxicological data were acceptable, the basis for the FDA's 1972 calculations would be seriously questionable.

The chemical nature of PCBs may help to explain in part why they are subacutely and chronically toxic. The

chemicals have a low solubility in water but are highly soluble in fat.¹³ Therefore, they tend to accumulate and concentrate in the body's fat stores rather than being rapidly metabolized and excreted. It appears that the metabolism and excretion of PCBs, especially of Aroclors with high chlorine content, are even slower than for some of the most persistent pesticides, such as dieldrin and DDT.¹⁴ Thus, after ingestion, PCBs persist in the body for long periods of time and may thereby elicit chronic effects.

Excessive Intake Is Common

The FDA's acceptable daily PCB intake level of 75 to 210 micrograms per day for a 70-kilogram man seems debatable at best. As J. Wessel, scientific coordinator of the FDA, has pointed out,¹ "It is quite clear that the acceptable daily PCB intake of 75-210 micrograms per day could easily be exceeded by an adult consuming some of the heavily contaminated foods in his diet." Table 2, taken directly from Wessel's testimony,¹ illustrates this point clearly: Consuming a steady diet of combined foods from the list would produce consistent over-exposure to PCBs if each food had the maximum allowed concentration of PCBs (listed under the heading "Temporary Tolerance [ppm]").

As Wessel concludes, "Even when PCBs are present at the [permissible] tolerance level, it would be possible for an adult, with normal dietary habits, to exceed 175 micrograms [0.175 milligram] of PCBs per day." Furthermore, adults with "normal dietary habits" are not the only people who are vulnerable. Perhaps at even greater risk are those large groups within the population (such as sport fishermen or people who require special diets) who consume above-average quantities of PCB-contaminated foods, or nursing babies who ingest PCBs in their mothers' contaminated milk and who, for natural biochemical reasons, have a very limited capacity for metabolizing foreign substances such as PCBs. A. Kolbye, Jr., associate director for science, Bureau of Foods, FDA, and H. Blumenthal are clearly correct when they say, respectively:^{17,2}

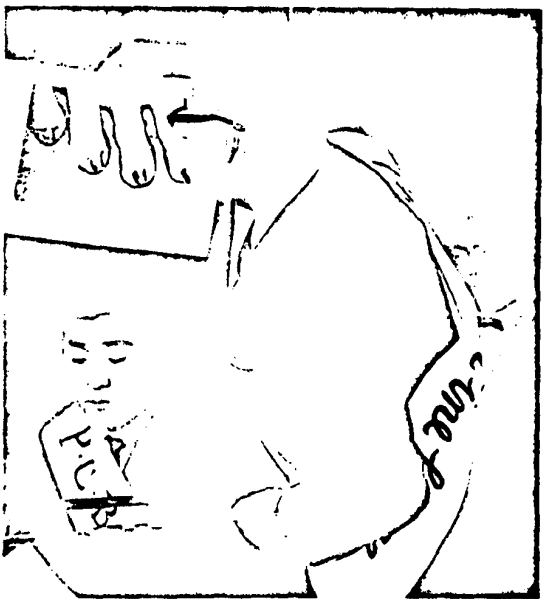
"Infants or young children could also readily exceed the tolerable daily exposure," and, "Thus, the best evidence indicates that children would be more susceptible to the poisoning caused by unmetabolized PCBs, since

TABLE 2

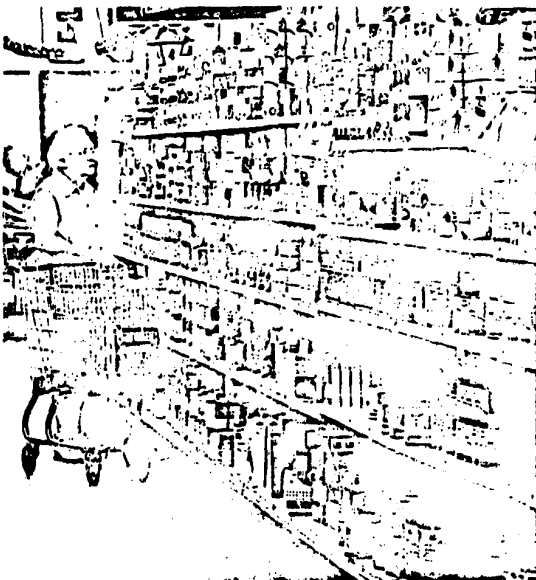
RELATIVE CONTRIBUTION TO ACCEPTABLE DAILY INTAKE (175 mcg/s) BY FOODS SUBJECT TO PCB TEMPORARY TOLERANCES

Type of Food	Serving of Food	Temporary Tolerance (ppm)	Equivalent Amount PCBs in Food	Relative Contribution to ADI
Milk	800 ml (3.5 cups)	2.5 (fat basis) 0.1 (whole product)	80 mcg	46%
Cheese	100 g (3.5 oz)	2.5 (fat basis) 0.6 (whole basis, assuming 25% fat)	60 mcg	35%
Poultry	200 g (7 oz)	5.0 (fat basis) 0.5 (whole basis, assuming 10% fat)	100 mcg	58%
Eggs	100 g (2 eggs)	0.5	50 mcg	29%
Fish	200 g (7 oz)	5.0	1,000 mcg	580%
Packaged Food	100-200 g (3.5-7.0 oz)	10.0 (Packaging)	10-60 mcg*	6-35%

*Based on 1971 FDA Survey that 10 ppm PCBs in packaging can result in migration of 0.1 - 0.6 ppm PCBs to packaged food.



Japanese doctor explaining the effects of PCB poisoning of rice to victims in 1968.



PCBs can seep from food packaging materials into food itself.

of photographs and lead-in excerpts, photo captions, and preparation of most graphs and illustrations which appear in *Environment* articles.

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they have not developed effective ways of eliminating them from the body."

Need for Reevaluation

The FDA's temporary tolerances for PCBs in food are both inadequate and inappropriate; the acceptable daily dose calculated in 1972 is now obsolete in light of more recent findings. More importantly, however, the appropriateness of any "acceptable daily dose" for PCBs is highly questionable in light of new evidence demonstrating the carcinogenicity of these compounds in experimental animals.¹⁵ Since there is at present no scientific evidence that any level of intake of a

carcinogen is safe, the entire concept of an acceptable daily dose for carcinogenic substances such as PCBs is contradictory and scientifically unsupportable.

Our growing concern about the continued contamination of food supplies by PCBs reflects an ever-increasing awareness of the extreme health hazards posed by these compounds. Federal sampling and surveillance programs have continually demonstrated the presence of PCBs in our food supply, and the existing temporary tolerance levels for PCBs established by the FDA have been insufficient to protect the public health. A new regulatory posture is needed which takes into account recent evidence of chronic effects caused by low-level dietary intakes of PCBs and, even more importantly, the new evidence substantiating the carcinogenicity of these compounds in laboratory animals.¹⁶ □

The USDA Surveillance Program consists of periodic sampling and analysis of meats and poultry for chemical contaminants, including PCBs.

The title of this article as originally submitted was "PCBs in Foods: A Look at Federal Government Responsibilities." The Publisher and Editors of *Environment* are responsible for the published titles and subtitles, selection

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