December 10, 1969

PCB -- Industrial Brother of DDT

DEC 17 1969

by [redacted]

In the alphabet of environmental contamination, PCB may become as well known as DDT, and for some of the same reasons. Like DDT, PCB contains chlorine, hydrogen and carbon; like DDT, it is not soluble in water, it is soluble in fat, and it is extremely persistent in the environment.

The chemical similarity between the two also suggests similar physiological effects in the bodies of fish, birds, and perhaps people. In one particular respect, which will be discussed later, this has been confirmed in the laboratory. ?

A striking difference is that DDT has been used in the open environment with intent, because of its toxic effects, to destroy insects; PCB, although known to be toxic, has been used for its other qualities, and has escaped into the environment by accident. It is not yet known exactly how and where this happens, and how much is escaping. The fate of PCBs is another example of the fact that every manufactured product is eventually thrown away, and in one way or another reaches the environment.

Polychlorinated biphenyls, to give PCBs their proper name, are chemical compounds manufactured in this country by the Monsanto Chemical Company and in Europe and Japan by other manufacturers. Aroclor, the Monsanto trade name, came on the market in the thirties and began to be used extensively during World War II.

PCBs have a wide variety of uses. Production and sales figures

are not available, but a Monsanto bulletin (1) lists many products which may contain PCBs as plasticizers, flame retardants, insulating fluids, or to impart some other useful quality. Among these products are natural and synthetic rubber, electrical products, floor tile, printers ink, coatings for paper and fabric, brake linings, automobile-body sealants, paints, varnishes, waxes, asphalt, and many kinds of adhesives and resins. PCBs are also recommended by Monsanto for mixing with chlorinated insecticides to suppress their vaporization and extend their "kill-life."

The amounts being used are suggested by the fact that PCBs are available in containers ranging from 50-pound cans to 600-pound steel drums and by the tank carload.

As with many of the chemical compounds so widely used in this age of synthetics, concern about the hazards of PCBs has been limited to the possible exposure of industrial workers. Until 1966 it did not occur to anyone to ask whether PCBs might be released into the environment or if they were, what they might do to birds or fish or human beings.

The first revelation that these compounds had been dispersed in ways which made them available to living things came in 1966 with a report by Soren Jensen, a Swedish scientist, that he had identified PCBs in the bodies of 200 pike taken from different parts of Sweden and in other fish as well as an eagle. (2) Dr. Jensen had also analyzed eagle feathers which had been preserved in a museum, some of them

going back as far as 1880. He found PCBs first in feathers dated 1944.

A colleague of Dr. Jensen, Gunnar Widmark, reported later that year to an international commission studying pesticide analysis. One of the problems in isolating and identifying residues of DDT had been the presence of what Dr. Widmark described as "unknown but chlorine-containing compounds" so similar to DDT that they may sometimes have been mistaken for it. These compounds he and Dr. Jensen had identified as PCBs. The polychlorinated biphenyls had been found in fish and sea birds, in conifer needles, and in some samples of human fat. (3)

Scientists in Britain (4) and the Netherlands (5) have now identified PCBs in wildlife in their respective countries. Jensen and his colleagues have recently reported details of their analysis of Swedish wildlife. (6)

The detection of PCBs in North America began in the spring of 1967 when Monte Kirven of the San Diego Natural History Museum was searching the southwest for peregrine falcons, those swift-flying and now fast-disappearing birds of prey. He found a nest with a single, unhatched, abandoned egg and brought it to our laboratory at the Institute of Marine Resources, University of California in Berkeley. The egg was found to contain five milligrams of DDE, the DDT compound that is apparently the most abundant synthetic pollutant in the environment. The analysis also showed the presence of other compounds which could not be identified. These same compounds had been evident in the analysis of other species of birds and of fish.

Because of the importance of the peregrine, an attempt was now made to identify them, but without success until the Swedish papers came to our attention. With this clue, further laboratory work made it clear that the hitherto unknown compounds were polychlorinated biphenyls. (7)

The Monsanto Company, according to its statement of March 3, 1969 (see page ___) is not yet ready to accept the conclusion that formerly unknown chlorinated compounds are indeed PCBs. However, because similar results were obtained independently in the laboratories of Sweden, England, Scotland, the Netherlands and the United States, supported by detailed descriptions of the analytical techniques used, including mass spectrometry and the results obtained, this conclusion is now generally accepted in the scientific community. The amounts found are frequently not in the parts per billion range, as the Monsanto statement suggests, but are up to a million times higher. Amounts as large as 1,980 PCB per million parts of body fat in North American peregrine falcons and 17,000 parts per million in the fat of the white tailed eagle in Sweden have been reported. (8)

The Monsanto statement suggests that the chemical compounds identified as PCBs may be something else, synthesized by marine organisms. The only species known to synthesize PCBs is man. The distribution of these compounds is highest in the areas where men are concentrated and carry on industrial activity. They are found in lower amounts in the sea.

The Monsanto statement refers to the chemical stability of PCBs and their insolubility in water. These are among the qualities which make them desirable for various industrial uses. They are also among the qualities which make them a great concern as environmental pollutants. Because they have these characteristics, they are extremely persistent--just how persistent is not known--and they can be carried, like DDT, far and wide over the globe by air currents. (DDT is one of the most stable and least water-soluble of the chlorinated hydrocarbon pesticides.)

The one insecticide use mentioned in the Monsanto bulletin would not be adequate as a source to explain the PCBs that have already been found in the environment. There are four other possible sources, all of which should be explored. The first is the stacks of the plants which manufacture Aroclor and the plants which manufacture the products incorporating Aroclor. The second is other forms of industrial waste. For example, PCBs, other synthetics and oil are mixed to make a fire-resistant hydraulic fluid, one of the "closed system" uses of PCBs referred to in the Monsanto statement. Yet the amount of hydraulic fluid that escapes from this "closed system" is suggested in a Mobil advertisement stating that "industry wastes almost four times as much hydraulic oil as it uses. The third possible source is gradual wear and weathering of Aroclor-containing products which may result in slow release of PCBs as vapor or minute particulate matter into the atmosphere: For example, asphalt and tires are both subjected to constant

Sen. Fred Harris's statement

friction, and may be sources of either vapor or particulate matter containing PCBs. Fourth, many products containing PCBs eventually wind up on the city dump, where they may be burned. Although fire resistant, at sufficiently high temperatures PCBs may be released as a vapor in the form of highly toxic fumes. (9) PCBs have not yet been detected in samples of airborne particulates, but have been shown to be present in rainwater. (10) More study is obviously needed to show the ways in which PCBs enter the environment.

The worldwide distribution of DDT and related pesticides by a variety of mechanisms, but primarily by air currents, is now known (See "Earth, Air, Water," Environment, July-August, 1969). Even without knowing the exact sources of PCBs, it would be possible to discover, by the analysis of various forms of wildlife, whether a similar distribution of PCBs is taking place, or whether PCB contamination is local. In the past two years our own work in California, and that of scientists elsewhere in this country, Britain and Sweden has provided mounting evidence for the worldwide dispersion of PCBs. The levels of PCBs found in wildlife near industrial centers may be very high, frequently higher than the amount of DDT. Birds and fish from more remote areas may contain less PCB than DDT.

In the table below, the amounts of DDT and PCB found in various tissues in two peregrine falcons from California and two from the Arctic are shown. The final column shows the ratio of DDT to PCB--in the California birds, between one and two times as much DDT; in the

arctic birds, from three and a half to six times as much DDT as PCB.

Table 1 - DDT and PCB in Peregrine Falcons

Concentrations in parts per million, wet weight, dry weight or lipid (fat) weight. (Note: Total DDT includes p,p' DDT, p,p' DDE, p,p' DDD, p,p' DDMU, o,p' DDT and o,p' DDE)

	Total DDT	PCB	DDT/PCB
Immature bird, California			
breast muscle (wet)	14.4	9.4	1.5
liver (wet)	7.7	4.5	1.7
brain (wet)	2.8	1.5	1.9
brain (lipid)	36	19.3	1.8
carcass (wet)	20.2	10.8	1.9
carcass (lipid)	300	160	1.9
Adult bird, California			
breast muscle (wet)	127	98	1.3
liver (wet)	77	57	1.4
brain (wet)	49.5	34.6	1.4
brain (lipid)	595	415	1.4
carcass (wet)	85	65	1.3
carcass (lipid)	2,600	1,980	1.3
Immature bird, Arctic			
breast muscle (wet)	2.3	0.16	14
breast muscle (dry)	7.8	0.54	14
liver (wet)	1.0	0.10	10
brain (wet)	0.43	0.032	12
body fat (wet)	50.3	3.2	16
carcass (wet)	9.3	0.80	12
carcass (lipid)	63.7	5.5	12
Second year bird, Arctic			
breast muscle (wet)	99	28	3.5
breast muscle (dry)	296	84	3.5
brain (wet)	85	21	4.7
carcass (wet)	70	19.7	3.5
carcass (lipid)	5,000	1,420	3.5

The contrast between the immature and the older birds from both areas is also interesting. The immature California bird had 160 ppm in its carcass fat, while the adult California bird had 1,980 ppm. The contrast

between the immature and older migrant birds from the Arctic was even greater, with an increase from 5.5 ppm in the young bird to 1,320 in the older one. The birds migrate through the U.S. and winter in the tropics. Evidently they pick up both DDT and PCB during their migration and on their wintering grounds.

Analyses of Western Gull eggs show that eggs from San Francisco Bay contained more PCB than eggs from the Farallon Islands, 27 miles west of the Golden Gate Bridge, which in turn contained more PCB than eggs from Baja, California.

Samples of bird tissue and eggs from a variety of species were collected off California. (See Table 2) Some of them breed in New Zealand and Australia, others on the coasts of British Columbia and Alaska, still others in Alaska and the Canadian Arctic. It is not clear how much DDT and PCB the birds ingested in their native breeding grounds, and how much in the vicinity of California. What is quite clear is that PCB is widely distributed among marine birds which are at the top of a complex mesh of food chains in the sea. One source of PCBs in the birds is clearly the fish on which they prey, as PCBs have been found in a variety of fish from many different waters. (11)

PCBs have also been found in freshwater fish and terrestrial birds, although there is less published data than for marine organisms.

What species?

Table 2 - PCB Content of Eggs of Sea Birds

(PCB in micrograms. Where sample included more than one egg, value given is average)

Species	Locality	No. Samples	PCB
Brandt's Cormorant	Farallon Islands	17	113
Pelagic Cormorant	San Mateo County, Calif.	2	128
Murre	Farallon Islands	6	558
Pigeon Guillemot	Farallon Islands	1	20
	San Mateo County	1	62
Cassin's Auklet	Farallon Islands	2	15.
Western Gull	Farallon Islands	1	118
	San Mateo County	1	112
	San Francisco County	1	480
Black-crowned night heron	San Francisco Bay	1	330
	San Francisco Bay	1	24
Caspian Tern	San Francisco Bay	2	805
	San Diego Bay	5	1,010
Forsters Tern	San Diego Bay	2	114
Least Petrel	Baja, California	2	3.1
Peregrine Falcon	Baja, California	1	471

The American Conference of Governmental Industrial Hygienists (ACGIH) recommends levels of hazardous materials which should not be exceeded in occupational situations. The ACGIH recommendation for PCB containing 42 percent chlorine is the same as for DDT, namely, a maximum or "threshold limit value" of one milligram per cubic meter of air. But some PCBs are more heavily chlorinated and for those with 54 percent chlorine, the ACGIH threshold limit value is half as much (0.5 milligrams per cubic meter of air). (12) Among the Aroclors listed for sale by Monsanto are chlorinated biphenyls that are up to 68 percent chlorine and mixtures of chlorinated biphenyls and chlorinated triphenyls that are up to 65 percent chlorine.

The more heavily chlorinated PCBs have been reported in wildlife both in the U.S. and in Europe, and have been found to be highly persistent in the bodies of fish and birds, while the more lightly chlorinated PCBs are somewhat less persistent, and are metabolized or excreted more rapidly. (13) This was confirmed by a laboratory experiment by Koeman in which Japanese quails were fed a mixture of PCBs. After the birds died, it was found that some of the PCBs, particularly the more lightly chlorinated ones, had been metabolized. However, this is not necessarily reassuring. Little is known about the metabolites--the breakdown products--of PCB; one of the metabolites of DDT (DDE) affects the reproductive systems of birds.(14)

PCBs are described in"Dangerous Properties of Industrial Materials" as affecting human skin and liver at high concentrations, although how high is not stated. The toxicology is described as follows:

> The lesion produced in the liver is an acute yellow atrophy. This hepatotoxic action of the chlorinated diphenyls [same as biphenyls] appears to be increased if there is exposure to carbon tetrachloride at the same time. The higher the chlorine content of the diphenyl compound, the more toxic it is likely to be. Oxides of chlorinated diphenyls are more toxic than the unoxidized materials.
>
> The skin lesion is known as chloracne, and consists of small pimples and dark pigmentation of the exposed areas, initially. Later comedones [blackheads] and pustules develop. In persons who have suffered systemic intoxication the usual signs and symptoms are nausea, vomiting, loss of weight, jaundice, edema and abdominal pain. Where the liver damage has been severe, the patient may pass into a coma and die.

Cases of occupational chloracne and of systemic damage were reported in the early years of PCB use. Experiments with laboratory animals also revealed the high toxicity of PCBs injected, fed or applied to the skin. (15)

There is no evidence to indicate that PCBs in the environment are likely to reach levels which would cause severe injury or death from short-term exposure. The PCB hazard, like so many environmental hazards, is one of long-term, low-level exposure and of possible synergistic effects with other poisons. There is also the more indirect, but no less real danger of destroying

other forms of life, part of the vast interconnected web of species of which man is but one part, and on which he depends.

The accumulating evidence that no part of the world is now free of pesticide residues, the products of atomic explosions, or of a variety of industrial pollutants has induced subtle but profound changes in our concept of our position in the global ecosystem and in our individual conceptions of remoteness and isolation. There has never been any question that local ecosystems could be irreversibly changed by the introduction of synthetic chemicals, whether these introductions be purposeful or incidental. Since the sea, however, is the dominant feature of the worldwide ecosystem, interpretations of environmental contamination in terms of local ecosystems, whether terrestrial or aquatic, may be misleading. The accumulation of significant amounts of several pollutants, including PCBs, in marine organisms, pollutants which are insoluble in water, but soluble in fat, not only elicits an uncertainty about the long-term utilization of the sea as a source of human food, but has suddenly raised the question of the ultimate survival of a number of species of sea birds. These species comprise a very large fraction of the world's wildlife and doubts about their future would have been considered preposterous and untenable only three or four years ago. Massive reproductive

failures have been noted in the brown pelican and the double-crested cormorant in Southern California. The jack mackerel fishery out of Los Angeles has just been closed because of the high levels of DDT in the fish.

In terms of the exposure of many forms of life and of people of all ages and states of health to PCBs, is there really a "threshold limit value"? Is there really an amount of polychlorinated biphenyl which is safe to breathe or to ingest? (A "threshold" is an amount below which no effect is noted.) No threshold has been found for the effect of DDE on eggshell thickness.

The close similarity of DDT and PCB suggested the possibility that PCB might also have biological effects. Accordingly, David Peakall investigated the effect of PCB on pigeons, and found that, like DDT, it induced enzymes in the liver, enzymes which broke down estradiol, a sex hormone.

The entire reproductive cycle in birds, from the development of the male and female reproductive organs to the laying of the egg and the birth of the chick is mediated by various sex hormones. When the work of a hormone has been done, it is broken down by enzymes in the liver. However, if the enzyme activity is artificially increased by the introduction into the body of a chlorinated hydrocarbon, it appears that the hormone will be broken down at a much faster rate.

Some aspect of the reproductive process may not be completed normally. Among the effects of chlorinated hydrocarbon pesticides that have been found in birds are delayed laying of eggs, thin egg shells (sometimes so thin as to break) and chicks that die at hatching.

In Peakall's experiment, four groups of four birds each were used. The first group was given no chlorinated hydrocarbon. The second was injected with DDE (the persistent breakdown product of DDT); 40 milligrams of DDE per kilogram of bodyweight were injected. The third group was injected with 40 milligrams of DDT per kilogram of bodyweight, and the fourth with 20 milligrams of PCB per kilogram of bodyweight. The PCB used was Aroclor 1262, which is approximately 62 percent chlorine. One week later, the birds were killed, and the portion of the liver containing enzymes was removed. To each liver was added a tiny amount of the sex hormone, estradiol. The breakdown of the estradiol was increased two-and-a-half times by DDE, three-and-a-half times by DDT and almost six times by PCB, in spite of the fact that only half as much PCB was used. (On a weight basis, the PCB had an estradiol-degrading potential four times that of DDT and 5.6 times that of DDE). (Note: Results were in average amounts of polar metabolites formed in millimicromoles for each group of four birds:

Control	29.3 ± 6.5
DDE	76.2 ± 13.1
DDT	93.1 ± 11.2
PCB	160.0 ± 10.5

MONS 031604

J.C. Street and his co-workers at Utah State University investigated the effect of a series of ten PCBs ranging from 21 to 68 percent chlorine on female rats. All of them stimulated enzyme induction; the amount of stimulation increasing regularly with the percentage of chlorine in the PCB. PCBs containing more than 50 percent chlorine were as potent in enzyme induction as DDT.(16)

Species of birds like the peregrine falcon, threatened with extinction, may prove to be the victims of a combination of poisons, rather than of DDT alone. All the white-tailed eagles (another threatened species) studied by Jensen in Sweden contained PCBs, mercury compounds and breakdown products of DDT. Steps now being taken to reduce the amount of DDT used may accomplish less than their proponents have hoped in reversing the hazard to wildlife, if the use of PCB continues to increase.

Very little data are available about PCBs in people, although both Swedish and English scientists have reported finding them in a few samples of human fat. DDT has been found in human fat, liver and brain tissue in widely varying amounts, and PCBs should be looked for in these same tissues, because concern about possible hazards to people are pointed up by the results of the experiments with hormones in birds and rats. Hormones in various animals--including man--are much alike. The

reproductive systems of all mammals are very similar, so that a chemical compound which affects reproduction in one mammal--whether by breaking down a sex hormone or in some other way--may very well have a similar effect in another.

A thorough investigation of the physiological effects of both agricultural and industrial chlorinate hydrocarbons, including their effects on the reproductive systems of both birds and mammals, is urgent. We would do well to act on the basis of data from laboratory animals and wildlife rather than waiting for definitive knowledge from studies of people.

Effects on human reproductive systems would not show up for many years, and would be extremely difficult, if not impossible to relate to one specific cause. However, a study of workers with a long history of exposure to PCBs should be undertaken.

A major effort is also needed to fill the gap in our present understanding of what happens between the manufacture of PCBs and their appearance in the bodies of birds, fish and people. Unless the sources can be established and control measures devised, the problem will resolve itself into the economic desirability of continuing to use PCBs, versus the biological desirability of banning them.

(Dr. [redacted] will add some new data on PCBs in human milk.)

References:

(1) Technical Bulletin O/PL-306, Organic Chemicals Division, Monsanto, St. Louis, 1968.

(2) "Report of a New Chemical Hazard," New Scientist, 32:612, 1966.

(3) Widmark, G., "Possible Interference by Chlorinated Biphenyls" in "Pesticide Residues" Report of the IUPAC Commission on the Development, Improvement, and Standardization of the Methods of Pesticide Residue Analysis, Journal of the Association of Official Analytical Chemists, 50:1069, No. 5, 1967.

(4) Holmes, D.C., J.H. Simmons and J. O'G Tatton, "Chlorinated Hydrocarbons in British Wildlife," Nature, 216:227, 1967 and Holden, A.V. and K. Marsden, Nature, 216:1274, 1967.

(5) Koeman, J.H., M.C. Ten Neoven, De Brauw and R.H. De Vos, "Chlorinated Biphenyls in Fish, Mussels and Birds from the River Rhine and the Netherlands Coastal Area," Nature, 221:1126, March 22, 1969.

(6) Jensen, S. et al "DDT in Marine Animals from Swedish Waters," Nature, 224:247, 1969.

References continued.....

(7) Note: Laboratory procedures in this and subsequent analyses for PCBs are described in the papers listed above and in the author's scientific papers. See especially, Risebrough, R.W., P. Reiche, D.B. Peakall, S.G. Herman and M.N. Kirven, "Polychlorinated Biphenyls in the Global Ecosystem," Nature, 220:1098, 1968 and Risebrough R.W., P. Reiche and H.S. Olcott, "Current Progress in the Determination of the Polychlorinated Biphenyls," Bulletin of Environmental Contamination and Toxicology, 4:192, 1969.

(8) Risebrough, et al, op cit and Jensen, S. et al, "Polychlorinated Biphenyls in the Global Ecosystem," op cit.

(9) Sax, N. Irving, "Dangerous Properties of Industrial Materials," 2nd Ed. Reinhold Pub. Co., New York, 1963, p. 34.

(10) (Reference to come)

(11) Risebrough, R.W. "Chlorinated Hydrocarbons in Marine Ecosystems" in "Chemical Fallout," Morton W. Miller and George C. Berg, editors, Charles C. Thomas, 1969.

(12) Sax, N. Irving, op cit. p. 34.

(13) Jensen, S. et al, op cit.

(14) Koeman et al, op cit.

References continued.....

(15) Miller, J.W. "Pathologic Changes in Animals Exposed to a Commercial Chlorinated Diphenyl," U. S. Public Health Record, 59:1085, 1944 and McLaughlin, Joseph, Jr., Jean-Pierre Marliac, M. Jaqueline Verrett, Mary K. Mutchler and O. Garth Fitzhugh, "The Injection of Chemicals into the Yolk Sac of Fertile Eggs prior to Incubation as a Toxicity Test," Toxicology and Applied Pharmacology, 5:760, 1963.

(16) Street, J.C., F.M. Urry, D.J. Wagstaff and A.D. Bisu, "Comparative Effects of Polychlorinated Biphenyls and Organochlorine Pesticides in Induction of Hepatic Microsomal Enzymes,"