378

Confidential! Not to be released until published!! 10/4/66 EAW.

Silver Spring, Md. 20904

CHLORINATED HYDROCARBONS IN MARINE ECOSYSTEMS

R. W. Risebrough
Institute of Marine Resources
Department of Nutritional Sciences
University of California, Berkeley

ABSTRACT

Polychlorinated biphenyls, which are widely used as plasticizers and in the manufacture of many industrial products, and the DDT compounds have been found to be widely distributed in marine ecosystems of the Pacific Ocean. Marine birds contain higher concentrations than fish; tissues of the Peregrine Falcon have contained the highest concentrations which have been so far recorded. The polychlorinated biphenyls have not yet been detected in samples of airborne particulates but their observed distribution in the sea indicates that they are dispersed by wind currents. Their effects upon natural populations, including man, are as yet unknown. The DDT compounds are also accumulating in the sea and are present in highest concentrations in marine birds. Their distribution indicates that coastal areas are not the primary sources of contamination of the pelagic species. A quantitative approach to the problem of aerial transport of pesticides to the sea has been made by analysing the pesticide content of airborne particulates. The results indicate that wind transport can account for the observed distribution of DDT compounds in the California waters and that the amount of pesticides entering the tropical Atlantic as fallout from the Northeast Trades is comparable to that entering the sea from a major river system.

REPORT

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 produced subtle but profound changes in our concepts of remoteness, isolation, and of our own position in the world-wide ecosystem. There has never been any question that local ecosystems may be irreversibly changed by the introduction of synthetic chemicals, whether these introductions be purposeful or incidental. The sea is the dominant feature of the world-wide ecosystem. The accumulation of significant amounts of several pollutants in marine organisms, pollutants which are non-polar and therefore water-insoluble but lipid-soluble, not only elicits an uncertainty about the long-term utilization of the sea as a source of human food, but has suddently raised the question of the ultimate survival of a number of species of sea birds. These comprise a very large fraction of the world's wildlife and doubts about their future would have been considered preposterous and untenable only 3 or 4 years ago.

DSW 026653

STLCOPCB4010614

Perhaps because fish kills have provided the most dramatic evidence of pesticide contamination, pesticide distribution studies in the United States and elsewhere have frequently consisted of monitoring the levels in major river systems [2,7]. In spite of the low solubility of chlorinated hydrocarbons in water [4], their tendency to pass into the vapour phase [16,1,5,6,12], their persistence in soils [3,10,18], and their presence in very low concentrations in some streams draining areas of intensive application [17], rivers do transport large amounts of stable, persistent pesticides to the sea. A large proportion of these pesticides are adsorbed to silt particles which are part of the runoff from areas of application. Thus, the Sacramento and San Joaquin rivers, which drain the Central Valley of California, one of the most heavily pesticided areas of the world, annually bring about 1,900 kg of chlorinated hydrocarbons into San Francisco Bay [2,21], and the Mississippi contributes about 10,000 kg of the Gulf of Mexico [7,21].

When the Institute of Marine Resources began a study of the distribution of chlorinated hydrocarbons in marine fish, we expected to find, therefore, much higher levels in the fish from San Francisco Bay than in the fish from the Pacific Ocean [24]. The data of Table I, however, show that collections of the Northern Anchovy and of the English Sole from the coastal waters contained as much as or significantly more total DDT residue than did the collections from San Francisco Bay. Total DDT concentrations were highest in the collection from waters off Los Angeles, but anchovies from the Channel Islands area off Port Hueneme also contained significantly more residue than the anchovies of San Francisco Bay. Since no major river system enters the Pacific Ocean from Southern California, the source of these pesticides could not be agricultural drainage waters. Similarly, the data of Table II, which set forth the distribution of total DDT residues and of polychlorinated biphenyls in several collections of marine fish, including Shiner Perch from San Francisco Bay, Hake, Jack Mackerel and English Sole from the coastal waters of California, Bluefin and Yellowfin Tunas from waters off Baja California and Central America, and Skipjack Tuna from South America and the Central Pacific, show that almost all total DDT residue levels fall within the range of 0.2 and 2.0 parts per million, wet weight, including those in the Bluefin and Yellowfin Tunas, which are more pelagic. Only the Yellowfin Tuna from the Galapagos region and the Skipjack Tuna contained lower total pesticide residues. The majority of fresh-water fish from Wisconsin analysed by the Wisconsin Conservation Department in 1965 contained less than 0.2 ppm of total DDT residue [27]. Moreover, fish from a stream in Wisconsin which drained an orchard where chlorinated hydrocarbons had been intensively applied over the years contained lower pesticide residue than did the marine fish from the Pacific Ocean [17]. These combined observations indicate that it is very unlikely that pesticides in marine fish have originated only in coastal waters contaminated by local agricultural runoff.

The polychlorinated biphenyls (PCB) occurring in fish and other marine organisms are assumed to be industrial pollutants. They are used extensively in industry as plasticizers and in the manufacture of paints, resins, electrical insulators and other products, and are available in railway car amounts. Since they are very stable, resist degradation, have significant vapour pressures, are poorly soluble in water and highly soluble in lipid, it is inevitable that they should be concentrated in biological systems. Their chemical structure is

STLCOPCB4010615

in some respects similar to that of several chlorinated hydrocarbon pesticides, including DDT and the benzene hexachlorides, so that it is to be expected that they would behave similarly in their movements through ecosystems. They are highly toxic to man when inhaled as vapours [9,25] and the more heavily chlorinated components have greater toxicity. Their presence in concentrations as high as one part per million on a wet weight basis, and several times higher on a lipid weight basis, in species which are used as human food, should be a matter of considerable concern. At the present time it is not entirely clear how the PCB enter marine food chains, but they could easily be introduced into the atmosphere as vapours during manufacturing processes, when high temperatures increase vapour pressure, or by gradual volatilization over a period of time or even when materials containing them are incinerated.

The PCB peaks had been evident in chromatograms of our extracts of marine fish and birds but remained unidentified until late in 1967. Extracts of tissues and of an unhatched egg of a Peregrine Falcon, a species whose decline and local extinction has frequently been attributed to pesticides, contained exceptionally high amounts of the unknown compounds [22]. Attempts to identify them were unsuccessful until reports appeared from Sweden [30] and from Great Britain [14] that PCB had been identified in wildlife with the use of mass spectrography and gas-liquid chromatography, respectively. The published chromatogram [14] was virtually identical with those we had obtained from the Peregrine Falcons extracts.

The commercial PCB preparations are mixtures of several compounds. Of these, those having the following retention times relative to p,p'-DDE on DC-200 and QF-1 columns have been detected in marine fish and birds:

DC-200: 1.25, 1.48, 1.75, 2.05, 2.41, 2.50, 2.90, 3.41, 3.88, 5.53

dieldrin: 1.00; p,p'-DDD: 1.27; p,p'-DDT: 1.68

QF-1: 1.10, 1.33, 1.40, 1.65, 1.72, 2.14, 2.59, 3.23, 3.88, 4.84

dieldrin: 1.49; p,p'-DDD: 1.75; p,p'-DDT: 1.91.

The retention times of the three principal peaks are underlined. Similar values for the DC-200 column have been reported from extracts of seals from the North Atlantic [13]. Because of the similarity of retention times, it is evident that large amounts of PCB will interfere with the determination of p,p'-DDD and p,p'-DDT on DC-200 columns and with p,p'-DDD on QF-1 columns. Conversely, in many extracts, the PCB content can be determined only after DDD and DDT have been removed by saponification with alcoholic KOH. The quantitative measurements of the PCB compounds were made as follows: it was assumed that each produces the same peak height in the electron capture detector as the same amount, by weight, of p,p'-DDE. After summing the contributions of the individual peaks, the total was multiplied by 4.2. Measurements of standard solutions by this method yielded results close to the predicted values and to those obtained by measuring the total halogen content with the microcoulometric detector.

Measurements of the PCB in the fish extracts were made from pooled extracts. Unlike the DDT compounds, the PCBs appear to be relatively more abundant in San

STLCOPCB4010616

Francisco Bay, but their presence in Yellowfin Tuna taken off Central America indicates a wide geographical distribution.

Concentrations of the chlorinated hydrocarbons, both pesticide and PCB, tend to be an order of magnitude higher in marine birds than in fish. In Table III are listed the total DDT and PCB concentrations in tissues and in entire birds of several species. Table IV lists the DDT and PCB content of those eggs so far analysed. The latter values will eventually be also expressed as parts per million, lipid weight.

From the data in Tables III and IV it is evident that both DDT and the polychlorinated biphenyls are widely distributed among marine birds, which are the terminal carnivores of a complex variety of food chains in the sea. Sooty and slender-billed shearwaters are pelagic species which breed in New Zealand and Australia, respectively and which spend the southern winter in the northern Pacific. Rhinoceros Auklets and Ancient Murrelets breed on the coasts of British Columbia and Alaska, whereas Kittiwakes, Fulmars and Red Pharalopes breed in Alaska and the Canadian Arctic. All of these specimens were collected in California, so that it is not clear how much DDT and PCB the birds had ingested on their breeding grounds. In the pelagic bird species, in most of the fish from the ocean and in the other specimens from areas remote from sites of application, such as Baja California, the ratios of DDT to PCB are of the same order of magnitude and most values are between 5 and 15. If both PCB and DDT were dispersed around the world by the same transport system, their relative concentrations in "remote" areas would very likely be similar. Additional data are needed to support this hypothesis; eggs from tropical regions and from Antarctica have been obtained and will eventually be analysed.

In many of the specimens from San Francisco Bay and San Diego Bay this ratio is close to one. Absolute concentrations of PCB also tend to be higher, suggesting that the most important sources of contamination are local.

The sample sizes of Tables III and IV are very small; many samples, however, remain to be analysed. Only tentative suggestions can be made from the data, but a consistent pattern is evident. Thus, the Western Gull egg from San Francisco Bay and the Pigeon Guillemot egg from the coast near San Francisco contained more PCB than the egg of each species from the Farallons. Of the two night heron eggs analysed, one had an "ocean" profile: high DDE, low p, p'-DDT and DDD, high DDT/PCB ratio; the other had a "bay" profile. The Caspian and Forster's Terns nest side by side on the dikes in San Diego Bay, yet the DDT/PCB ratio is much lower in the Caspian Terns, which feed primarily along the coast, than in the Forster's Terns, which feed along the brackish and fresh-water dikes and canals. The two California Peregrine Falcons had both been feeding in San Francisco Bay prior to capture; in both the ratios of DDT to PCB are near unity.

The high DDT concentrations recorded in the Bermuda Petrel [32] and in the Skua from Antarctica [26], which are species of the high seas, also indicate that coastal regions were not the primary source of contamination.

A quantitative approach to the problem of aerial transport of pesticides over the sea has been made by measuring pesticide concentrations in airborne

STLCOPCB4010617

particulates [21]. Over the past year, Goldberg, Griffin and others at the Scripps Institution of Oceanography have collected airborne particulate matter on nylon mesh screens coated with glycerin which had been mounted on the Scripps pier in La Jolla. Dust collected on such screens in several areas of the world had previously been shown to contain the mineral talc in concentrations much higher than expected on the basis of its natural distribution [31]. Talc has been extensively used as a diluent for pesticides, in addition to other industrial applications. Moreover, the rate at which talc has been deposited on glaciers shows a significant increase after 1940 [31]. Analysis of the airborne dust collected on the Scripps Pier showed that pesticides, predominately p,p'-DDT were present in concentrations ranging from one to eighty-one parts per million, corresponding to a mean value of 7×10^{-11} grams per cubic meter of air. The method fractionates against those materials carried on particles less than several micra or present as vapours. This value is therefore a lower limit of the amount of chlorinated hydrocarbons in the air. No PCB peaks were observed in the chromatograms of the dust extracts. These were then pooled, concentrated, and saponified in order to degrade DDT and DDD. PCB was not present in the saponified extracts and a maximum concentration of 5 ppb was calculated for the dust samples, 10,000 times lower than that of the total pesticides. Since the ratios of PCB to DDT observed in fish and birds is much higher, it would appear that the PCBs remain as vapours in the air and do not adsorb to particulate matter. In the near future we hope to obtain air samples which have been passed through a cold chamber in which PCBs, pesticides and other compounds present as vapours would condense.

The winds at the Scripps pier in La Jolla are predominantly landward with an unknown fraction of air from nearby agricultural areas. Although the lack of knowledge about air circulation patterns and fallout rates makes difficult at the present time a calculation of the amounts of pesticides brought to the coastal waters of California by winds, such a transport system could account for the unexpected geographical distribution of the DDT compounds in fish.

The Science Research Council of the United Kingdom had earlier mounted a similar screen on the eastern tip of the island of Barbados in an attempt to collect extraterrestrial dust of meteorite origin. The Northeast Trades at Barbados have blown over 5000 kilometers of the tropical Atlantic and it was therefore considered highly unlikely that any dust accumulating on the screen could be of continental origin. This assumption was, however, in error, since significant quantities of airborne particulate material were collected on the screen which upon mineralogical and biological examination proved to be most likely from Africa and Europe. Fallout rates of the dust over the tropical Atlantic and its contribution to the bottom sediments were calculated [8]. Gram samples were subsequently made available to us for analysis. The concentrations of pesticides in the dust ranged from less than one part per billion to 164 ppb, with an average value of 41. Knowledge of the fallout rate over the ocean made possible an estimate of the quantity of pesticides entering the tropical Atlantic between the Equator and 30° N. This figure, a minimum value since the recovery of very small particulate materials was low, was 600 kilograms a year, which compares with the value of 1,900 kilograms entering San Francisco Bay in the San Joaquin and Sacramento Rivers [21]. It cannot be concluded that these pesticides had originated only in Africa; the DDT residues accumulating

STLCOPCB4010618

the rare and declining Bermuda Petrel [32] might have come from any part of the world.

Most of the chlorinated hydrocarbon content of the particulate material of marine air consists of p,p'-DDT, p,p'-DDE, however, comprises the large proportion of the total DDT in marine fish and birds. The percentage of DDE is lower in fish from San Francisco Bay and Puget Sound; DDT and DDD are comparatively more abundant. DDE, however, comprises only 22% of the total DDT residue entering San Francisco Bay in the San Joaquin River [2] and in bottom mud, where anaerobic conditions might prevail, DDT can be expected to be converted to DDD rather than to DDE [15,29]. From mammalian and insect toxicity studies it has been concluded that DDE is relatively harmless. Recent research at the Patuxent Wildlife Research Center, however, have indicated that the toxicity of DDE to birds is much higher than expected and may be about one half that of p,p'-DDT [28]. Like p,p'-DDT, p,p'-DDE is capable of inducing liver epoxidase enzymes [11]. Such enzymes degrade sex hormones by hydroxylating them [19] and thereby may affect calcium metabolism. No evidence of abnormal calcium metabolism has yet been observed in sea birds, but a significant decrease in eggshell weight of several birds of prey in Britain after the Second World War is evidence of a fundamental change in the environment [20].

The effects of the PCB compounds upon natural populations remain unknown. The highest concentrations so far recorded were in an adult female Peregrine Falcon from California, where the species has recently undergone a sharp decline [22].

Application of the persistent biocides and the release of nondegradable waste products into the environment have frequently been justified by arguments that point out that local populations of organisms remain relatively uncontaminated and that, especially in the case of the chlorinated hydrocarbon insecticides, even the persistent compounds disappear with time. It is abundantly clear that these arguments have become misleading, irrelevant, and wrong. Pollutants do go everywhere. Those which are non-polar, water-insoluble and which have finite vapour pressures will eventually appear in marine food chains. The DDT compounds and the polychlorinated biphenyls have already done so to an alarming degree.

STLCOPCB4010619

TABLE I

esidues[a] in collections of Northern Anchovy (Engraulis mordax) and ish Sole (Parophrys vetulus) from San Francisco Bay and California stal waters.

Species locality, date	N	Mean Wt (g)	p,p'-DDE[b]	Total DDT[b]
Northern Anchovy				
San Francisco Bay July 29, 1965	17	12.4 (6-22)	0.21 ±0.05	0.59 ±0.11
San Francisco Bay November 4, 1965	29	4.0 (2-5)	0.11 ±0.05	0.33 ±0.04
Monterey November 30, 1965	30	26.5 (21-35)	0.68 ±0.19	0.90 ±0.22
Morro Bay June 16, 1965	29	25.9 (19-33)	0.45 ±0.12	0.74 ±0.22
Port Hueneme February 24, 1966	15	11.6 (2.8-21.5)	2.44 ±0.77	3.04 ±1.00
Terminal Island Los Angeles, June 25, 1965	44	11.7 (6.5-20)	10.2 ±2.1	14.0 ±1.9
English Sole				
San Francisco Bay July 29, 1965	18	14.3 (5-35)	0.14 ±0.02	0.55 ±0.07
San Francisco Bay November 4, 1965	33	17.3 (7.5-53)	0.13 ±0.03	0.55 ±0.12
San Francisco lightship December 1, 1965	15	253)175-306)	0.12 ±0.03	0.19 ±0.04
Monterey February 15, 1966	15	195 (89-262)	0.53 ±0.13	0.76 ±0.16

[a] from R. W. Risebrough, D.B. Menzel, D. J. Martin, Jr., and H. S. Olcott, in preparation. DDT residues include the two isomers of DDT,p,p'-DDT

DSW 026659

STLCOPCB4010620

TABLE I, continued

and o,p'-DDT and their metabolic derivatives: p,p'-DDE, o,p'-DDE p,p'-DDD and p,p'-DDMU.

[b] wet weight parts per million, means, standard errors, 95% confidence limits

DSW 026660

STLCOPCB4010621

TABLE II

DDT [a] and polychlorinated biphenyl (PCB) residues in marine fish. [b]

Species, locality, date	N	Mean Wt. (g)	Total DDT	% p,p'-DDE	PCB	DDT/PCB
Northern Anchovy						
Terminal Island June 25, 1965	44	11.7 (6.5-20)	14.0 ±1.9	83	1.0	14
Shiner Perch						
San Francisco Bay October 20, 1965	14	5.5 (4-8)	1.0 ±0.1	28	1.2	0.8
San Francisco Bay October 20, 1965	10	26.7 (10-48)	1.4 ±0.3	35	0.4	3.5
San Francisco Bay November 4, 1965	15	15.3 (8-49)	1.1 ±0.1	33	1.2	0.9
English Sole						
San Francisco Bay July 29, 1965	18	14.3 (5-35)	0.55 ±0.07	25	0.11	5
San Francisco Bay November 4, 1965	33	17.3 (7.5-53)	0.55 ±0.12	24	0.11	5
San Francisco lightship, Dec. 1, 1965	15	253 (175-306)	0.19 ±0.04	63	0.05	4
Monterey February 15, 1966	15	195 (89-262)	0.76 ±0.16	70	0.04	19
Jack Mackerel						
Channel Islands November 22, 1965	31	81.8 (45-141)	0.56 ±0.10	57	0.02	28
Hake						
Puget Sound January 29, 1966	22	281 (185-350)	0.18 ±0.05	23	0.16	1.1

DSW 026661

STLCOPCB4010622

TABLE II, continued

Hake						
Channel Islands February 24, 1966	6	384 (61-872)	1.8 ±1.1	68	0.12	15
Bluefin Tuna[c]						
Body muscle	7	--	0.56 ±0.24	45	0.04	14
Liver	9	--	0.22 ±0.13	45	0.04	6
Yellowfin Tuna						
Liver[d]	13	--	0.07 ±0.02	13	ND[e]	>7
Liver[f]	13	--	0.62 ±0.19	30	0.04	15
Skipjack Tuna						
Liver[g]	3	--	0.057	23	0.1	0.6
Body Muscle[g]	13	--	0.051 ±0.014	18	ND	>30
Liver[h]	25	--	0.056 ±0.023	11	ND	>30
Liver[i]	12	--	0.029 ±0.008	21	ND	>20

[a] from Risebrough et al. [24] Concentrations in wet weight, parts per million. Means, standard errors, 95% confidence limits

[b] Northern Anchovy (Engraulis mordax), Shiner Perch (Cymatogaster aggregata), English Sole (Parophrys vetulus), Pacific Jack Mackerel (Trachurus symmetricus), Hake (Merluccius productus), Bluefin Tuna (Thunnus thynnus), Yellowfin Tuna (Thunnus albacares), Skipjack Tuna (Euthynnus pelamis).

[c] Isla Geronimo, Baja California, Aug. 29, 1965

[d] Tower Island, Galapagos Archipelago, Nov. 6, 1965

[e] N.D. not detected, less than 0.01

DSW 026662

STLCOPCB4010623

TABLE II, continued

[f] Central America (90° W, 8° N) Aug. 23, 1965

[g] Hawaii, November, 1965

[h] Galapagos Islands, November and December, 1965

[i] Cape Pasado, Ecuador, September 1, 1965

DSW 026663

STLCOPCB4010624

TABLE III

DDT[a] and PCB residues in marine birds[b] and in the Peregrine Falcon[k]

Species, locality, date	Total DDT[c]	% DDE	PCB	DDT/PCB
Cassin's Auklet[d]	5.8	98	0.16	36
Ancient Murrelet[e]	0.75	90	0.15	5
Fulmar[f]	0.41	76	0.08	5
Fulmar[f]	3.4	89	0.34	10
Red Phalarope[g]	0.78	79	0.10	8
Rhinoceros Auklet[h]	2.7	97	0.36	8
Slender-billed shearwater[i]	32.0	92	2.1	15
Sooty shearwater[j]	12.3	94	1.2	10
Sooty shearwater[j]	10.3	86	0.9	12
Peregrine Falcon[k]				
breast muscle, second year female, migrant from Arctic	104	99	22	4.5
breast muscle, immature California	13	99	10.5	1.2
breast muscle, adult female, California	112	98	109	1.0

a: from Risebrough et al. [23] and Risebrough, Kirven and Herman [22]; b: entire bird analysed, except Peregrine Falcons; c: includes p,p'-DDT, p,p'-DDD, p,p'-DDE, p,p'-DDMU, p,p'-DDE; parts per million, wet weight; d: Ptychoramphus aleuticus, adult female, Farallon Islands, April, 1966; e: Synthliboramphus antiquus, Monterey Bay, Nov. 1, 1966; f: Fulmarus glacialis, Monterey Bay, Nov. 1, 1966; g: Phalaropus fulicarius, Monterey Bay, Nov. 1, 1966; h: Cerorhinca monocerata, Monterey Bay, Nov. 1, 1966; i: Puffinus tenuirostris, Monterey Bay, Dec. 12, 1966; j: Puffinus griseus, Monterey Bay, Nov. 1, 1966; k: from Risebrough, Kirven and Herman [22].

DSW 026664

STLCOPCB4010625

TABLE IV

DDT and PCB content in eggs of several bird species.

Species, locality	N	Total DDT[a]	% p,p'-DDE	PCB[a]	DDT/PCB
Brandt's Cormorant[b] (Phalacrocorax penicillatus)					
Farallon Islands	17	326	91	113	2.9
Pelagic Cormorant (Phalacrocorax pelagicus)					
San Mateo Co. Calif.	2	128 (125-130)	90	62 (48-75)	2.1
Murre Uria aalge					
Farallon Islands	6	1945 (932-3621)	96	558 (364-1010)	3.5
Pigeon Guillemot Cepphus grylle					
Farallon Islands	1	110	95	20	5.5
San Mateo Co.	1	103	91	62	1.7
Cassin's Auklet Ptychoramphus aleuticus					
Farallon Islands	2	147 (127-167)	97	15 (12-18)	10
Western Gull Larus occidentalis					
Farallon Islands	1	423	95	118	3.6
San Mateo Co.	1	235	94	112	2.1
San Francisco Bay	1	458	87	480	0.95

DSW 026665

STLCOPCB4010626

TABLE IV, continued

Black-crowned Night Heron *Nycticorax nycticorax*					
San Francisco Bay	1	541	89	330	1.6
	1	869	99	24	36
Caspian Tern *Hydroprogne caspia*					
San Francisco Bay	2	1269 (1216-1322)	89	805 (660-950)	1.7 (1.3-2.0)
San Diego Bay	5	1430 (991-2430)	88	1010 (550-1600)	1.4
Forsters Tern *Sterna forsteri*					
San Diego Bay	2	665 (598-732)	89	114 (91-137)	5.8
Least Petrel *Halocyptena microsoma*					
Baja California	2	30 (23-37)	84	3.1 (1.2-5.0)	10
Peregrine Falcon[c] *Falco peregrinus*					
Baja California	1	4830	97	471	10

a: total micrograms; b: samples pooled for PCB analysis, p,p'-DDE content ranged from 63 to 1240 micrograms; c: from Risebrough, Kirven, Herman [22].

STLCOPCB4010627

REFERENCES

[1] ACREE, F., JR., BEROZA, M., BOWMAN, M.C. J. Agr. Food Chem. 11 (1959) 278.

[2] BAILEY, T.E., HANNUM, J.R., J. Sanit. Eng. Div. Amer. Soc. Civil Engrs. 93 (1967) 27.

[3] BARTHA, R., LANZILOTTA, R.P., PRAMER, D., Appl. Microb. 15 (1967) 67.

[4] BOWMAN, M.C., ACREE, F., JR., CORBETT, M.K., J. Agr. Food Chem. 8 (1960) 406.

[5] BOWMAN, M.C., ACREE, F., JR., LOFGREN, C.S., BEROZA, M., Science 146 (1964) 1480.

[6] BOWMAN, M.C., ACREE, F., JR., SCHMIDT, C. H., BEROZA, M., J. Econ. Entomol. 52 (1959) 1038.

[7] BREIDENBACH, A.W., GUNNERSON, C.G., KAWAHARA, F.K., LICHTENBERG, J.J., GREEN, R.S., Public Health Rept. 82 (1967) 139.

[8] DELANEY, A.C., DELANEY, A.C., PARKIN, D.W., GRIFFIN, J.J., GOLDBERG, E.D., REIMANN, B.E.F., Geochim. Cosmochim. Acta 31 (1967) 885.

[9] Documentation of Threshold Limit Values, 41 (Committee on Threshold Limit Values, American Conference of Governmental Industrial Hygenists) 1966.

[10] FAHEY, J.E., BUTCHER, J.W., TURNER, M.E., Pesticides Monit. J., 1 4 (1968) 30.

[11] GILLETT, J.W., CHAN, T.M., TERRIERE, L.C., Agr. Food Chem. 14 6 (1966) 540.

[12] HARRIS, C.R., LICHTENSTEIN, E.P., J. Econ. Entomol. 54 (1961) 1038.

[13] HOLDEN, A.V., MARSDEN, K., Nature 216 (1967) 1274.

[14] HOLMES, D.C., SIMMONS, J.H., TATTON, J.O'G., Nature 216 (1967) 227.

[15] JOHNSON, B.T., GOODMAN, R.N., GOLDBERG, H.S., Science 157 (1967) 560.

[16] LICHTENSTEIN, E.P., ANDERSON, J.P., FUHREMANN, T.W., SCHULZ, K.R., Science 159 (1968) 1110.

[17] MOUBRY, R.J., HELM, J.M., MYRDAL, G.R., Pesticides Monit. J. 1 4 (1968) 27.

[18] NASH, R.G., WOOLSON, E.A., Science 157 (1967) 924.

[19] PEAKALL, D.B., Nature 216 (1967) 505.

[20] RADCLIFFE, D.A., Nature 215 (1967) 208.

[21] RISEBROUGH, R.W., HUGGET, R.J., GRIFFIN, J.J., GOLDBERG, E.D., Science 159 (1968) 1233.

[22] RISEBROUGH, R.W., KIRVEN, M.N., HERMAN, S.G., in preparation

[23] RISEBROUGH, R.W., MENZEL, D.B., MARTIN, D.J., JR., OLCOTT, H.S., Nature 216 (1967) 589.

[24] RISEBROUGH, R.W., MENZEL, E.B., MARTIN, D.J., JR., OLCOTT, H.S., in preparation.

[25] SAX, N.I., Dangerous Properties of Industrial Chemicals, Reinhold, New York, (1963).

DSW 026667

STLCOPCB4010628

REFERENCES, continued

[26] TATTON, J.O'G., RUZICKA, J.H.A., Nature 215 (1967) 346.
[27] THOMPSON, D.R., Wisconsin Conservation Department Survey Report, February 14, 1966.
[28] United States Department of the Interior, Fish and Wildlife Service; Wildlife Research Problems Programs Progress 1966 Resource Publication 43, Washington, D.C.
[29] WEDEMEYER, G., Science 152 (1966) 647.
[30] WIDMARK, G., J. Ass. Off. Anal. Chem. 50 (1967) 1069.
[31] WINDOM, H., GRIFFIN, J.J., GOLDBERG, E.D., Environ. Sci. Technol. 1 (1967) 923.
[32] WURSTER, C.F., JR., WINGATE, D.B., Science 159 (1968) 979.
[33] The National Science Foundation, (grant GP 6362) provided financial support. I thank P. L. Ames, E. D. Goldberg, S. G. Herman, M. N. Kirven, H. S. Olcott, P. Reiche and R. L. Rudd for their assistance. The Monsanto Chemical Company generously provided samples of Aroclor 1254 and 1262.

DSW 026668

STLCOPCB4010629