OCCURRENCE AND SIGNIFICANCE OF DDT COMPOUNDS AND OTHER CONTAMINANTS IN FISH, WATER, AND SEDIMENT FROM THE YAKIMA RIVER BASIN

by

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EXECUTIVE SUMMARY

Routine monitoring between 1979 and 1984 done as part of the Washington State Department of Ecology Basic Water Monitoring Program (BWMP) showed higher levels of DDT and metabolites DDE and DDD in Yakima River fish than elsewhere in Washington State. In response to requests by the Ecology Central Regional Office, the Yakima Indian Nation, and the National Marine Fisheries Service, a study of this problem was conducted in 1985 by Ecology's Water Quality Investigations Section. Study objectives were to evaluate the hazards to human health and aquatic life, identify sources, and determine if contamination was primarily due to recent illegal use or historical applications.

Fish, water, and bed sediment were analyzed. Resident fish were collected at four locations on the river--Cle Elum (river mile [r.m.] 179-181), Wymer (r.m. 134-136), Buena (r.m. 93-95), and Kiona/Benton City (r.m. 20-23). Returning spring chinook and out-migrating juvenile salmonids also were collected. Whole fish, muscle, and eggs were analyzed. Water and sediment samples were collected from the Yakima main stem and from major tributaries (including irrigation return flows) considered to have the greatest potential for DDT contamination.

Target chemicals analyzed were DDT, DDE, and DDD, fifteen additional persistent organochlorine pesticides, polychlorinated biphenyls (PCBs), and mercury. Selected water samples were screened for a broad spectrum of toxic chemicals.

The major organochlorine compounds detected in fish were DDT, DDE, dieldrin, and PCB-1260. DDD, endosulfan, and endrin also were detected in a few samples. Fish in the lower river had higher concentrations than in the upper river. Salmonids had substantially lower contamination than resident species.

Concentrations of t-DDT (i.e., total DDT; the sum of DDT, DDE, and DDD), dieldrin, PCB-1260, and mercury in edible fish tissue were well below the Food and Drug Administration (FDA) "action levels" of 5,000 ug/Kg (parts per billion) t-DDT, 300 ug/Kg dieldrin, 2,000 ug/Kg PCBs, and 1,000 ug/Kg mercury. FDA has not established an action level for endosulfan, also detected at low concentrations in a few edible tissue samples,

U.S. Environmental Protection Agency (EPA) methodology for assessing the risk to humans from ingesting contaminants was used to evaluate the relative risks among the contaminants identified in fish from the lower river. Based on animal studies, EPA's Carcinogen Assessment Group has classed DDT, DDE, DDD, dieldrin, and PCBs as probable human carcinogens. Endosulfan and mercury are not carcinogenic. Risk analysis showed carcinogenic risks from dieldrin and PCB-1260, although present at relatively low concentrations, were greater than for DDT compounds. Endosulfan and mercury were within EPA acceptable daily intakes (ADI's) at the highest ingestion rate evaluated.

The potential for adverse effects on fish reproduction in the Yakima main stem was evaluated in part by comparing the concentrations of t-DDT, dieldrin, and PCB-1260 measured in egg samples with effects thresholds (e.g., hatching success, fry mortality) indicated in the literature. All concentrations were below apparent effects thresholds.

The potential for adverse effects on predators (i.e., fish-eating birds) feeding on Yakima River fish was assessed by comparing concentrations of t-DDT, dieldrin, endosulfan, endrin, PCB-1260, and mercury in whole fish samples with National Academy of Sciences (MA S) maximum recommended concentrations. With the exception of t-DDT, concentrations of these contaminants were generally within NAS recommendations.

Concentrations of t-DDT in whole fish from the lower Yakima River at Kiona were in the 1,100 to 3,000 ug/Kg range which exceeds the 1,000 ug/Kg concentration recommended by NAS. This raised the concern of possible adverse effects on reproduction of fish-eating and raptorial birds due to the eggshell-thinning effect of DDE. Based on a recent review of this subject, it appears likely that predatory birds which are sensitive to DDE and feed exclusively on fish from the lower Yakima River would produce eggs with shell thickness somewhat below normal. The reduction in shell thickness anticipated, however, would probably not be sufficient to prevent maintenance of stable populations.

DDT, DDE, DDD, dieldrin, and endosulfan were detected in water samples. The detection of these compounds was almost exclusively limited to tributaries. DDE and dieldrin were detected in only one instance in the main stem of the Yakima River; concentrations were in the 0.006 to 0.007 ug/L (parts per billion) range. Maximum concentrations in tributaries were 0.07 ug/L t-DDT, 0.02 ug/L dieldrin, and 0.16 ug/L endosulfan. The broad spectrum screen of selected water samples revealed only one additional compound of potential concern--diazinon. This organophosphorus pesticide was detected at 1.7 ug/L in Birchfield Drain.

Of the eleven tributaries monitored, Sulphur Creek, Birchfield Drain, Granger Drain, and Spring/Snipes Creek were identified as sources of DDT compounds. Sulphur Creek contributed the largest load of t-DDT to the Yakima River. Six tributaries were sources of dieldrin. Endosulfan was detected only in Birchfield Drain. The maximum combined loads measured for these compounds were 0.1 pound/day t-DDT, 0.03 pound/day dieldrin, and 0.05 pound/day endosulfan.

The concentrations of DDT compounds, dieldrin, and endosulfan measured in water were below those known to be acutely toxic to aquatic life. However, concentrations in a number of tributaries exceeded EPA criteria for protection of freshwater life for chronic (long-term) exposure and NAS recommended maximum concentrations for water. This suggests aquatic life may be adversely affected, and that resident fish in these tributaries may be unsuitable for consumption by wildlife. Bioaccumulation of DDT compounds and dieldrin by fish in tributaries with the highest concentrations may result in FDA action levels for human consumption being exceeded. There did not appear to be evidence of a toxicity problem due to organochlorine concentrations in water in the main stem of the Yakima.

DDT compounds and dieldrin were the predominant organochlorines detected in bed sediments from the Yakima main stem and tributaries. Maximum concentrations occurred in Sulphur Creek where 234 ug/Kg t-DDT and 14.9 ug/Kg dieldrin were measured. Endosulfan, aldrin, and isodrin also were detected in one sample each. An extremely high aldrin concentration of 1,065 ug/Kg was present in the sediment sample from Spring/Snipes Creek. Criteria for protection of aquatic life have not been developed for organochlorines in sediment.

DDE is more persistent than DDT or DDD. Its predominance in Yakima River fish indicates the source is primarily historical application. Although there may be instances of illegal use in the Yakima drainage, the following evidence supports the conclusion that the source is primarily historical:

- o Neither FDA nor the Washington State Department of Agriculture has detected field application of DDT since the ban in 1972.
- o Localized contamination was not observed in this study.
- o Concentrations in Yakima River fish are not extremely high.

U.S. Geological Survey (USGS) and U.S. Fish and Wildlife Service historical data show substantial decreases in the concentrations of both t-DDT and dieldrin in Yakima River fish and water since the early 1970s. USGS historical data and results from the present survey indicate transport of DDT compounds and dieldrin to the Yakima River occurs primarily during the irrigation season. Examination of DDT:DDE ratios in water and-sediment samples from tributaries suggests that DDT has a long half-life in Yakima soils. The potential for the miticide dicofol (trade name Kelthane) being a source of DDT compounds is discussed.

Study recommendations include erosion control to reduce losses of DDT and dieldrin from agricultural land; improved management of endosulfan and diazinon--two currently used pesticides; study of the potential for sublethal effects on fish in the lower river; additional sediment samples in Spring/Snipes Creek to verify high aldrin concentrations; bioassay of bed sediments to screen for toxicity; and collection of fish samples from the Columbia River pool below the Yakima confluence as part of Ecology's 1987 BWMP survey.

INTRODUCTION

The Washington State Department of Ecology began doing chemical analyses on fish and aquatic invertebrates in 1979 as part of its Basic Water Monitoring Program (BWMP). These analyses, done annually, have consistently demonstrated the presence of p,p'-DDT and two metabolites, p,p'-DDE and p,p'-DDD, in Washington State freshwater fishes (Hopkins, et al., 1985).

The detection of DDT compounds a decade or more after the use of this pesticide was banned in the United States is not unusual. Because of the former widespread use of DDT, continued use in some countries, and the persistence of both parent compound and metabolites, these contaminants are still routinely found in fish and other organisms throughout the world (McEwen and Stephenson, 1979).

Of special interest in the Ecology data, however, was the elevated level of DDT compounds in fish from the Yakima River. Table 1 summarizes the data available as of early 1985.

Table 1. Ecology data on t-DDT (DDT+DDE+DDD) in fish † tissue samples from Washington State rivers 1978-1984 (ug/Kg, wet; i.e., parts per billion).

		V	hole Fi	sh	Muscle			iver				
Location	Year(s)	n =	Mean	Range	Year(s)	n =	Mean	Range	Year	n =	Mean	Range
Yakima R @ Birchfield Drain	1983	3	2,900	1,000-5,100	1983-84	6	1,500	570-2,800	1984	4	8,100	1,800-26,000
Yakima River @ Kiona	1980-83	8	1,800	580-3,100	1984	2	2,100	2,000-2,200	1984	2	14,000	13,000-15,000
Okanogan R. @ Okanogan	1978-83	2	1,300	810-1,800	1984	2	2,500	1,800-3,200	1984	2	9,000	3,100-15,000
Lk. Chelan Outlet	1982-83	4	2,300	1,400-4,500	1983-84	4	880	460-1,200	1984	2	4,200	2,100-6,400
Wenatchee R @ Wenatchee	1982	2	480	410-560	1984	2	850	300-1,400	1984	2	1,100	920-1,200
Other Eastern Wash. rivers	1978-83	27	260	8.2-990	1983-84	9	250	44-780	1984	5	360	110-670
Western Wash.	1979-83	23	45	0.8-220	1983-84	7	410	1.4-1,500	1984	4	970	18-2,000

Source: Hopkins, et al. (1985).

In light of these findings and in response to requests from the Ecology Central Regional Office, the Yakima Indian Nation (Yallup, 1984), and the National Marine Fisheries Service (Evans, 1985) for an investigation of the problem, Ecology's Water Quality Investigations Section initiated a series of

[†]Primarily largescale sucker, bridgelip sucker, northern squawfish, and mountain whitefish.

surveys in the Yakima River in March of 1985. The primary objectives of this work were two-fold:

First, to determine the concentrations of DDT and other organochlorine compounds in fish and invertebrates from different parts of the Yakima River and evaluate the significance for human health and well-being of aquatic life.

Second, to measure concentrations of these chlorinated compounds in water and bed sediment from major discharges to the Yakima and in the river main stem to identify sources and ascertain whether contamination is the result of historical use or recent application.

Field work was completed in October 1985. A description of these investigations, the results obtained, and a discussion of the significance of the findings follow.

METHODS

Survey Plan

Figure 1 shows the locations where fish, water, and sediment were collected.

The fish species selected for sampling were generally those widely distributed in the Yakima River and/or consumed by humans. The species collected were rainbow trout, mountain whitefish, largescale sucker, bridgelip sucker, northern squawfish, smallmouth bass, and channel catfish. Timing of the collections and species availability were largely dictated by life history and habitat requirements. Sampling was done between May and October; the main collection took place on August 19-20. Specimens of up to four fish species were collected from each of four reaches in the Yakima River; Cle Elum (river mile [r.m.] 179-181), Wymer (r.m. 134-136), Buena (r.m. 93-95), and Kiona/ Benton City (r.m. 20-23). Two invertebrates, mussels and crayfish, also were collected at Cle Elum and Wymer, but were not available in the lower river. The Cle Elum reach was intended to reflect background conditions. The other three sites were below each of the three major irrigated valleys and population centers in the basin. Returning spring chinook were obtained from the adult trap at Rosa diversion dam (r.m. 128); downstream migrating chinook and steelhead juveniles were taken from the smolt trap at Prosser on Chandler Canal (r.m. 46). Table 2 summarizes the samples obtained.

Table 2. Fish and invertebrate samples collected for Ecology's 1985 study of DDT compounds and other contaminants in the Yakima River.

Location	Approx. River Mile	Common Name	Scientific Name*	Collection Date	Tissues Analyzed .
Yakima R. 8 Cle Elum	179-181	Mountain whitefish, " Largescale sucker Rainbow trout Crayfish Mussel	Prosopium williamsoni " Catostomus macrocheilus Salmo gairdneri Pascifastacus sp. Margaritifera falcata	May 6 August 19 October 11 May 20 September 17, 18 September 12	muscle eggs muscle entire soft parts
Yakima R. @ Wymer	134-136	Mountain whitefish " Largescale sucker Bridgelip sucker Northern squawfish Rainbow trout Crayfish Mussel	Prosopium williamsoni Catostomus macrocheilus Catostomus columbianus Ptychochelius oregonensis Salmo gairdneri Pascifastacus sp. Margaritifera falcata	August 19 October 11 May 20 August 19 " September 23, 24 September 12	muscle, whole fish eggs "muscle, whole fish ", " entire soft parts
Yakima R @ Rosa Dam adult trap	128	Spring chinook	Oncorhynchus tshawytscha	May 21 July 23	muscle , eggs
Yakima R. @ Buena	93-95	Mountain whitefish " Largescale sucker Bridgelip sucker " Northern squawfish	Prosopium williamsoni Catostomus macrocheilus Catostomus columbianus Ptychochelius oregonensis	August 20 October 11 May 20 May 6 August 20	muscle, whole fish eggs whole fish muscle. whole fish
Prosser, Chandler Canal smolt trap	46	Spring chinook Steelhead	Oncorhynuchus tshawytscha Salmo gairdneri	May 21	whole fish
Yakima R. @ Kiona	20-23	Largescale sucker Northern squawfish Smallmouth bass Channel catfish	Catostomus macrocheilus Ptychochelius oregonensis Micropterus dolomieu Ictalurus punctatus	May 21 August 20 May 20	eggs muscle, whole fish

^{*}American Fisheries Society (1980).

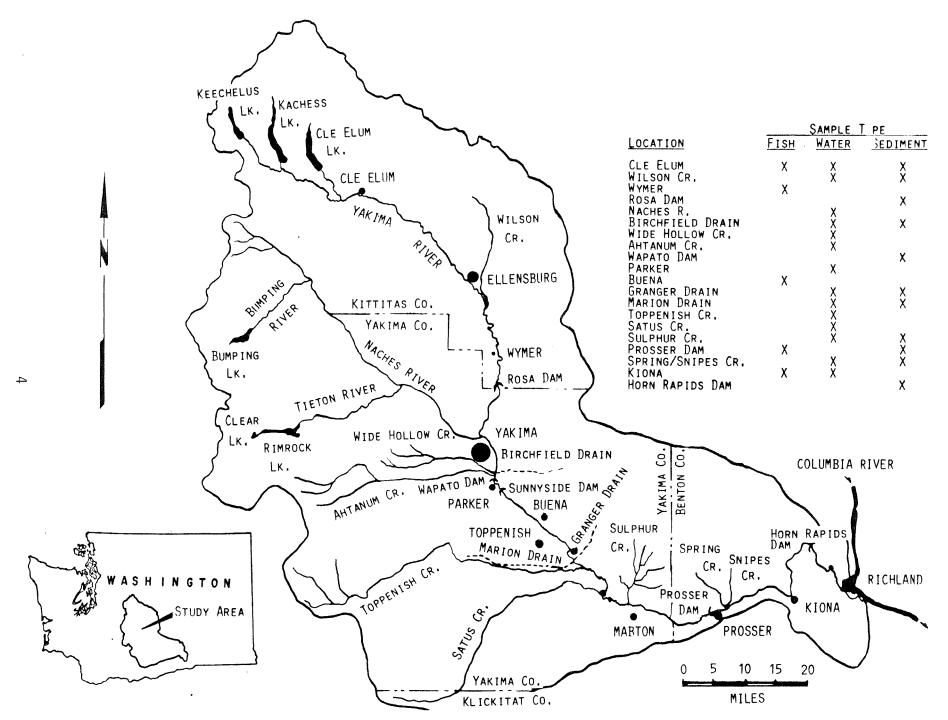


Figure 1. Locations of fish, water, and sediment samples collected for Ecology's 1985 study of DDT and other contaminants in the Yakima River basin.

Chemical analyses were done on whole fish, muscle, and eggs. Whole fish data were needed to assess potential impacts to aquatic life and wildlife by comparing these data with National Academy of Sciences recommendations (NAS, 1973). Whole fish data also were compared with existing data from Ecology's BMP surveys and the U.S. Fish and Wildlife Service (USFWS) National Pesticide Monitoring Program which are both primarily on whole fish. Data on muscle tissue were required to evaluate the hazard to human health from eating Yakima River fish or invertebrates. Data on contaminants in fish eggs were obtained to assess the potential for adverse reproductive effects which were considered most likely to occur during the sensitive life history stages of embryonic development and yolk sac absorption.

Eleven tributaries to the Yakima and three sites on the river main stem were selected for water sampling--these are described in Table 3. The selection criterion for tributaries was large discharge coupled with substantial irrigation return flow contribution as determined using information supplied by the U.S. Bureau of Reclamation Yakima Office, and other reports (U.S. Army Corps of Engineers, 1978; U.S. Department of Agriculture, 1978; U.S. Bureau of Reclamation, 1975). Municipal and industrial discharges in the Yakima basin are unlikely sources of the principal contaminant of concern, DDT (Johnson and Newman, 1983), and, therefore, were not sampled. Mainstem stations were chosen to reflect background water quality (Cle Elum station at r.m. 183.1) and conditions in the middle and lower river (Parker and Kiona stations at r.m. 104.6 and 29.8, respectively). Water samples were collected June 24 early in the irrigation season, August 5 during peak irrigation demand, and October 29 after diversions for irrigation were turned off.

Table 3. Locations of water samples collected for Ecology's 1985 study of DDT compounds and other contaminants in the Yakima River.

Station Name	Sampling Location	River Mile [†]
Yakima River at Cle Elum	Cle Elum bridge	183.1
Wilson Creek	Highway 821 bridge	147.0(L)
Naches River	Highway 82 bridge	116.3(R)
Birchfield Drain	Mouth at Thorp Road	107.6(L)
Wide Hollow Creek	At Highway 82 culvert	107.4(R)
Ahtanum Creek	USBR gage	106.9 (R)
Yakima River at Parker	Parker bridge	104.6
Granger Drain	Mouth at Granger STP	82.8(L)
Marion Drain	Indian Church Road bridge	82.6(R)
Toppenish Creek	Highway 22 bridge	80.4(R)
Satus Creek	North Satus Road bridge	69.6(R)
Sulphur Creek	McGee Road bridge	61.0(L)
Spring/Snipes Creek*	Below Chandler Canal	41.8(L)
Yakima River at Kiona	Kiona-Benton City highway bridge	29.8

[†]L and R refer to left and right bank facing downstream.

^{*}Spring and Snipes Creeks merge before flowing into the Yakima River.

Collection of bed sediment samples was done September 24 and 25 at or immediately downstream of the mouths of six of the above tributaries and behind each of four diversion dams on the Yakima (see Table 4). The tributaries sampled were those where DDT compounds or other organochlorine pesticides had been detected in the June or August water collections.

Table 4. Locations of bed sediment samples collected for Ecology's 1985 study of DDT compounds and other contaminants in the Yakima River.

Station Name	Sampling Location *	Method	River Milet
Yakima River at Cle Elum Wilson Creek Yakima River at Rosa Dam Birchfield Drain Yakima River at Wapato Dam Granger Drain Marion Drain Sulphur Creek	Mid-channel 200 ft. below Cle Elum bridge Near left bank at upstream side of Burlington Northern RR br. Mid-channel 500 ft. above dam Mouth Near left bank 200 ft. above dam Near river bank 5 - 10 feet downstream of mouth Near river bank 5 - 10 feet downstream of mouth Mouth	hand collected pipe dredge ponar grab pipe dredge	183.1 147.0(L) 127.9 103.8 ^{6(L)} 82.8(L) 82.6(R) 61.0(L)
Yakima River at Prosser Dam Spring/Snipes Creek* Yakima River at Horn Rapids Dam	Near left bank 3,000 feet upstream of dam Near river bank 5 – 10 feet downstream of mouth	ponar grab pipe dredge ponar grab	47.1 41.8(L) 18.0

[†]L and R refer to left and right bank facing downstream.

Ecology had historical data on one tributary, Birchfield Drain, which indicated it was a source of DDT compounds (Johnson and Newman, 1983). Efforts were made to locate sources within this drainage by collecting a series of bottom sediment samples on March 25, prior to the start of the irrigation season, and a series of water samples on July 2 and August 21.

Chemicals Analyzed

Table 5 shows the chemicals analyzed in fish, water, and sediment. Both p,p' and o,p' isomers of DDT, DDE, and DDD were quantified in all samples. Samples also were routinely analyzed for fifteen additional persistent organochlorine pesticides, four polychlorinated biphenyl (PCB) mixtures, and mercury. PCBs and mercury were included among the target chemicals because concentrations appeared somewhat elevated in Yakima River fish samples collected during Ecology's 1984 BBMP survey (Hopkins, et al., 1985).

Selected water samples were screened for a wide variety of chemicals including several classes of pesticides, phenols, aromatic hydrocarbons, halogenated hydrocarbons, and thirteen metals.

A number of ancillary parameters were measured to help interpret the organics and metals data. Fish tissue samples were analyzed for percent dry weight and percent lipid. Flow, temperature, pH, specific conductivity, total suspended solids, and total organic carbon were determined at each water sampling site. Percent dry weight, total organic carbon, and grain size distribution were analyzed in the sediment samples.

^{*}Spring and Snipes Creeks merge before flowing into the Yakima River.

Table 5. Target chemicals and corresponding detection limits for Ecology's 1985 study of DDT compounds and other contaminants in the Yakima River (parts per billion).

A. ALL SAMPLES

Organochlorine Pesticides

	fish	water	sediment
p,p' DDT p,p' DDE	40 ug/Kg, wet 20 "	0.02 - 0.01 ug/L 0.01 - 0.005 "	0.1 ug/Kg, dry
p,p' DDD o,p' DDT	40 " 40 "	0.02 - 0.01 " 0.02 - 0.01 "	H
o,p' DDT o,p' DDE	20 "	0.02 - 0.01	II
o,p' DDD	40 "	0.02 - 0.01 "	H
dieldrin	20 "	0.01 - 0.005 "	11
endosulfan 1/11	20 "	0.01 - 0.005 "	H H
alpha-BHC gamma-BHC	100 " 100 "	0.005 - 0.003 " 0.005 - 0.003 "	11
beta-BHC	100 "	0.005 - 0.003 "	11
de l t a-BHC	100 "	0.005 - 0.003 "	II
heptachlor	10 "	0.005 - 0.003 "	II
aldrin	10 "	0.005 - 0.003 "	II II
heptachlor epoxide endrin	100 '' 20 ''	0.005 - 0.003 " 0.01 - 0.005 "	'' 'I
endrin aldehyde	200 "	0.01 - 0.005	It
endosulfan sulfate	200 "	0.02 - 0.01 "	II
methoxych lo r	100 "	0.1 -0.05 "	i (
chlordane	100 "	0.1 - 0.05 "	H .
t o x aphene	2000 "	1.0 - 0.5	4.6
			ative .

Polychlorinated Biphenyls

	fish	water	sediment
PCB-1242 PCB-1248 PCB-1254	100 ug/Kg, wet 100 " 100 "	0.05 ug/L 0.05 " 0.05 "	1.5 ug/Kg, dry
PCB-1260	100 "	0.05 "	EI

Metals

	fish	water	sediment
mercury	3 ug/Kg, wet	0.05 ug/L	3 ug/Kg, wet

Table 5 - continued.

B. SELECTED WATER SAMPLES[†]

Organophosphorous	Pesticides	Carbamate/Phen	ylurea Pesticides	Herbicides	
phosdrin	0.5 ug/L	ox amyl	5.0 ug/L	simazi ne	3 µg/L
t hi met	0.5	methomyl	5.0 "	cyanozine	2
diazinon	0.5 "	carbaryl	5.0 "	dicamba	0.1
di-syston	0.5 "	propham	5.0 "	2,4-D	0.1
dioxathion	0.5 "	chloropropham	5.0 "	silvex	0.1 "
dimethoate	0.5 "	methioc arb	5.0	2,4,5-T	0.1^{-11}
chlorpyrifos	0.5	baygon	5.0	MCPA	3 "
methyl parathion	0.5	furadan	5.0 "		
malathion	0.5	f l uometuron	5.0 "		
ethyl parathion	0.5	monuron	5.0		
DEF	0.5	di uron	5.0 "		
eth ion	0.5 "	linuron	5.0 "		
trithion	0.5	neburon	5.0 "		

Acid/Base-Neutral Compounds

•		Committee Commit	
aniline	1 ug/L	2,6-dinitrotoluene	1 ug/L
pheno l	1 "	acenaphthene	1 "
bis(2-chloroethyl)ether	1 "	2,4-dinitrophenol	1 "
2-chl orophenol	1 "	dibenzofuran	1 "
1,3-dichlorobenzene	1 "	4-n itrophenol	2 "
1,4-dichlorobenzene	1 "	2,4-dinitrotoluene	1 "
1,2-dichlorobenzene	1 "	fluorene	1 "
benzyl alcohol	1 "	diethyl phthal ate	1 "
2-methylphenol	1 "	n-nitrosodi phenyl ami ne	2 "
bis (2-chloroisopropyl) ether	1 "	4-chlorophenyl phenyl ether	1 "
hexachl oroethane	1 "	4,6-dinitro-2-methylphenol	1 "
4-methyl pheno l	1 "	4-bromophenyl phenyl ether	1 "
n-nitroso-di-n-propylamine	2 "	hex ach l'orobenzene	1 "
nitrobenzene	1 "	pentachl orophenol	1 "
isophorone	1 "	phenanthrene	1 "
2-nitrophenol	1 "	anthracene	1 "
2\$4-dimethylphenol	1 "	di-n-butylphthal ate	1 "
bis(-2-chloroethoxy)methane	1 "	fluoranthene	1 "
2,4-dichlorophenol	1 "	benzidi ne	1 "
1,2,4-trichlorobenzene	1 "	pyrene	1 "
benzoic acid	1 "	but ylbenzyl phthal ate	1 "
naphthal ene	1 "	benzo(a)anthracene	1 "
4-chioroani line	1 "	3,3'-dichlorobenzidine	1 "
hexachlorobutadiene	1 "	chrysene	1 "
4-chloro-3-methylphenol	1 "	bis (2-ethylhexyl) phthalate	1 "
2-methyl naphthal ene	1 "	di-n-octyl phthalate	1 "
hexachl orocyc lopentadiene	1 "	benzo(b)fluoranthene	1 "
2,4,6-trichlorophenol	1 "	ben zo(k) fluoranthene	1 "
2,4,5-trichlorophenol	1 "	ben zo(a) pyrene	1 "
2-chloronaphthalene	1 "	indeno(1,2,3-cd)pyrene	1 "
3-nitroaniline	3 "	dibenz(a,h)anthracene	1 "
acenaphthyl ene	1 "	benzo(g,h,i)perylene	1 "
dimethyl phthal ate	1 "		

Table 5 - continued.

Volatile Compounds

chloromethane	1	ug/L	1,2-dichloropropane		ug/L
bromometh ane			trans-1,3-dichloropropene	5	
vinyl chloride	10	11	trich loroethane	5	81
chloroethane	10	11	dibromochloromethane	5	11
methyl ene chloride	1	11	1,1,2-trichloroethane	5	11
acetone	10	11	benzene	5	11
carbon disulfide	5	4.6	cis-1,3-dichloropropane	5	11
1,1-dichloroethene	5	11	2-chloroethylvinylether	10	11
1,1-dichloroethane	5	I i	bromoform	5	11
trans-1,2-dichloroethene	5	H	2-hex anone	10	11
chloroform	5	11	4-methyl-2-pentanone	10	11
1,2-dichloroethane	5	11	tetrachloroethene	5	11
2 - butanone	5	11	toluene	5	H
1,1,1-trichloroethane	5	it	chlorobenzene	5	11
carbon tetrachloride	10	Iŧ	ethylbenzene	5	11
vinyl acetate	10	8 6	styrene	5	11
bromodich loromethane	5	II	total xylenes	5	11
1,1,2,2-tetrachloroethane	5	ii	<u> </u>		

Metals			Miscellaneous	
copper zinc nickel chromi um cadmi um lead beryl li um tin arsenic sel eni um silver antimony thalli um	1 1 1 0.1 1 0.1 1 1 0.1	ug/L II II II II II II II II II	cyanide 20	ug/L

[†]Organophosphorus, carbamate, and phenylurea pesticides analyzed in all June 24 water samples. Acid/base-neutrals, volatiles, metals, and cyanide analyses restricted to June 24 water samples from Wilson Creek, Marion Drain, Sulphur Creek, and Yakima River at Kiona. Herbicide analysis done on August 5 water samples from Wilson Creek, Birchfield Drain, Granger Drain, Sulphur Creek, and Yakima River at Kiona.

Sampling Procedures

Fish other than salmonids were collected by electroshocking. The Yakima Indian Nation supplied a boat equipped with a Smith-Root electroshocker and operated by a tribal biologist for each collection. Salmon and trout samples were provided by the Yakima Nation from traps as previously mentioned, or in the case of rainbow trout, caught on hook and line. Mussels and crayfish were hand-collected by Jim Chatters, Central Washington University.

Fish and invertebrate samples were wrapped in aluminum foil, put in plastic bags, and placed in coolers containing ice in polyethylene cubitainers. The foil had been previously rinsed with 10 percent HCl, deionized water, nanograde methylene chloride, and nanograde acetone.

Water samples were whole, unfiltered water taken as surface grabs from midchannel, either collected by hand or by strapping the sample bottle to an upstream-orienting sounding weight and lowering it on a polypropylene line. Temperature and pH were measured in the field with a precision mercury thermometer and Orion Ionalyzer pH meter. Flow was either gaged in the field using a Marsh-McBirney magnetic flow meter and top-setting rod, or obtained from records of the U.S. Bureau of Reclamation, Yakima office or Wapato Irrigation District.

Sediment samples were taken with a Ponar grab in slow-moving water or an Emery pipe dredge in fast water; both were constructed of stainless steel. Samples taken for analysis from the Ponar grab were of the top two on surface layer. Depth of penetration of the pipe dredge ranged up to approximately 4 cm, based on observations in shallow water. One grab was taken at each sampling site. An effort was made to sample fine rather than coarse material. Sediment samples were homogenized by stirring with stainless steel spoons in large stainless steel beakers. Sediment samplers, spoons, and beakers were washed in Liqui-Nox detergent, de-ionized water, 10 percent HCl, nanograde methylene chloride, and nanograde acetone between samples.

Table 6 shows the sample containers, cleaning procedures, and preservatives used for water and sediment samples. All samples were placed on ice immediately after collection.

Water samples for organochlorine pesticides, PCBs, and organophosphorus/carbamate/urea pesticides analyses were shipped by air freight to California Analytical Laboratories, Inc. (CAL) in West Sacramento, California, within 24 hours of collection. All other samples were returned to the Ecology/EPA Manchester, Washington, laboratory.

Fish and invertebrates were dissected at Manchester using stainless steel instruments and working on aluminum foil. Total lengths and weights of the specimens used for analysis were recorded. Whole fish, muscle, and egg samples were composite tissues from two or more individuals of the same species where sample size permitted. Muscle samples did not include skin. Each tissue sample was homogenized in a Waring blender or a Hobart grinder. Sample containers for tissue were glass jars with teflon-lined lids cleaned as described for other organochlorine samples in Table 6. Tissue samples were frozen at Manchester for later shipment to CAL.

Table 6. Sample handling information for water and sediment samples collected for Ecology's 1985 study of DDT compounds and other contaminants in the Yakima River.

Sample Type and Parameter	Container	Cleaning procedure?	Preservative*
Water			
Organochlorine Pesticides/PCBs	1 quart amber glass ^{††}	virgin bottles, rinsed with methylene	
Organophosphorus Pesticides Carbamate/Urea Pesticides	ни н ,	chloride and drained	
Herbicides	1 quart clear glass ^{††}	detergent wash, tap water rinse, nitric rinse, deionized water rinse, methylene chloride rinse, oven dry, QC checked	
Acid/Base-Neutrals	1 gallon clear glass ^{††}	detergent wash, tap water rinse, nitric rinse, deionized water rinse, methylene chloride rinse, oven dry, QC checked	
Volatiles	40 mL clear glass ^{††}	detergent wash, tap water rinse, deionized water rinse, oven dry, QC checked	
Mercury	250 mL borosilicate glass ^{††}	detergent wash, HNO_3 rinse, high temperature baking	HNO ₃ (5% V/V) + K ₂ Cr ₂ O ₇ (.05% V/V)
Other Metals	1 quart high density polyethylene	HCl/HNO ₃ /deionized water leaching	HNO ₃ to pH <2
Cyanide	250 mL polyethylene	virgin bottles, deionized water rinse	NaOH to pH >10
Total Organic Carbon	8 ounce clear glass ^{††}	detergent wash, tap water rinse, deionized water rinse, oven dry	H ₂ SO ₄ to pH <2
Specif. Conduct./Tot. Susp. Solids	500 mL polyethylene	detergent wash, deionized water rinse, air dry	N- 4
Sediment			
Organochlorine Pesticides/PCBs/Mercury % Dry Wt, % Total Organic Carbon	8 ounce clear glass ^{††}	detergent wash, tap water rinse, nitric rinse, deionized water rinse, methylene chloride rinse, oven dry, QC checked	

[†]See appendix I for details.
*All samples iced immediately after collection.
††Teflon-lined lids.

Analysis

Table 7 summarizes the analytical methods employed and shows where the analyses were done. The method used for organochlorine pesticides and PCBs in tissue was developed at the Columbia National Fisheries Research Laboratory (CNFRL), Columbia, Missouri, for use in the USFWS National Pesticide Monitoring Program. In summary, homogenized tissue is mixed with Na₂SO₄ and eluted with methylene chloride. The concentrated eluate is cleaned up by gel permeation chromatography, florisil column chromatography, and silicagel column chromatography. The resulting three fractions are analyzed by influent-split, dual-column gas chromatography-electron capture (GC-EC) according to EPA protocol (EPA method 608). Because this method is not described in standard analytical methods manuals, the analysis scheme is given in its entirety in Appendix II.

Quality Assurance[†]

Interlaboratory Comparison -- In order to ensure that CAL's organochlorine analysis of tissue was comparable to that performed at the Ecology/EPA Manchester laboratory where all of Ecology's previous Yakima River work had been done, two fish tissue homogenates were split between the laboratories prior to initiation of the main survey. One homogenate, muscle from mountain whitefish collected near Cle Elum, had a low level of contamination; the other, whole largescale sucker from Buena, had relatively high contamination. The results of this exercise are in Table 8.

Table 8. Results of May 1985 fish tissue interlaboratory comparison exercise between the Ecology/EPA Manchester laboratory and California Analytical Laboratories (ug/Kg, wet).

Tissue Type	Mount	ain Whit	efish =	muscle	Largescale Sucker - whole fish							
Laboratory	ECOLO	GY/EPA	C	AL .	ECOLO	GY/EPA	C	CAL				
Sampl e Number	7002	7003	7002	7003	7004	7005	7004	7005				
p,p'-DDE	25	23	20	20	560	570	660	640				
p,p'-DDT	8	5u	40u	40u	190	210	60	60				
p,p'-DDD	4	5u	40u	40u	110	120	520	200				
dieldrin	2u	5u	200u	200u	24	24	200u	200u				
% lipid	5.29			5.29	1.16	1.02	4.97	4.84				
% dry weight	27.4	27.4	27.1	26.3	25.8	26.0	24.9	24.4				

NOTE: PCBs and other organochlorine pesticides not detected. u = Not detected at detection limit shown.

[†]QA information for the broad spectrum scans done on water is available on request.

Table 7. Analytical methods employed for Ecology's 1985 study of DDT compounds and other contaminants in the Yakima River.

Parameter	Analysis	Method Number	Reference	Laboratory
		TISSUE		
organochlorine pesticides/PCBs	GC/EC	**	Appendix II	California Analytical, W. Sacramento, CA
percent lipid	granmetric		11	11
percent dry weight	II		II .	H
mercury	cold vapor	245.5-modified	EPA (1979a)	Ecology/EPA, Manchester, WA
		WATER		
organochlorine pesticides/PCBs	GC/EC	608-modified	EPA (1984)	California Analytical, W. Sacramento, CA
organophosphorus pesticides	GC/NPD	622	H	н
carbamate & phenylurea pesticides	HPLC	632	31	T.
herbicides	GCIEC	625-modified	EPA (1984)	Ficology/EPA, Manchester, VM
acid/base-neutrals	GC/MS	625	н	
volatiles	11	624	**	H .
mercury	cold vapor	245.2-modi fied	EPA (1979a)	п
other metals	AA	various	1	R
cyanide	ti trimetric	335.2	II .	H
total suspended solids	gravimetric	209C	APHA (1985)	u
specific conductivity	conductivity cell	205	•	II .
total organic carbon	combustion-infrared	505A	II	Laucks, Seattle, WA
		SEDIMENT		
organochlorine pesticides/PCBs	GC/EC	608	EPA (1984)	Ecology/EPA, Mancheser, WA
mercury	cold vapor	245.5-modi fied	EPA (1979a)	11
percent dry weight	gravimetric	209F	ĀPHA`(1985)	ii
total organic carbon	combustion-CO ₂		in-hoke	Laucks, Seattle, WA
grain size	sieve and pipette		Holme & McIntyre (1971)	Parametrix, Bellevue, WA

Concentrations reported by both laboratories for p,p'-DDE were in agreement. While the results for dieldrin did not disagree, CAL's higher detection limits precluded quantification. An order of magnitude lower detection limits were achieved for dieldrin in the main survey. Concentrations of p,p'-DDT reported by CAL were lower than Ecology/EPA. In the opinion of the CAL analyst, this was due to p,p'-DDT splitting into both the florisil and silica fractions--a problem remedied before further analyses were initiated (Soderquist, 1985). The CAL results for p,p'-DDD showed poor precision. P,p'-DDD was not detected in subsequent duplicates (see below) and rarely detected in the main fish survey (see results). Because p,p'-DDD is a minor constituent of the total of DDT compounds (t-DDT), potential poor precision of CAL's p,p'-DDD analysis in the main survey was not considered to significantly influence measurement of t-DDT in tissue. A final problem identified through the intercomparison exercise was the low percent lipid reported by Ecology/EPA for samples 7004 and 7005. This result was never satisfactorily explained.

Accuracy and Precision -- The accuracy of organochlorine analyses on tissue, water, and sediment was assessed by analysis of spiked samples. Spike recovery was not used to correct the data. Selected tissue and water samples were spiked at CAL with gamma-BHC, heptachlor, aldrin, dieldrin, endrin, p,p'-DDT, and PCB-1254. Two water samples from the June 24 collection were also spiked with ethylparathion and carbaryl. Spike concentrations were 0.20 - 0.50 ug/L and 100 - 250 ug/Kg. Percent spike recoveries (mean \pm standard deviation) in tissue were 102 \pm $18_{n=23}$ for organochlorine pesticides and 92 \pm $13_{n=3}$ for PCB-1254. Spike recoveries in water were 82 \pm $3_{n=54}$ for organochlorine pesticides, 82 \pm $10_{n=2}$ for PCB-1254, 85 \pm $17_{n=4}$ for ethylparathion, and 82 \pm $8_{n=4}$ for carbaryl.

Sediment samples were spiked at the Manchester laboratory with p,p' and o,p' isomers of DDE, DDT, and DDD. Percent recoveries were 94 \pm 25_{n=18}.

The precision of the organochlorine analyses was evaluated by analyzing duplicate samples, duplicate spikes, and field replicates. The results of analysis of duplicate tissue samples are in Table 9.

Seven organochlorine pesticides and PCB-1260 were detected in the tissue duplicates. Concentrations of p,p'-DDE, o,p'-DDE, p,p'-DDT, endosulfan sulfate, endrin aldehyde, PCB-1260, and lipid and dry-weight percentages were within 10 percent of the mean of duplicate measurements. Quantification of dieldrin and endosulfan was less precise, ranging up to 33 percent about the duplicate mean. (The mean values in Table 9, after rounding to significant figures, are used in the remainder of this report.)

Recoveries of duplicate spikes on water samples were within 20 percent of the duplicate mean for both organochlorine pesticides and PCB-1254. P,p' and o,p' isomers of DDE, DDT, and DDD recoveries in sediment were within 15 percent of the mean of triplicate spikes.

The accuracy and precision of the mercury analyses done at Manchester were determined by analyzing standard reference materials and spiked samples. The results for reference material analyses, shown in Table 10, indicate good precision but a tendency to underestimate the mercury content of tissue samples. Spiked recoveries (percent) also were low--52 \pm $16_{n=3}$.

Results of organochlorine analyses by California Analytical Laboratories on duplicate fish tissue samples from Ecology's Yakima River fish collections in 1985 (ug/Kg, wet). Table 9

Parameter	Sample N	umbers	Mean ± Range	Range as % of Mean
p,p'-DDE p,p'-DDT dieldrin endosulfan % lipid % solids	8121 190 50 400 20 6.14 31.0	8124 200 50 460 30 5.90 38.7	195 ± 5 50 ± 0 430 ± 30 25 ± 5 6.02 ± 0.12 34.5 ± 3.9	3 0 7 33 2 11
p,p'-DDE p,p'-DDT o,p'-DDT dieldrin endosulfan endosulfan sulfate endrin aldehyde % lipid % dry weight	8122 560 110 60 720 850 290 40 5.70 38.5	8123 510 110 60 460 590 280 40u 5.75 36.5	535 ± 25 110 ± 0 60 ± 0 590 ± 130 720 ± 130 285 ± 5 (at detection limit) 5.72 ± 0.03 37.5 ± 1.0	5 0 0 22 18 2 0.5
p,p'-DDE % lipid % dry weight	8078 190 1.8 22.9	8078D 200 1.7 N/A	195 ± 5 1.75 ± 0.05	3 3
p,p'-DDE o,p'-DDE PCB-1260 % lipid % dry weight	910 20 130 3.8 25.4	20u 110 3.7 N/A	830 ± 80 (at detection limit) 120 ± 10 3.75 ± 0.05	10 8 1

u = Not detected at detection limit shown. N/A = Not analyzed.

Table 10. Results of mercury analyses of reference materials by the Ecology/EPA Manchester laboratory in 1985.

Reference Material	Manchester Results	Reference Value					
"Mercury in Water" NBS Standard Reference Material 1642b	1.60 \pm 0.05 ug/L (mean \pm range; n = 2)	1.49 f 0.06 ug/L					
"Trace Metals in Fish" EPA/EMSL	1.60 \pm 0.06 mg/Kg (mean \pm s.d.; n = 4)	2.52 ± 1.28 mg/Kg					
"Albacore Tuna" NBS Research Material 5	0.63 \pm 0.06 mg/Kg (mean \pm s.d.; n = 4)	0.95 ± 0.1 mg/Kg					

Organochlorine and mercury analyses on replicate field samples showed that, in general, variability was low. These data are presented later in the report.

Blanks -- All organochlorine and mercury analyses included methods blanks as a check against contamination arising from sample handling and analysis. Neither organochlorines nor mercury were detected. Field blanks were analyzed for all water sample collections. Both transport blanks (sample bottles filled with organic-free water and carried unopened through the survey) and transfer blanks (organic-free water transferred to empty sample bottles while in the field) were included. All field blanks were below detection limits.

Q/A Review -- The organochlorine data produced by CAL were reviewed by Mike Schlender of the Ecology/EPA Manchester laboratory or through contract with Ecology & the Environment, Inc., Seattle.

Limitations of the Data

Although the surveys reported here involved hundreds of physical/chemical measurements and represent the most extensive investigation of its type yet undertaken by Ecology, the reader should be aware of the limitations of the data obtained. These include the following:

- o Sample Size and Frequency -- Organics analyses are costly and time-consuming. As a result, relatively few fish, water, or sediment samples could be analyzed for any one period in time or at any single station. The results obtained, therefore, give no information on short-term variability or local variation in concentration. Short-term variability is less of a concern with fish and sediment which retain organochlorines over extended periods of time. Small sample size also does not lend itself to determination of the statistical significance of differences in concentration between locations, times, or species sampled. Good agreement between replicate samples and the fact that consistent patterns in contamination emerged from the data offset the above short-comings somewhat.
- o Biological Variability -- Contaminant levels in fish and aquatic invertebrates have been shown to vary with species, tissue type, lipid content, time of year, age, length, feeding, sex, spawning activity, and water

- quality (temperature, suspended solids, hardness, pH). In this study, only the influence of species, lipid, and tissue differences was evaluated. Specimens selected for analysis represented the average size encountered in the field. To the extent possible, a similar size range of individuals of each species was analyzed from each location sampled.
- o Number of Tributaries Monitored -- There are fifty or more irrigation returns, creeks, and rivers tributary to the Yakima. This study monitored eleven of the largest considered to have the greatest potential for organochlorine contamination. It is likely that other discharges have detectable concentrations of organochlorine pesticides. The extent to which the present study accounted for the major organochlorine pesticide loads to the Yakima River is discussed later in the report.
- on fish produced by three different laboratories—Ecology/EPA, CAL, and CNFRL. Sample handling, extraction efficiencies, analytical schemes, instrumentation, and other factors vary between laboratories and often within the same laboratory from year to year. For this reason, it is not unusual for the amount of analyte found by different laboratories when analyzing the same material to vary by 50 percent or more. The inter-laboratory comparison exercise (see Quality Assurance) and consequent adoption of the CNFRL method for tissue analysis were done in an effort to maximize comparability of data sets.
- o Analysis for Pesticides Other Than Organochlorines -- Organophosphorus and chlorophenoxy/triazine herbicides analyses were conducted using gas chromatography; carbamatelphenylurea pesticides analyses were done by high-pressure liquid chromatography. Extraction occurred within seven days of sample collection; analysis was completed within 30 days. the ease with which they are hydrolized, oxidized, or otherwise metabolized, organophosphoruslcarbamate pesticides and chlorophenoxy herbicides are considered non-persistent in soil and water; phenylureas and triazines are considered moderately persistent (Menzer, 1980). Losses of these compounds are likely unless analysis promptly follows sample collection. There is additional opportunity for loss in the analysis itself during extraction (extraction efficiencies are not well known for many compounds) or as a result of heating during gas chromatography. These factors and the generally high detection limits of the method (see Table 5) should be kept in mind when interpreting the results of the screen for these compounds in water.

RESULTS[†]

Fish Tissue

Table 11 shows the results of analyses for organochlorine pesticides, PCBs, and mercury in Yakima River whole fish and mussels. The organochlorine compounds detected were p,p'-DDE, o,p'-DDE, p,p'-DDT, p,p'-DDD, dieldrin, endosulfan, endrin, and PCB-1260. Mercury was above detection limits in all samples. 0,p' isomers of DDT and DDD and other target organochlorine pesticides were not detected.

Concentrations of total DDT compounds $(t-DDT^{\dagger\dagger})$ ranged from below detection limits (20 ug/Kg for p,p'-DDE) to 3,000 ug/Kg. The major component of t-DDT was consistently p,p'-DDE. DDT compounds were detected in all whole-fish samples; they were not detected in mussels, probably due to their low lipid concentrations. DDT compounds, and organochlorines in general, are much more soluble in lipid than in water. As a result, aquatic organisms accumulate these compounds as a function of their lipid content.

For those species available at multiple locations in the river--mountain whitefish, suckers (mixed species), and northern squawfish--concentrations of t-DDT increased by a factor of two to six at each successive downstream location sampled. For example, concentrations in whole suckers were 80 ug/Kg at Wymer (r.m. 134-136), 390 ug/Kg (replicate mean) at Buena (r.m. 93-95), and 1,100 ug/Kg at Kiona (r.m. 20-23).

Downstream migrating spring chinook and steelhead smolts intercepted at Prosser in the Chandler Canal trap (r.m. 46) had t-DDT concentrations of 570 ug/Kg and 100 ug/Kg, respectively. These concentrations fall in the middle to lower range of those observed in resident adult fish of other species.

Dieldrin, endosulfan, and endrin were detected in whole fish less frequently and at lower concentrations than DDT compounds. Detection occurred primarily in fish from the lower river. Dieldrin was detected in one species each at Wymer and Buena (confirmed in replicate samples), and in both species analyzed from Kiona. The maximum dieldrin concentration observed was 240 ug/Kg in largescale suckers at Kiona. Endosulfan and endrin were detected only at Buena. Maximum concentrations were 170 ug/Kg endosulfan in whitefish and 50 ug/Kg endrin in suckers. Concentrations of endosulfan and endrin measured in replicate samples of these species agreed closely.

Comparatively low levels of PCBs, identified as PCB-1260, were measured in Wymer squawfish and in most species analyzed below Wymer. Concentrations were between 110 and 300 ug/Kg.

[†]All organics and metals concentrations are expressed in parts per billion; i.e., ug/Kg for tissue and sediment and ug/L for water. Tissue concentrations are per unit wet weight; sediment concentrations are per unit dry weight.

 $^{^{\}dagger\dagger}$ t-DDT is the sum of p,p' and o,p' isomers of DDE, DDT, and DDD.

Table 11. Concentrations of organochlorine pesticides, PCBs, and mercury measured in samples of whole fish and mussels (less shell) collected by Ecology from the Yakima River in 1985 (ug/Kg, wet; detected compounds underlined).

Location and Species	Coll . Date	Sam- ple No.	N≈	Length Range† (mm)	Weight Range (grams)	% Dry Wt.	% Lipid	p,p'-	o,p'-	p,p'-	o,p'-	p,p'-	o,p'-	t- DDT	Diel- drin	Endo- sul- fan	En- drin	PCB- 1260	Mer- cury
Cle Elum (r.m. 179-181) Mountain whitefish Mussels	08/19 09/12	8069 8142	3 10	290-317 105-122	252 -3 56 57-99	31.7 13.0	11.5 0.76	50 20u	20u 20u	40u 40u	40u 40u	40u 40u	40u 40u	<u>50</u>	20u 20u	20u 20u	20u 20u	100u 100u	20 34
Wymer (r.m. 134-136) Mountain whitefish Bridgelip sucker Northern squawfish Mussels	08/19 !! 09/12	8065 8067 8063 8143	3 2 2 6	322-338 330; 391 364; 371 87-145	403-508 441; 501 404; 475 47-203	34.2 29.7 20.1 12.3	12.0 6.6 3.7 0.15	280 '86 560 20u	20u 20u 20u 20u	50 40u 40u 40u	40u 40u 40u 40u	40u 40u 40u 40u	40u 40u 40u 40u	330 80 560	90 20u 20u 20u 20u	20u 20u 20u 20u 20u	20u 20u 20u 20u 20u	100u 100u 270 100u	22 21 333 38
Buena (r.m. 93-95) Mountain whitefish " Suckers (mixed spp.*) , Northern squawfish	08/20	8072 8073 8074 8075 8079	3 2 2 3	240-286 232-295 343; 420 341; 359 285-331	130-253 110-252 480: 901 479; 559 230-297	34.6 34.9 28.2 28.6 25.4	11.7 12.8 8.4 10.0 3.7	550 540 370 210 890	20u 20u 20u 20u 20u 20u	80 110 20 80 40u	40u 40u 40u 40u 40u 40u	40u 40u 40u 40u 40u	40u 40u 40u 40u 40u 40u	630 650 490 290 890	20u 20u 30 40 20u	170 140 50 20u 20u	20u 20u 50 20 20	200 200 100 100 120	45 43 56 30 215
Kiona (r.m. 20-23) Largescale sucker Northern squawfish	08/20	8080 8082	2 2	462; 466 300; 336	935; 1150 241; 377	30.0 26.3	11.1 5.5	830 2900	30 20u	90 40u	40u 40u	140 130	40u 40u	1100 3000	240 90	20u 20u	20u 20u	110 300	50 96
Prosser (r.m. 46) Spring chinook (smolts) Steel head (smolts)	05/21	8103 8104	14 6	86-142 177-280	5.4-25.7 43.2-94.1	20.5	2.4 2.1	520 100	20u 20u	50 40u	40u 40u	40u 40u	40u 40u	570 100	20u 20u	20u 20u	20u 20u	100u 100u	24 32

NOIE See Table 5 Part A for detection limits of organochlorine pesticides analyzed for but not detected. u = Not detected at detection limit shown.

[†]Total length. *Pooled bridgelip and largescale suckers.

Mercury concentrations in whole fish and mussels ranged from 20 - 333 ug/Kg. Concentrations generally increased in a downstream direction except for squawfish. Squawfish consistently had much higher mercury concentrations than other species analyzed, Chinook and steelhead smolts had low mercury concentrations of 24 and 32 ug/Kg, respectively.

The data on muscle tissue are in Table 12. The organochlorine compounds detected were the same as those in whole fish except that one additional DDT isomer (o,p') was quantified and endrin was not detected. As was the case for whole fish, DDT compounds were detected in the muscle of most species analyzed. Only rainbow trout at Cle Elum and crayfish at both upper river locations sampled had no detectable DDT compounds in muscle.

Concentrations of DDT compounds were generally lower in muscle than in whole fish. This is probably a reflection of the relative amounts of lipid in these two types of tissue. The highest concentrations of t-DDT in muscle were 1,400 ug/Kg in whitefish at Buena (only 290 ug/Kg in the replicate sample) and 1,500 ug/Kg in channel catfish at Kiona. Re-analysis of the whitefish samples verified a wide variation in concentration. P,p'-DDE was, again, the predominant DDT compound present. The downstream trend in increasing t-DDT concentration in muscle was not as marked as in whole fish.

Dieldrin and endosulfan were infrequently detected in muscle. All detections in resident species were in the lower river. Dieldrin was also detected in muscle from one returning spring chinook. Dieldrin concentrations were 20 - 120 ug/Kg; concentrations above the 20 ug/Kg detection limit only occurred in resident fish from Kiona. As with whole fish, the detection of endosulfan in muscle was limited to samples from Buena; concentrations were in the 20 - 30 ug/Kg range.

PCB-1260 was detected at low concentrations in only two fish muscle samples; one of two whitefish samples at Buena had 230 ug/Kg, the channel catfish sample at Kiona had 180 ug/Kg.

Muscle tissue had more mercury than whole fish, Concentrations ranged from 32 - 447 ug/Kg with the highest concentrations again occurring in squawfish.

Muscle from returning spring chinook had lower contaminant burdens than resident species. Concentrations of the two organochlorine pesticides detected, p,p'-DDE and dieldrin, were near or below detection limits (20 ug/Kg) except for p,p'-DDE in one sample at 390 ug/Kg. PCBs were not detected in chinook muscle. Mercury concentrations were 48 - 62 ug/Kg, also low. Because chinook are migratory, it is not possible to attribute the presence of p,p'-DDE or dieldrin in returning adults to the Yakima system.

Egg samples were analyzed from whitefish, largescale suckers, and spring chinook (Table 13). The contaminants in eggs were generally the same as those in whole fish and muscle. Whitefish eggs had higher organochlorine levels than either suckers or chinook. Two compounds detected only in eggs were endosulfan sulfate and endrin aldehyde, metabolites of their respective parent compound. These compounds were identified in eggs from whitefish at Buena.

Table 12. Concentrations of organochlorine pesticides, PCBs, and mercury measured in samples of fish and crayfish muscle collected by Ecology from the Yakima River in 1985 (ug/Kg, wet; detected compounds underlined).

Location and Species	Coll. Date	Sam- ple No.	N=	Length Range† (mm)	Weight Range (grams)	% Dry Wt.	% Lipid	p,p'-	o,p'-	p,p'	- o,p'- DDT	p,p'-	o,p'-	t- DDT	Diel- drin	Endo- sul- fan	En- drin	PCB- 1260	Mer- cury
Cle Elum (r.m. 179-18	31)																		
Mountain whitefish Rainbow trout Crayfish	08/19 09/17-18 09/12	8068 8125 8127	3 3 10	335-356 214-217 23-41	387-500 105-110 2.7-29.0	31.7 23.9 17.5	7.8 0.94 0.59	90 20 u 20u	20u 20u 20u	40u 40u 40u	40u 40u 40u	40u 40u 40u	40u 40u 40u	90	20u 20u 20u	20u 20u 20u	20u 20u 20u	100u 100u 100u	42 33 N/A
Wymer (r.m. 134-136) Mountainwhitefish Bridgelip sucker Northern squawfish Rainbow trout Crayfish	08/19 " 09/23-24 09/12	8064 8066 8062 8126 8128	3 2 2 3 16	335-346 368; 380 356; 432 220-319 25-32	396-510 587; 763 400; 729 109-380 4.6-10.5	27.2 22.6 22.6 27.0 19.9	9.0 3.3 1.3 3.3 1.0	150 60 190 30 20u	20u 20u 20u 20u 20u 20u	40u 40u 40u 40u 40u	40u 40u 40u 40u 40u	40u 40u 40u 40u 40u	40u 40u 40u 40u 40u	150 190 30	20u 20u 20u 20u 20u 20u	20u 20u 20u 20u 20u 20u	20u 20u 20u 20u 20u 20u	IOOu 100u 100u 100u 100u	32 38 447 25 N/A
Buena (r.m. 93-95) Mountain whitefish " Suckers (mixed spp.*) Northern squawfish	08/20 11 11	8879 8877 8078	2 2 2 2 2	307; 331 305; 318 395; 408 447; 465	293; 316 266; 312 593; 727 874; 907	31.4 29.2 25.8 28.8 22.9	8.1 5.7 4.1 3.7 1.8	1100 290 130 130 200	284 284 20u	200 40 40 40u 40u 40u	40u 40u 40u 40u	70 u 40u 40u 40u	40u 40u 40u	1400 330 170 130 200	20 20 20 20 20 20 20 20 20	30 30 20 20u 20u 20u	20u 20u 20u 20u 20u	230 100u 100u 100u 100u	73 83 96 87 388
Kiona (r.m. 20-23) Largescalesucker Northern Squawfish Smallmouth bass Channel catfish	08/20 " " 05/20	8081 8083 8084 8085	3 3 4 4	423-440 286-305 226-276 352-395	764-1080 217-252 145-285	23.2 23.6 21.8 27.8	4.9 1.4 0.49 8.3	80 250 40 1300	20u 20u 20u 30	50 40u 40u 130	40u 40u 40u 40u	40u 40u 40u 80	40u 40u 40u 40u	130 250 '40- 1500	80 30 20u 120	20u 20u 20u 20u 20u	20u 20u 20u 20u 20u	100u 100u 100u 180	127 136 77 95
Rosa Dam (r.m. 128) Spring chinook	05/21 07/23	8105 8106 8099 8101	2 1 1 1	7750; 8000 7370 7550 7540	 	32.9 33.2 25.1 27.4	13.2 10.7 3.6 6.8	390 50 20 30	20u 20u 20u 20u 20u	40u 40u 40u 40u 40u	40u 40u 40u 40u 40u	40u 40u 40u 40u 40u	40u 40u 40u 40u 40u	390 50 20 30	20u 20 20u 20u 20u	20u 20u 20u 20u 20u	20u 20u 20u 20u 20u	100u 100u 100u 100u	48 53 72 62

NOTE: See Table 5 Part A for detection limits of other organochlorine pesticides analyzed for but not detected. u = Not detected at detection limit shown. N/A = Not analyzed.

[†]Total length except carapace length for crayfish. *Pooled bridgelip and largescale suckers.

Table 13. Concentrations of organochlorine pesticides, PCBs, and mercury measured in fish egg samples collected by Ecology from the Yakima River in 1985 (ug/Kg, wet; detected compounds underlined).

Location and Species	Coll. Date	Sam- ple No.	N=	Lensth Range [†] (mm)	Weight Range (grams)	% Dry Wt.	% Lipid	p,p'-	o,p'- DDE	p,p'-	o,p'-	p,p'-	o,p'- DDD	t- COT	Diel- drin	Endo- sul- fan	En- drin	PCB- 1260	Mer- cury
Cle Elum (r.m. 179-18	31)																		
Mountainwhitefish Largescale sucker	10/11 05/20	8120 8107	3 2	335-505 459; 462	475-1720 1188; 1296	38.4 37.1	5.9 6.3	30 14	20u 20u	40u 40u	40u 40u	40u 40u	40u 40u	30	200 20u	20u 20u	20u 20u	IOOu IOOu	14 5
Wymer (r.m. 134-136) Mountainwhitefish Largescale sucker	10/11 05/20	8121/24 8108	3 2	354-360 495; 502	537-582 1378; 1554	34.8 36.0	6.0 6.0	200 230	20u 20u	50 40	40u 40u	40u 40u	40u 40u	250 270	430 20u	20 20u	20u 20u	100u IOOu	11 9
Buena (r.m. 93-95) Mountainwhitefish Largescale Sucker	10/11 05/20	8122123 8109 8110	6 1 1	272-334 516 524	223-408 1325 1434	37.5 38.2 35.9	5.7 5.2 4.8	540 760 150	20u 20u 43	110 160 290	60 40u 150	40u 60 230	40u 40u 40u	710 980 820	590 20u 63	720* 20u 20u	20tt 20u 20u	IOOu 100u 460	15 37 22
Kiona (r.m. 20-23) Largescale sucker	05/20	8111	1	445	1131	35.8	5.8	490	20u	60	40u	40u	40u	540	30	20u	20u	100u	11
Rosa Dam (r.m. 128) Spring chinook	7/23	8100 8102	1	7550 7540		45.0 47.5	15.4 18.2	20u 40	20u 20u	40u 40u	40u 40u	40u 40u	40u 40u	<u></u>	230 180	20u 20u	20u 20u	100u 100u	11 11

NOTE: See Table 5 Part A for detection limits of other organochlorine pesticides analyzed for but not detected. u = Not detected at detection limit shown.

[†]Total length. *Endosulfan sulfate also detected at 280 ug/Kg, wet. ††Endrin aldehyde detected at 40 ug/Kg, wet, in #8122 only.

Although dieldrin was a minor contaminant in whole fish and muscle, its detection frequency and concentration level in eggs were comparable to those for DDT compounds. DDT compounds were detected in all resident species egg samples at concentrations from 14 - 980 ug/Kg. Dieldrin's detection frequency in eggs was five of eight samples, with concentrations reaching 590 ug/Kg.

Only one egg sample had detectable PCBs. One of the two samples of largescale sucker eggs collected at Buena had a PCB-1260 concentration of 460 ug/Kg, the highest observed during the present study.

Chinook eggs had measurable concentrations of p,p'-DDE and dieldrin only. 40 ug/Kg p,p'-DDE was detected in one of the two samples analyzed; both samples had detectable dieldrin at 230 ug/Kg and 180 ug/Kg. As mentioned above, these contaminants may have been accumulated elsewhere than in the Yakima River.

Mercury concentrations were much lower in eggs than in whole fish or muscle. A range of 11 - 37 ug/Kg was observed.

Table 14 summarizes the data on whole fish, muscle, and eggs.

Water

The results of organochlorine pesticides, PCBs, and mercury analyses of water samples collected from the Yakima River and eleven tributaries on June 24, August 5, and October 2, and results from one tributary, Birchfield Drain, where additional samples were collected on July 2 and August 21 are shown in Table 15.

The organochlorines detected in water were the same compounds that predominated in fish; i.e., p,p'-DDE, p,p'-DDT, p,p'-DDD, dieldrin, and endosulfan. PCBs and mercury were below detection limits (0.05 ug/L) in all water samples analyzed. There appeared to be a general trend toward decreasing concentration and frequency of detection for these compounds over the course of the study. No compounds were detected during the final survey in October.

Four of the eleven tributaries monitored had detectable concentrations of DDT compounds during the surveys conducted in June through August. DDT compounds were consistently detected in Birchfield Drain, Granger Drain, and Sulphur Creek, and once in Spring/Snipes Creek. Concentrations of t-DDT ranged from 0.02 to 0.07 ug/L. In contrast to the previously described results for fish tissue, p,p'-DDT concentrations were equal to or greater than p,p'-DDE. P,p'-DDD was detected in one instance only. Results from analysis of replicate samples collected from Granger Drain and Sulphur Creek in August were in close agreement. All four of the above tributaries shared the common characteristic of high suspended solids concentrations (61 - 380 mg/L).

Dieldrin was detected in tributaries more frequently, although at lower concentrations, than DDT compounds. Granger Drain, Sulphur Creek, and Spring/Snipes Creek had measurable dieldrin during both the June and August surveys; it was detected in Wilson, Toppenish, and Satus Creeks in the June survey only. The range in dieldrin concentration in these six tributaries was 0.003 - 0.02 ug/L. Concentrations in Wilson Creek, Granger Drain, and Sulphur Creek replicate samples were, again, similar.

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Table 14. Summary of organochlorine pesticides, PCBs, and mercury analyses of fish and invertebrates collected by Ecology from the Yakima River in 1985 (ug/Kg, wet).

				t-DD	t-DDT		Dieldrin		E	Endosulfan		tndrin			PCB-1260				Mercur	у
Location	Species	Tissue	Det 1 Freq.	Mean*	Range	Det∎ Freq.	Mean	Range	Det. Freq.	Mean	Range	Det. Freq.	Mean	Range	Det. Freq.	Mean	Range	Det. Freq.	Mean	Range
Cle Elum	Resident	Whole	1/2	30	20u-50	0/2			0/2			0/2			012			2/2	30	20-34
(r.m. 179-181) Wyer	spp.	11	3/4	250	20u-560	1/4	30	20u-90	0/4			0/4			1/4	100	100u-270	4/4	100	21-333
(r.m. 134-136) Buena	н	и	5/5	590	290-890	2/5	20	20u-40	3/5	80	20u-170	2/5	20	20u-50	3/5	120	100u-200	5/5	78	30-215
(r.m. 93-95) Kiona (r.m. 20-23)	u	u	2/2	2000	1100-3000	2/2	160	90-240	0/2	••		0/2		~~	2/2	200	110-300	2/2	70	50-96
Cle Elum	Resident	Muscle	1/3	40	20u-90	0/3			0/3			0/3			0/3			2/2	38	33-42
(r.m. 179-181) Wyer	spp.	ii	4/5	90	20u-190	0/5			0/5			0/5			0/5			4/4	136	25-447
(r.m. 134-136) Buena	H	14	5/5	450	130-1400	1/5	10	20u-20	3/5	20	20u-30	015			1/5	90	100u-230	5/5	145	73-388
(r.m. 93-95) Kiona (r.m. 20-23)	и	И	4/4	480	40-1500	314	60	20u-120	0/4	••		0/4			1/4	80	100u-180	4/4	109	77-136
Cle Elum	Resident	Eggs	2/2	20	14-30	1/2	100	20u-200	0/2			0/2			0/2			2/2	10	5-14
(r.m. 179-181) Wyer	spp.	11	2/2	260	250-270	1/2	220	20u-430	1/2	15	20u-20	0/2			0/2		**	2/2	10	9-11
(r.m. 134-136) Buena	u		313	840	710-980	2/3	220	20u-590	1/3††	250	20u-720	0/3**			1/3	190	100u-460	3/3	25	15-37
(r.m. 93-95) Kiona (r.m. 20-23)	11	H	1/1	540	540	1/1	30	30	0/1			0/1			0/1			1/1	11	11
Prosser (r.m. 46)	Salmonid smolts	Whole	2/2	340	100-570	0/2			0/2			0/2			0/2			2/2	28	24-32
Rosa	Spring	Muscle	4/4	120	20-390	1/4	10	20u-20	0/4			0/4			0/4			4/4	59	48-72
(r.m. 128)	chinook "	Eggs	1/2	20	20u-40	2/2	200	180-230	0/2			0/2			012		+-	2/2	11	11

u = Not detected at detection limit shown.

†Detection frequency = number of samples with a detectable concentration/total number of samples analyzed.

*One-half of the detection limit used in calculating mean where compound not detected.

†tEndosulfan sulfate detected in one sample.

**Endrin aldehyde detected in one sample.

Concentrations of organochlorine pesticides, PCBs, and mercury measured in water samples collected by Ecology from the Yakima River and tributaries in 1985 (detected compounds underlined). Table 15.

Sample Location	Sam- ple No.	Flow (cfs)	Temp.	рН (S.U.)	Sp. Cond. (umhos/cm)	Total Susp. Solids (mg/L)	Total Org. Carbon (mg/L)	p,p'- DDE (ug/L)	p,p'- DDT (ug/L)	p,p'- DDD t-DD (ug/L) (ug/		Endo- sulfan (ug/L)	PCBs (ug/L)	
June 24														
Yakima R. @ Cle Elum Wilson Creek " " Naches River Birchfield Drain Wide Hollow Creek Ahtanum Creek Yakima R. @ Parker Granger Drain Marion Drain Toppenish Creek Satus Creek Sulphur Creek Sulphur Creek Spring/Snipes Cr. Yakima R. @ Kiona	7091 7092 7093 7094 7095 7096 7098 7099 7100 7101 7102 7103 7104 7105 7106	3606 142 1280 57 17 7 478 61 90 3.2 135 291 64 1320	9.2 11.3 N/A 12.7 15.5 14.4 15.4 17.1 18.2 19.6 18.6 19.6 19.6 19.7	6.5 6.7 6.8 6.7 6.8 6.7 6.8 6.7 6.8 7.1 6.6 7.1 6.6 7.9	43 234 234 56 262 267 282 86 305 232 244 219 277 196 247 250	<1 27 27 <1 212 <1 41 278 30 5 15 217 61 13 18	June 24 1.6 4.4 4.0 N/A 3.2 2.2 2.8 2.1 3.1 4.8 2.9 3.0 5.1 2.6 3.2 3.2	0.01u 0.01u 0.01u 0.01u 0.013J 0.01u 0.01u 0.01u 0.01u 0.01u 0.01u 0.01u 0.01u 0.01 0.01	0.02u 0.02u 0.02u 0.02u 0.02u 0.02u 0.02u 0.02u 0.02u 0.02u 0.02u 0.02u 0.02u	0.02u 0.02u 0.02u 0.02u 0.02 0.02u 0.02u 0.02u 0.02u 0.02u 0.02u 0.02u 0.03 0.02u 0.02u 0.06 0.02u 0.06 0.02u 0.06 0.02u 0.06	0.01u 0.01u 0.01u 0.02 0.01u 0.004J 0.003J 0.02 0.01u 7 0.006J	0.01u 0.01u 0.01u 0.01u 0.01e 0.01u	0.05u	0.05u
July 2														
Birchfield Drain	7303	64	N/A	N/A	249	380	5.1	0.02	0.04	0.01 0.07	0.01u	0.03	0.05u	0.05u
Yakima R. @ Cle Elum Wilson Creek Naches River Birchfield Drain Wide Hollow Creek Ahtanum Creek Yakima R. @ Parker Granger Drain Marion Drain Toppenish Creek Satus Creek Sulphur Creek " Spring/Snipes Cr. Yakima R. @ Kiona	8000 8001 8003 8004 8005 8007 8008 8015 8009 8011 8002 8012 8013 8014	4006 532 434 89 29 11 342 60 128 9 80 331 127 1730	16.7 15.3 17.3 17.9 16.4 16.9 18.5 18.4 N/A 19.0 19.3 19.9 N/A 22.1 23.5	6.6 6.8 7.3 N/A 6.7 7.1 6.7 6.9 6.7 7.0 6.8 7.0 7.1	36 186 84 206 252 265 78 254 244 197 198 256 224 223 140 201	6 71 9 160 8 9 26 240 250 34 32 22 150 220 120 50	August 5 2.2 7.2 2.8 5.6 3.6 5.2 2.3 3.7 5.8 3.7 4.1 3.9 N/A 4.4 4.4 3.9 August 2	0.01u 0.01u 0.01u 0.01u 0.01u 0.01u 0.01u 0.02 0.01u 0.01u 0.01u 0.01u 0.01u 0.01u 0.01u	0.01u 0.01u 0.01u 0.01u 0.02u 0.02u 0.02 0.05u 0.02c 0.05u 0.02u 0.01 0.01u 0.01u	0.02u 0.02u 0.02u 0.02u 0.02u 0.02u 0.02u 0.02	0.01u 0.01u 0.01u 0.01u 0.01u 0.01u 0.01s	0.025u	0.05u 0.05u 0.05u 0.05u 0.05u 0.05u 0.05u 0.05u 0.05u 0.05u	0.05u
Birchfield Drain	8050	112	14.6	6.7	233	160	4.7	0.01	0.02	0.02u <u>0.03</u>	0.01u	0.02	0.05u	~0.05u
Yakima R. @ Cle Elum Wilson Creek Naches River Birchfield Drain Wide Hollow Creek Ahtanum Creek Yakima R. @ Parker Granger Drain " Marion Drain Toppenish Creek Satus Creek Sulphur Creek " Spring/Snipes Cr. Yakima R. @ Kiona	8157 8158 8159 8162 8160 8161 8163 8164 8165 8166 8167 8168 8170 8171 8172	117 890 20 21 12.5 2297 24 251 41 47 72	6.9 11.6 N/A 11.1 10.1 9.1 10.8 N/A 11.0	6.7 6.9 7.4 7.8 7.0 7.3 6.9 7.4 7.8 7.7 7.9 7.1 8.1 8.1 N/A	64 394 85 695 474 446 132 663 656 349 483 245 683 681 692 245	7 111 9 20 6 6 7 111 50 56 22 23 30 13 19 20 10 35	2.8 2.1 1.3 1.7 1.5 2.1 1.4 2.4 2.4 1.2 2.2 1.5 2.3 2.3 2.3 2.1 1.7	0.005u 0.005u 0.005u 0.005u 0.005u 0.005u 0.005u 0.005u 0.005u 0.005u 0.005u 0.005u 0.005u	0.01u 0.01u 0.01u 0.01u 0.01u 0.01u 0.01u 0.01u 0.01u 0.01u 0.01u 0.01u	0.01u	0.005u	0.005u 0.005u 0.005u 0.005u 0.005u 0.005u 0.005u 0.005u 0.005u 0.005u 0.005u 0.005u 0.005u 0.005u	0.05u 0.05u 0.05u 0.05u 0.05u 0.05u 0.05u 0.05u 0.05u 0.05u 0.05u 0.05u 0.05u	0.05u 0.05u 0.05u 0.05u 0.05u 0.05u 0.05u 0.05u 0.05u 0.05u 0.05u 0.05u 0.05u

NOTE: See Table 5 Part A for detection limits of organochlorine pesticides analyzed for but not detected u = Not detected at detection limit shown. J = Estimated concentration. N/A = Not analyzed.

The detection of endosulfan was limited to Birchfield Drain. A concentration of 0.16 ug/L—high relative to levels of DDT compounds and dieldrin—was present during the initial survey in June; subsequent samples contained 0.03 ug/L or less.

Although organochlorine pesticides were being discharged by a number of the tributaries monitored, these compounds were detected only once in water from the main stem of the Yakima River. Concentrations of p,p'-DDE and dieldrin were estimated at 0.006 - 0.007 ug/L in replicate samples collected in June from the lower river at Kiona. Endosulfan was not detected in Yakima River water.

Organochlorines were never detected in water from the Yakima River at Cle Elum, Yakima River at Parker, Naches River, Wide Hollow Creek, Ahtanum Creek, or Marion Drain.

Very few additional organic compounds were identified in the broad spectrum screen done on selected water samples.† Diazinon, an organophosphorus pesticide, was measured at a concentration of 1.7 ug/L in an August 5 sample from Birchfield Drain. No other organophosphorus, carbamate, or phenylurea pesticides or herbicides were detected. Potential shortcomings in these analyses have been previously discussed.

Low levels of ten organic compounds were detected by analysis for acid/base-neutrals and volatiles. These are listed in Table 16. All are commonly observed contaminants of surface waters. Potential sources include petroleum (aromatic hydrocarbons), refrigerants (chloromethane), fumigants (bromomethane), solvents (chlorinated ethylenes and 4-methylpentanone), and plastics (di-n-octylphthalate).

Table 16. Organic compounds detected by acid/base-neutral and volatiles analyses of water samples collected by Ecology from the Yakima River and tributaries on June 24, 1985 (ug/L).

Compound	Location: Sample No:	Wison Creek 7092	Marion Drain 7100	Sulphur Creek 7103	Yakima River at Kiona 7105/7106
Aromatic H to luene ethyl benz xyl ene 2-methylr		1J 1J 1J	1Ј	1J	13
Chlorinated trichloro tetrachlo		1J 1J			
Haloforms chloromet bromometh		1J	1J	1J 1J	1J 1J
Ketone 4-methyl	pentanone	1J			
Phthalate d-n-octy	lphthalate			1J	1 J

NOTE See Table 5, Part B for complete list of compounds analyzed and detection limits.

J = Estimated concentration

[†]Complete data set available on request.

The data from metals analyses of water are in Table 17. Low concentrations were observed in all samples analyzed.

Table 17. Results of metals analyses (other than mercury) on water samples collected by Ecology from the Yakima River and tributaries in 1985 (ug/L total recoverable metal).

Location	Date	Sample Number	Gı	Z'n	Ni	,Cr	a	Pb	Be	Sn	As	Se	Ag	Sb	TI
Wilson Creek	6/24 8/21	7092 8059	* 7	* 7	1u 	1u	0.1u	1u	0.1u not a					1u	1 u
Marion Drain	6/24 8/20	7100 8055	* 2	* 4	7 -	1u_	0.2	5 -	0.1u not a	lu naly	2 zed	_ l u	0.1u	_1u	l u
Sulphur Creek	6/24 8/20	7103 8053	* 4	* 10	8 -	1u_		7 -	0.1u not a	1u naly	5 zed	_1 <u>u</u>		_1 u	1u
Yakima R. at Kiona	6/24 6/24 8/20 8/20	7105 7106 8051 8052	* 33	* * 4	15 2 	1u 1u 	0.2	1 2 -	0.1u 0.1u not a not a	-	6 zed	_1u	0.lu 0.lu 		1u 1u

^{* =} Data not usable due to contamination in methods blank.

Sediment

The concentrations of organochlorines and mercury measured in bed sediments collected at five locations in the Yakima River and at the mouths of six tributaries are reported in Table 18. In line with the previously described findings for fish and water, DDT compounds and dieldrin were the predominant organochlorines detected in sediment. Endosulfan, aldrin, and isodrin were also detected, but in only one sample each—the latter two compounds being previously undetected in fish or water. Mercury was above detection limits in all but one sediment sample. PCBs could not be detected in sediment at a detection limit of 1.5 ug/Kq.

P,p'-DDE and p,p'-DDT comprised the major fraction of t-DDT in sediment. 0,p'-DDT and p,p'-DDD were also detected. Relatively high t-DDT concentrations of 234 ug/Kg and 91.7 ug/Kg were measured in replicate samples from the mouth of Sulphur Creek. As previously shown, concentrations of DDT compounds in water samples from this creek were among the highest of the eleven tributaries monitored. An elevated t-DDT concentration; 34.6 ug/Kg, also occurred in sediment for a second documented DDT source, Granger Drain.

Although Birchfield Drain had consistently detectable DDT compounds in water, the concentration of t-DDT in the sediment sample was only 4.0 ug/Kg. Sediment collected a short distance upstream from the mouth of Birchfield Drain (data presented later in Figure 2' and Appendix III) showed a range in t-DDT concentration of 27 - 31 ug/Kg. The low t-DDT concentration at the mouth of Birchfield Drain is probably due to the sample being 93 percent sand and gravel and having a total organic carbon (TOC) content of only 0.5 percent. Organochlorines have a high affinity for organic carbon and are preferentially associated with the silt-clay fraction in sediments. When the t-DDT data for Yakima tributaries are normalized to TOC, the following ranking is obtained: Sulphur Creek > Granger Drain > Spring/Snipes Creek > Wilson Creek, Birchfield Drain > Marion Drain.

u = Not detected at detection limit shown.

Table 18. Concentrations of organochlorine pesticides, PCBs, and mercury in bed sediment samples collected by Ecology from the Yakima River and tributaries on September 24 and 25, 1985 (ug/Kg dry).

Sample Location	Sample Number	% Dry Wt.	% Total Org. Carb.	Gravel (>2mm)	Grain Sand (2mm- 62um)	Size Silt (62um -4um)	Clay (<4um)	p,p'-	p,p'- DDT	o,p'-	p,p'-	t-DDT	t-DDT TDC Corrected	Diel- drin	Endo- sul- fan	Al- drin	PCBs	Mer- cury
Yakima R. @ Cle Elum Wilson Cr.	8129 8130	75 71	0.6 0.9	0	97.91 80.06	1.00 16.78	1.09 3.76	0.1J 4.9	0.lu 2.2	0.1u 0.66	0.1u 1.0	0.1 8.8	17 980	0.1u 2.6	0.lu 0.lu	0.1u 0.1u	1.5u 1.5u	28 9
Yakima R. @ Rosa Dam Birchfield Drain	8131 8132	48 80	3.1 0.5	0 9.10	38.02 84.35	58.73 1.40	3.25 5.15	17.4 1.3	27.3 1.8	3.8 0.62	16.9 0.33	65.4 4.0	2,100 800	8.2 0. l u	0.lu 0.lu		1.5u 1.5u	34 4u
Yakima R. @ Wapato Dam Granger Drain Marion Drain Sulphur Cr.	8133 8134 8135 8136 8137	46 75 70 67 87	2.3 0.3 2.3 1.5 0.4	0.08 0 3.19 0 0.77	46.13 87.61 90.33 48.61 86.35	49.89 2.50 4.37 48.47 11.21	3.90 9.89 2.11 2.92 1.67	65.7 10.5 7.0 129 33.2	11.1 19.3 2.1 59.2 50.2	2.1 1.2 0.32 8.4 1.6	15.4 3.6 2.2 37 6.7	94.3 34.6 11.6 234 91.7	4,100 12,000 500 16,000 23,000	2.4 0.7 0. l u 14.9 3.1	4.1 0.1u 0.1u 0.1u 0.1u	0.1u 0.1u 0.1u 0.1u 0.1u	1.5u 1.5u 1.5u 1.5u 1.5u	32 8 11 19 7
Yakima R @ Prosser Dam Spring/Snipes Cr.†	8138 8141 8139	79 79 79	0.2 0.1 0.3	0 0 0.79	97.86 98.72 89.24	1.46 0.68 7.70	0.68 0.60 2.34	1.5 1.2 4.1	0.41 0.31 3.8	0.10 0.13 0.81	0.41 0.35 1.0	2.4 1.9 9.7	1,200 1,900 3,200	0.1J 0.1u 10.1	0.1u 0.1u 0.1u	0.lu 0.lu 1065	1.5u 1.5u 1.5u	24 18 4
Yakima R @ Horn Rapids Dam	8140	64	1.0	0.01	52.44	44.31	3.24	30.1	3.4	0.71	5.3	39.5	4,000	1.7	0.1u	0.1u	1.5u	22

NOTE: See Table 5 Part A for detection limits of organochlorine pesticides analyzed for but not detected

t = Isodrin also tentatively identified at an estimated concentration of 17 ug/Kg dry. u = Not detected at detection limit shown. J = Estimated concentration.

T-DDT concentrations in sediment from the mainstem of the Yakima River ranged from the detection limit of 0.1 ug/Kg in the upper river at Cle Elum (r.m. 183.1) to 94.3 ug/Kg behind Wapato Dam (r.m. 103.8). With the exception of sediment collected at Prosser, there is a general downstream trend in increasing t-DDT concentration when the data are normalized to TCC.

Dieldrin concentrations in sediment were typically an order of magnitude lower than t-DDT. The highest concentration, 14.9 ug/Kg, occurred in Sulphur Creek.

The detection of 4.1 ug/Kg endosulfan in Wapato Dam sediment may reflect the previously documented chronic discharge of this pesticide by Birchfield Drain 3.8 miles upstream. Endosulfan was not detectable in the Birchfield sediment sample, again probably due to TOC and grain size characteristics.

An extremely high aldrin concentration of 1,065 ug/Kg was measured at the mouth of Spring/Snipes Creek. Aldrin was not detected in any other fish, water, or sediment samples collected during the study. The unusual nature of this finding is discussed later in the report. Isodrin, an isomer of aldrin, was also tentatively identified in Spring/Snipes Creek sediment. Isodrin is not a commercially produced agricultural chemical.

Mercury concentrations in Yakima River and tributary sediment were between less than 4 ug/Kg and 32 ug/Kg. These concentrations are typical of soils and rocks (Parker, 1967).

Birchfield Drain Investigations

The results of attempts to identify sources of organochlorine pesticides by analysis of sediment and water from one Yakima tributary, Birchfield Drain, are depicted in Figure 2 (t-DDT) and Figure 3 (endosulfan). The data from which these figures were prepared are in Appendices III and IV. As previously noted, dieldrin was not detected in this discharge.

The initial survey involved a series of bed sediment samples collected on March 25. t-DDT concentrations ranged from 9 - 98 ug/Kg. Results from analysis of replicate samples collected from Hubbard Ditch and lower Birchfield Drain indicated a variability at those sites of 9 percent and 48 percent about the mean (see Appendix III). Considering this variation in sample concentration observed at individual locations, there was no evidence of significant differences in the level of t-DDT in sediment between upper and lower portions of the drain. However, two laterals, Postma Ditch and Hubbard Ditch, did have higher t-DDT concentrations than other parts of the drainage--71 and 98 ug/Kg, respectively. (Normalizing these data to TOC and percent fines [silt + clay] gave no additional information on gradients in contamination.)

A water sample collection on July 2 followed the sediment survey and included samples from several points in Birchfield Drain and in Hubbard Ditch, Postma Ditch, and the other major lateral, Union Gap Canal. As shown in the figure, water concentrations of t-DDT in Postma and Hubbard Ditches were at or below detection limits and did not reflect the elevated DDT observed earlier in sediment at these sites. The third lateral, Union Gap Canal, had an elevated t-DDT concentration of 0.06 ug/Kg, also not predicted by the sediment analysis.

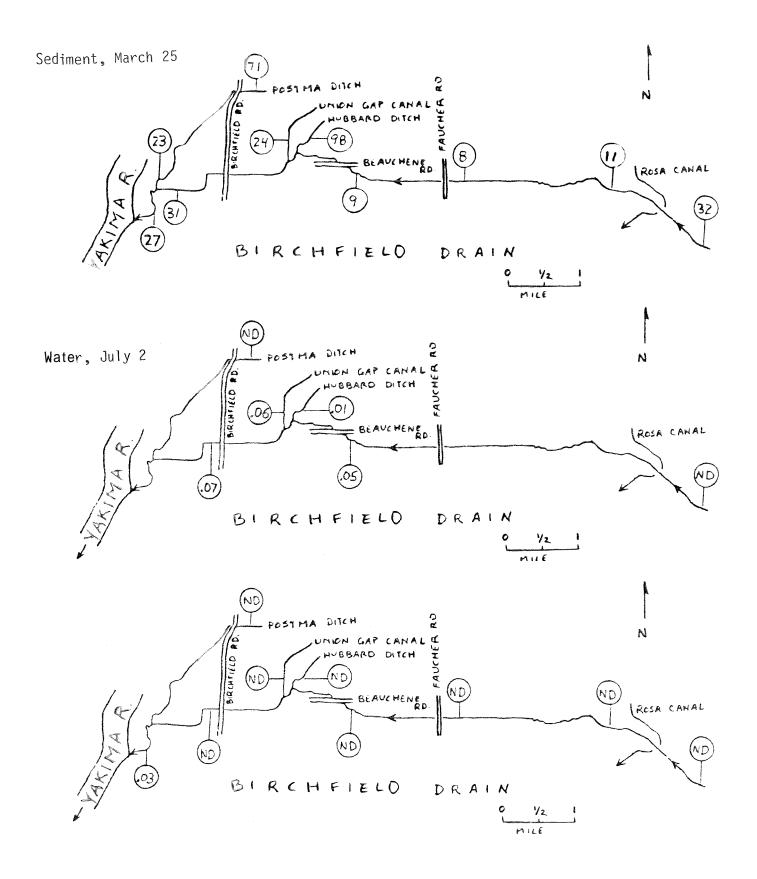
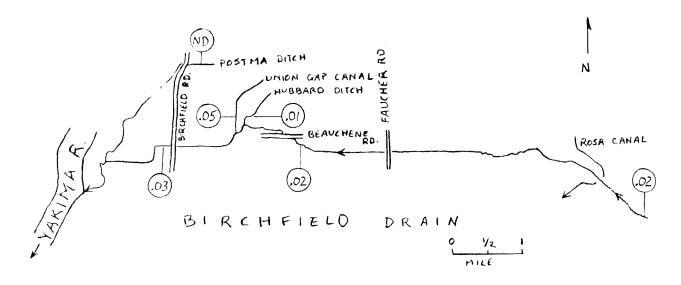


Figure 2. Concentrations of t-DDT measured in Birchfield Drain sediment and water samples collected by Ecology in 1985. (Detection limit = 1 ug/Kg, dry for sediment and 0.01 ug/L for water; ND = not detected.)



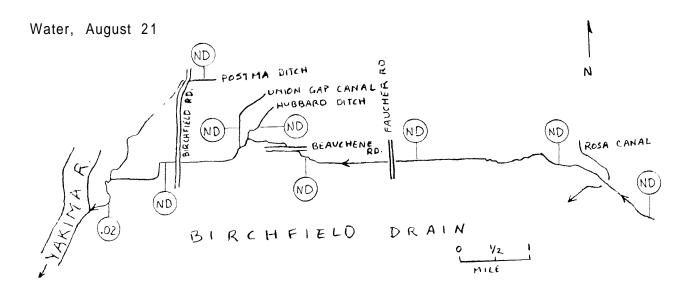


Figure 3. Concentrations of endosulfan measured in Birchfield Drain water samples collected by Ecology in 1985. (Detection limit = 1 ug/L; ND = not detected.)

Data on the main stem of Birchfield Drain above all three laterals showed t-DDT increased from below detection limits in the upper drain above Rosa Canal to 0.05 ug/L above the confluence of the laterals. This value was confirmed by analysis of a replicate (see Appendix IV). The water concentration in Birchfield Drain below Hubbard Ditch and Union Gap Canal was 0.07 ug/L. Based on these data, it appeared that during this sampling effort a source or sources above the major laterals to Birchfield was having a greater impact on t-DDT in water than Hubbard Ditch, Union Gap Canal, and Postma Ditch.

In a follow-up water collection on August 21, two sampling sites were added between Rosa Canal and Hubbard Ditch. The results shown in the figure indicate reduced t-DDT concentrations throughout the drainage. T-DDT reached a detectable level, 0.03 ug/L, only at the mouth of Birchfield Drain. Flow, suspended solids, and total organic carbon were much lower on August 21 than on July 2 (see Appendix IV).

The distribution of endosulfan in water on July 2 and August 21 parallelled that described for t-DDT (see Figure 3).

Results from the surveys described above and data presented previously on water and sediment from other parts of the Yakima River suggest DDT compounds are widely distributed in Birchfield Drain and in the Yakima basin as a whole and that the level of contamination in Birchfield Drain is similar to several other drains and creeks. The search for localized DDT sources in Birchfield Drain was not pursued further.

The Washington State Department of Agriculture (WSDA) also conducted an investigation of DDT in Birchfield Drain in 1985. This investigation included a determination of evidence for current use of DDT, analysis of the miticide Kelthane (dicofol) a suspected source of DDT compounds by way of impurities, and a survey for evidence of discarded DDT containers at dump sites. DDT sources were not found (Faulconer, 1985).

DISCUSSION

Implications for Human Health - During the present study, DDT compounds, dieldrin, endosulfan, PCB-1260, and mercury were detected in muscle samples from Yakima River fish. The Food and Drug Administration (FDA) has established the "action levels" shown below in Table 19, when these compounds are present in fish used for human consumption (FDA, 1984, 1985). FDA has no action level for endosulfan. The action levels represent a judgment by FDA balancing potential for adverse health effects against economic impacts. When a product exceeds an action level, FDA can legally remove it from the market.

Table 19. FDA "action levels" for contaminants identified in Yakima River fish muscle samples collected by Ecology in 1985.

Contaminant	Action Level	Reference
t-DDT [†] Dieldrin PCBs Mercury*	5,000 ug/Kg, wet 300 " 2,000 " 1,000 "	Compliance Policy Guideline 7120.23-D " 7120.23-A Federal Register, 1984. 49(100):21514-21520 Compliance Policy Guideline 7108.07

[†]DDT, DDE, DDD either singly or in combination; FDA does not count individual compound concentrations below 200 ug/Kg as part of t-DDT.
*methylmercury

Figure 4 plots the Yakima River fish muscle data by sample location and shows the relationship to FDA action levels. All samples are well below action levels. As noted earlier in this report, the data on mercury in tissue may underestimate actual concentrations. The mercury concentrations in these samples would not, however, be expected to exceed the 1,000 ug/Kg FDA action level even after allowing for the low results on spikes and reference materials.

Chinook eggs are part of the diet of the Yakima Indians. Although these data were not plotted, the concentrations of t-DDT, dieldrin, and mercury measured in spring chinook eggs easily meet FDA criteria. PCBs were not detected in chinook eggs.

EPA assesses the relative human health hazard from contaminants in fish by theoretical risk calculations for chemicals deemed carcinogens or by comparison with acceptable daily intakes (ADI's) for non-carcinogens. Carcinogens are considered to have no safe level of exposure because "...even a small dose must be assumed to elicit a finite increase in the incidence of response..." (EPA, 1980a). Non-carcinogens exhibit exposure (dose) thresholds below which adverse effects do not occur either due to "physiological reserve capacity" or "insignificant rate of damage." (EPA, 1980a). The concept of acceptable daily intake, therefore, is not applied to carcinogens.

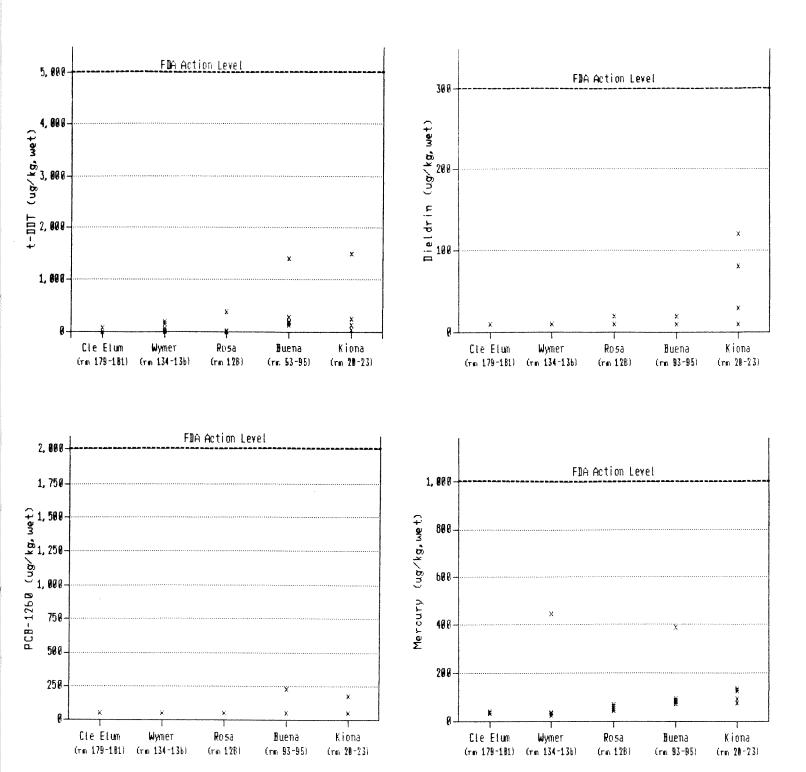


Figure 4. Comparison of FDA action levels with t-DDT, dieldrin, PCB-1260, and mercury concentrations in Yakima River fish and invertebrate muscle samples collected by Ecology in 1985. (Non-detections plotted at one-half the detection limit; equal concentrations at individual locations plot as single points.)

EPA considers the evidence for the carcinogenicity of DDT, DDE, DDD, dieldrin, and PCBs in humans to be inadequate. However, in consideration of evidence from animal studies, they have been classed as Group B2 - Probable Human Carcinogens by EPA's Carcinogen Assessment Group (EPA, 1985a; 1986a).

Increased lifetime (70-year) cancer risk for exposure to a carcinogen through fish consumption is calculated as shown below. Carcinogenic potency factors are expressed as a risk score which is the probability of effect per unit dose.

Chemical concentration in edible fish tissue (mg/Kg) x ingestion rate (kglday) \div 70 kg person = exposure (mg/Kg/day)

Exposure (mg/Kg/day) x carcinogenic potency factor $(mg/Kg/day)^{-1}$ = individual risk

Endosulfan and mercury are not carcinogenic. ADI's have therefore been calculated by EPA (1980d,e)--a 70-year daily exposure is also assumed. When the daily ingestion rate of a chemical exceeds the ADI (i.e., ingestion rate: ADI >1), adverse health effects are considered to potentially occur in sensitive subpopulations. Safety factors are applied in calculating ADI's to allow for adequacy of the supporting data.

A preliminary risk assessment following EPA methodology was done for consumption of Yakima River fish. For the purpose of the present study, the results are intended to allow comparison of relative risk from different carcinogens or amount of exposure not provide absolute quantification of risk.

The approach used in this analysis is conservative (i.e., worst-case assumptions are used). The carcinogenic potency factors employed represent the upper 95 percent confidence limit of the dose response from animal studies. The most up-to-date factors presently available were used (EPA, 1986b). Exposure is assumed to be constant over a 70-year lifetime. Tissue concentrations entered in the calculations were the average of the nine muscle samples analyzed from the lower Yakima River (Buena and Kiona) where the highest concentrations were observed. For samples where a chemical was not detected, one-half the detection limit was used in the calculation.

Information on the amount of fish eaten in the Yakima basin was not available, so a range of ingestion rates was evaluated. The lowest value, 6.5 glday (5.2 lbs/year) is EPA's estimate of the national average consumption of freshwater and estuarine fish and shellfish (EPA, 1980a). The U.S. Department of Agriculture has estimated a rate of 20 g/day (16 lbs/year) which includes freshwater, estuarine, and marine consumption (USDA, 1984). The highest of the three published rates evaluated was 165 g/day (133 lbslyear) which has been calculated to be the amount of fish consumed by 0.1 percent of the population (Finch, 1973).

Table 20 shows the results of the health assessment calculations described above as applied to eating fish from the lower Yakima River.

Table 20. Results of applying EPA health assessment methodology for consumption of fish from the lower Yakima River.

1. Carcinogens

Chemical	Mean Concen- tration in Fish Muscle (mg/Kg)	Carcino- genic Potency Factor	Average Lifet @ 6.5 g/day	ime (70-year) <u>@ 20 g/day</u>	Cancer Risk [†] @ 165 glday
p,p'-DDE p,p'-DDT p,p'-DDD Dieldrin PCB-1260	0.390 0.060 0.030 0.030 0.080	0.34 0.34 0.34 30.4 4.34	1 x 10-5 2 x 10-6 9 x 10-7 8 x 10-5 3 x 10-5	4 x 10-5 6 x 10-6 3 x 10-6 3 x 10-4 1 x 10-4	3 x 10-4 5 x 10-5 2 x 10-5 2 x 10-3 8 x 10-4
		2. N	lon-carcinogens	;	
<u>Chemical</u>	Mean Concen- tration in Fish Muscle (mg/Kg)	Accep- table Daily Intake (mg/day)	Ingestion Rat	e:Acceptable @ 20 g/day	Daily Intake @ 165 g/day
Endosulfan Mercury	0.020 0.130	0.28 0.02	0.0005 0.04	0.001 0.1	0.02 1

NOTE: See text for data sources.

The theoretical risks from intake of dieldrin and PCB-1260, although present at relatively low levels in Yakima River fish, are considerably greater than for DDT compounds. Endosulfan and mercury remain at or within acceptable daily intakes for all consumption rates evaluated.

Briefly, to put the above risk estimates in perspective, the lifetime cancer risk for the average smoker is 8 x 10^{-2} , while the risk from eating one-half pound of charcoal-broiled steak per week is 2 x 10^{-5} (Crouch and Wilson, 1984). The risks of dying in an automobile accident or being killed by lightning have been calculated at 2 x 10^{-2} and 4 x 10^{-5} , respectively (New York State Department of Health, 1981). A detailed discussion of risk assessment, especially as applied to eating fish, can be found in Tetra Tech (1986).

 $^{^{\}dagger}$ A lifetime risk of **1** x 10⁻⁶ is **1** in 1,000,000.

Potential Aquatic Life and Wildlife Impacts

The National Academy of Sciences (NAS, 1973) has recommended concentrations of toxic substances that should not be exceeded in whole fish if aquatic life and wildlife are to be protected. The recommendations include each of the contaminants detected in whole fish analyzed from the Yakima River during the present study. For "protection of predators," the following values were suggested: 1,000 ug/Kg (wet) for t-DDT; 100 ug/Kg (wet) for dieldrin, endosulfan, and endrin (singly or in combination); and 500 ug/Kg (wet) for PCBs. These organochlorine criteria were based on levels in water and fish and were designed to provide "... guidelines for water quality control. .." recognizing the fact that "... present knowledge is not yet sufficient to predict or estimate safe concentrations of these compounds in aquatic systems." The t-DDT recommendation incorporated the data available at that time on eggshell thinning induced by DDE in fish-eating and raptorial birds. (A diet containing as little as 2,800 ug/Kg, p,p'-DDE induced thinning in eggs of the American kestrel; i.e., sparrow hawk).

The NAS recommendation for mercury is 500 ug/Kg (wet) for protection of "selected fish and predatory species." This value was derived by considering that fish killed by methylmercury poisoning have contained 5,000 - 10,000 ug/Kg and that 100 - 200 ug/Kg is "apparently a usual background level for freshwater fish." Because data on safe mercury levels were not available, the FDA criterion of 500 ug/Kg was adopted, as it fell conveniently near background but well below toxic levels.

Figure 5 compares the Yakima River whole fish data with the NAS recommendations. The majority of samples are within a level considered protective for t-DDT. However, both largescale sucker (1,100 ug/Kg) and northern squawfish (3,000 ug/Kg) from the lower river at Kiona exceeded the NAS criterion of 1,000 ug/Kg. Sample size at Kiona was small but Ecology whole fish samples collected here since 1980 have consistently been in this range (See Table 1).

All but one of the fifteen samples analyzed for dieldrin were within the NAS recommendation. That sample, whole largescale suckers from Kiona, had 240 ug/Kg--more than twice the recommended level. One of the two sucker samples at Buena (sample number 8074, Table 11) also exceeds the 100 ug/Kg criterion when dieldrin, endosulfan, and endrin concentrations are summed, giving 120 ug/Kg (data point not shown in Figure 5).

All PCB-1260 and mercury concentrations in Yakima River whole fish were less than maximum levels recommended by NAS. Mercury concentrations would remain within NAS recommendations after allowing for the potential underestimation of sample concentrations described previously.

Other recent (i.e., since 1980) organochlorine analyses of tissue from Yakima River fish have been conducted by Ecology and USFWS. Several compounds not detected in the present study have been reported as present. These data are summarized in Table 21.

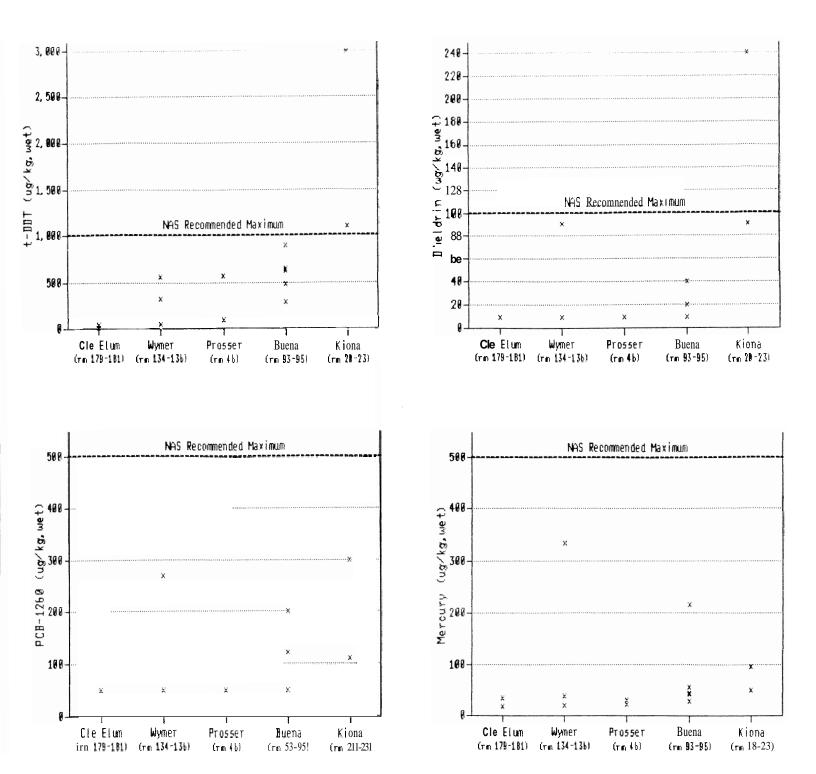


Figure 5. Comparison of NAS recommended maximum concentrations with t-DDT, dieldrin, PCB-1260, and mercury concentrations in Yakima River whole fish and invertebrate samples collected by Ecology in 1985. (Non-detections plotted at one-half the detection limit; equal concentrations at individual locations plot as single points.)

Table 21. Organochlorine compounds recently reported as present in fish tissue samples from the lower Yakima River but not detected in the Ecology 1985 study.

Compound	Detection Frequency [†]	Concentration Range (ug/Kg, wet)	Date	Investi- gator	Referenc	e		
a 1pha-BHC pentachlorophenol*	11/12 12/12	3 - 47 1 - 32	1984	Ecology	Hopkins,	e t	al.	(1985)
alpha-BHC	9/9	3.9 - 22	1983	•	u	n	11	u
alpha-BHC	2/2	6; 6	1982	11	ii	**	n	11
cis-chlordane trans-chlordane cis-nonachlor trans-nonachlor	2/2 2/2 2/2 1/2	93; 230 5.1; 17 32; 73 9.8	1981	н	н н н	#1 #1 #1	11 11 11	11 H H
alpha-BHC gamma-BHC cis-chlordane trans-chlordane trans-nonachlor	2/2 2/2 2/2 1/2 1/2	3.4; 3.7 0.3; 0.6 6; 21 9.2 6.6	1980	# #	11 H H	11 11 11	11 11 11	11 11 11 11
trans-chlordane cis-nonachlor trans-nonachlor toxaphene	2/3 1/3 1/3 1/3	10 10 10 400	# # # #	USFWS	Schmitt,	et u	al. 11	(1985) "

NOTE: Ecology data are on whole fish, muscle, liver, and viscera; USFWS data are on whole fish.

Ecology surveys done in the Yakima River between 1980 and 1984 detected alpha-and gamma-BHC (hexachlorocyclohexane), pentachlorophenol, and components of heptachlor (chlordane and nanochlor isomers). The USFWS detected heptachlor components and toxaphene in 1980. The concentrations reported by Ecology and USFWS have, for the most part, been low, rarely exceeding the NAS recommendation of 100 ug/Kg for organochlorine pesticides other than DDT compounds.

The relationship between the NAS t-DDT recommendation and concentrations in whole fish from the lower Yakima River raises the question of potential adverse effects on predatory bird populations. DDE reduces the production of birds by interfering with calcium metabolism which results in abnormally thin eggshells subject to early breakage (Ratcliffe, 1970; Heath 1969). The exact cause of thinning has not been established (McEwen and Stephenson, 1979). PCBs have also been shown to cause eggshell thinning (Heath, et al., 1970) but at concentrations well above those observed in Yakima River fish.

It is not universally agreed what dietary concentration of DDE constitutes a no-effect level. This subject has been recently reviewed in detail by Lincer (1985). The lowest levels of t-DDT in diets that have been reported as correlated with thin eggshells are from field observations; i.e., concomitant analysis of DDT compounds in prey items and measurement of thickness of

[†]Number of samples in which compound was detected/total number of samples analyzed.

^{*}Not analyzed in Ecology's 1985 study.

predator eggshells. These reported effect levels include 150 ug/Kg t-DDT in brown pelican diets (Anderson, et al., 1975) and 320 ug/Kg t-DDT in peregrine falcon diets (Enderson, et al., 1982). Lincer points out the following errors inherent in this type of correlative analysis:

- o The migratory nature of predator and prey were not considered.
- o The persistence of residues in predators and associated biological effects were more a reflection of historic than current environmental and body burdens (these observations were made during a period of rapid decline in environmental DDT levels following its ban).
- o Confounding effects of other chemicals and environmental factors were not considered.

It was concluded that controlled experiments are required to produce valid data. Under these conditions, 300 ug/Kg DDE resulted in no eggshell thinning of the American kestrel (Lincer, 1975); 2,800 ug/Kg DDE caused 9.7 percent thinning (Wiemeyer and Porter, 1970); and 3,000 ug/Kg DDE caused 15 percent eggshell thinning (Lincer, 1975).

These and other data (McClane and Hall, 1972) have shown that dietary concentrations of DDE must be greater than 3,000 ug/Kg for eggshell thinning to exceed 15 percent. Normally reproducing populations of both raptors and brown pelicans tolerate eggshell thinning in the range of 7 to 17 percent (Lincer 1985, and references therein). Lincer concluded that a chronic dietary level of up to 3,000 ug/Kg DDE will not result in population problems in most sensitive bird species.

The phenomenon of eggshell thinning has not been studied in the Yakima River There has been one study covering other parts of Washington and Oregon and Nevada which addressed the impact of DDE on black-crowned nightherons (Henny, et al., 1984). The study included two colonies in the Columbia River below the Yakima River confluence. The authors observed a "strong north-south clinal pattern" of DDE residues in eggs. Southern colonies were more contaminated than northern colonies. However, the two Columbia River colonies, Foundation [s] and 12 miles below the Yakima confluence and Three-Mile Island 70 miles below the confluence, had higher than expected residues which was interpreted as local contamination. Nevertheless, all northern colonies, including Foundation Island and Three-Mile Island, had adequate production to maintain a stable population. The most contaminated southern colony, Ruby Lake, Nevada, had eggshells that averaged 16.4 percent thinner than normal and below normal productivity (i.e., less than one young fledged per breeding pair). This demonstration of adverse effects at the population level for 16.4 percent eggshell thinning agrees well with the 15 percent criterion described above.

In conclusion, it appears likely that predatory birds which are sensitive to DDE, and feed exclusively on fish from the lower Yakima River would be expected to produce eggs with shell thickness somewhat below normal. The reduction in shell thickness experienced should not be sufficient, however, to prevent maintenance of a stable population.

The significance of the organochlorine concentrations measured in eggs from Yakima River fish and in downstream migrating salmonid smolts can be gauged by examination of Table 22 which summarizes the results of a literature search on effects of DDT compounds, dieldrin, and PCBs in eggs and fry of freshwater fishes. No information was found on endosulfan, endosulfan sulfate, or endrin aldehyde which were detected in one or two egg samples. Based on the weight of evidence in the available literature, the following generalizations can be made:

- o Concentrations of t-DDT less than 1,000 ug/Kg in fish eggs or fry do not cause substantial mortality.
- o Concentrations of t-DDT between 2,000 and 4,000 ug/Kg in fish eggs or fry may result in 10 to 30 percent mortality.
- o A dieldrin concentration of 800 ug/Kg in fish eggs does not adversely affect fry survival.
- PCB concentrations below approximately 2,000 ug/Kg in fish eggs do not cause substantial mortality.

The maximum concentrations of organochlorines measured in fish eggs from resident Yakima River species during the present study were 980 ug/Kg t-DDT, 590 ug/Kg dieldrin, and 460 ug/Kg PCB-1260. Eggs from returning spring chinook had up to 40 ug/Kg t-DDT and 230 ug/Kg dieldrin; downstream migrating chinook and steelhead smolts (whole fish) had 570 ug/Kg t-DDT and 100 ug/Kg t-DDT, respectively. In no instance do these concentrations exceed the effects thresholds indicated in the literature.

Salmonids are more sensitive to pesticides than warm-water fish (Jarvinen, 1977). Because nearly all the literature on this subject deals with salmonids, the margin of safety for warm-water species that spawn and rear in the lower Yakima River may be greater than suggested by the above comparison. However, only one sample of eggs was obtained from the lower Yakima River. The fact that organochlorine concentrations in whole fish at Kiona (r.m. 20-23) were much higher than at Buena (r.m. 93-95) brings the representativeness of the single egg value for Kiona of 540 ug/Kg into question since higher concentrations in eggs were measured upstream.

Macek (1985) evaluated potential adverse effects on fish reproduction by comparing whole body and egg t-DDT residues in female fish. His review of data in Burdick, et al. (1964), Macek (1968), and Locke and Havey (1982) indicated whole fish:egg ratios are typically 2:1. (Whole fish and egg data were not obtained simultaneously in the present study.) Whole-fish concentrations likely to result in t-DDT residues of 2,000 - 4,000 ug/Kg in eggs-and subsequent increased mortality--were therefore estimated to be on the order of 6000 ug/Kg, twice the maximum concentrations observed by Ecology in the lower Yakima River since 1980. (The potential for adverse reproductive effects on fish exposed to organochlorine levels characteristic of some Yakima River tributaries is discussed later in this report.)

Table 22. Effects of organochlorine residues in fish eggs and fry.

Species	Tissue	Concentration (ug/Kg, wet)	Effect	Mode of Exposure	Reference
			DDT Compounds		
Char Salvelinus alpinus	eggs	36-170 t-DDT	no fry mortality	natural accumulation	Monod (1985)
Brook trout Salvelinus fontinalis	eggs & fry	360-1000 t-DDT	no fry mortality	laboratory, DDT-3H (radio-labeled in feed)	Atchison (1970)
Coho salmon Onchorhynchus kisutch	eggs	1000-2000 t-DDT	<15% mortality	natural accumulation	Johnson & Pecor (1969)
Brook trout Salvelinus fontinalis	eggs	1200 t-DDT	40% fry mortality	natural accumulation	Currier, <u>et al</u> . (1967)
Brook trout Salvelinus fontinalis	eggs & fry	2000-3000 t-DDT	5-8% fry mortality	laboratory, technical grade DDT in feed	Macek (1968)
Lake trout Salvelinus namaycush	eggs	>2900 t-DDT	15% fry mortality	natural accumulation	Burdick, <u>et al</u> . (1964)
Coho salmon Onchorhynchus kisutch	fry	3400 t-DDT	30% fry mortality	natural accumulation	Williford, et al. (1969)
Rainbow trout Salmo gairdneri	fry	4630 t-DDT	increased mortality	natural accumulation	Dace & Scott (1971)
Coho salmon Onchorhynchus kisutch	eggs	6000-7000 t-DDT	15-73% mortality	natural accumulation	Johnson & Pecor (1969)
			<u>Dieldrin</u>		
Wal leye Stizostedion vitreum	eggs & fry	800 dieldrin	no significant fry mortality	laboratory, water	Hair (1972)
Wal leye Stizostedion vitreum	eggs & fry	12,000 dieldrin	increased fry mortality	laboratory, water	Hair (1972)
			PCBs		
Atlantic salmon	eggs	400-1900 PCBs	16-100% mortality	natural accumulation	Jensen, <u>et al</u> . (1970)
Brook trout Salvelinus fontinalis	eggs	1700-1800 PCB-1254	no adverse effects	laboratory, water	Snarski & Puglisi (1976)
Sheepshead minnow Cyprinodon variegatus	eggs	3700-9500 PCB-1254	16-19% fry mortality	laboratory, water	Hansen, et al. (1973)
			Mixtures		
Coho salmon Onchorhynchus kisutch	eggs	500-3500 t-DDT, 2000-9000 PCBs	no fry mortality	natural accumulation	Degurse, <u>et al</u> . (1973)
Rainbow trout Salmo gairdneri	eggs	90 t-DDT, 2700 PCB-1242	75% fry mortality	natural accumulation	Hogan & Brauhn (1975)
Lake trout Salvelinus namaycush	eggs	1410-5240 DDE, 3160-9900 PCBs	no effects on survival	natural accumulation	Stauffer (1979)
Lake trout Salvelinus namaycush	eggs	2500-7200 DDE, 3600-11,100 PCB-1254	no significant correla- tion with survival rate	natural accumulation	Mac, et al. (1981)
Atlantic salmon Salmo salar	eggs	710-3060 t-DDT, ND - 210 dieldrin, 1900-6500 PCB-1254	no significant correla- tion with hatchinq success	natural accumulation	Zitko & Saunders (1979)

ND = not detected

Significance of Sources Identified

Organochlorine analysis of the water samples collected in June, July, and August revealed detectable levels of p,p'-DDE, p,p'-DDT, p,p'-DDD, dieldrin, and endosulfan. This occurred almost exclusively in tributaries. The loads in pounds/day being discharged to the Yakima River by individual tributaries have been calculated in Table 23 and compared to the load carried in the main stem of the river.

The major source of DDT compounds identified among the eleven tributaries monitored was Sulphur Creek. T-DDT loads in this discharge were 0.09 pound/day on June 24 and 0.04 pound/day on August 5. Granger and Birchfield Drains appeared to be t-DDT sources of secondary importance, averaging about 0.01 pound/day. Spring/Snipes Creek was a lesser t-DDT source, with a measured load of 0.007 poundlday on June 24 only.

Dieldrin loads were lower than those for t-DDT by about a factor of 10. Sulphur Creek and Granger Drain were also the major dieldrin sources identified (0.003 - 0.02 pound/day). Smaller dieldrin loads in the 0.002 - 0.005 pound/day range were measured once each in Wilson, Satus, and Spring/Snipes Creeks. An extremely small load of 0.00007 pound/day was calculated for Toppenish Creek on June 24.

As noted previously, endosulfan was detected only in Birchfield Drain; loads up to 0.05 poundlday occurred.

The total load of t-DDT going into the Yakima River from the sources identified was 0.1 pound/day on June 24 and 0.05 poundlday on August 5. The total dieldrin loads measured on these dates were 0.01 pound/day and 0.03 poundlday, respectively. Based on these limited data and assuming transport of these compounds occurs only from the above-mentioned sources and primarily during the six-month irrigation season (April - September), total yearly loads were estimated to be on the order of 14 pounds for t-DDT, 4.5 pounds for dieldrin, and 3.6 pounds for endosulfan.

Comparison of the t-DDT load measured in discharges to the Yakima River with the load in the lower river at Kiona suggests that major sources have been accounted for. However, the bulk of t-DDT (and dieldrin) in natural waters occurs in association with particulate matter (Callahan & Slimak, 1979). As a result, DDT compounds do not behave as conservative constituents (i.e., concentration changes do not follow simple dilution). An unknown, and perhaps large, fraction of the t-DDT load to the Yakima River may be incorporated into and transported with the bed sediments (bed load).

The data obtained from the present study are not sufficient to definitively identify major sources. The tributaries monitored are, however, the major sources of sediment to the Yakima River (U.S. Army Corps of Engineers, 1978; U.S. Department of Agriculture, 1978). If the concentrations of organochlorine pesticides observed in these tributaries are typical of the Yakima basin, it follows that they are also the major contaminant sources.

Table 23. Organochlorine pesticide loads measured by Ecology in the Yakima River and tributaries in 1985 (poundsIday).

Di~charge	t-DDT	Dieldrin	Endosulfan			
		Ju	ne 24			
Yakima R. @ Cle Elum Wilson Creek Naches River Birchfield Drain Wide Hollow Creek Ahtanum Creek Yakima R. @ Parker Granger Drain Marion Drain Toppenish Creek Satus Creek Sulphur Creek Spring/Snipes Creek Yakima R. @ Kiona	ND ND O. 009 ND ND ND O.01 ND ND O.09 O.09 O.007	ND 0.005 ND ND ND ND 0.0007 ND 0.00007 0.002 0.003 ND 0.004	ND ND ND 1.05 ND ND ND ND ND ND ND ND ND			
total load to river load in river @ Kiona	0.1 0.05	0.01 0.04 July	0.05 ND			
Birchfield Drain	0.02	ND ND	0.01			
		August	5			
Yakima R. @ Cle Elum Wilson Creek Naches River Birchfield Drain Wide Hollow Creek Ahtanum Creek Yakima R. @ Parker Granger Drain Marion Drain Toppenish Greek Satus Creek Sulphur Creek Spring/Snipes Creek Yakima R. @ Kiona	ND N	ND ND ND ND ND ND ND ND ND O.005 ND	ND N			
total load to river load in river @ Kiona	0.05 ND	0.03 ND	ND ND			
	gelanifolio del Printi	August	21			
Birchfield Drain	0.02	ND	0.01			
	October 28					
	No orga	nochlorines d	etected			

Note: Flow (cfs) x concentration (ug/L) x 10^{-3} x 5.39 = load (pounds/day). See Table 14 for data.

ND = Not detected.

Although the organochlorine pesticide concentrations on which these loads were based are in the parts per trillion range (i.e., 0.003 - 0.16 ug/L), there remains the potential that aquatic life could be adversely affected due to their high toxicity and bioaccumulation potential. The most widely accepted criteria for judging the impact of contaminants in water are those published by EPA (1980a-d) and NAS (1973). The EPA criteria are under consideration by Ecology for adoption as state water quality standards (Moore, 1985). Present state standards address the issue of organochlorine pesticides by stipulating that (for Class B waters such as the lower Yakima River) "Toxic . . or deleterious material concentrations shall be below those of public health significance, or which may cause acute or chronic toxic conditions to the aquatic biota, or which may adversely affect characteristic water uses . . " (Washington Administrative Code, 1982). State water quality standards are applicable to irrigation wasteways as well as natural water bodies (Roe, 1969).

The EPA water quality criteria are two-part: (1) a concentration not to be exceeded (acute exposure), and (2) a 24-hour average concentration (chronic exposure). The criteria are based on results from laboratory experiments and data on contaminant residues in aquatic organisms. Where data are available on twenty or fewer species, the acute criteria represent the lowest concentration (LC50 or EC50) found to elicit a toxic response; a calculated value is used when there are data on more than twenty species. The calculated acute value is not the lowest value at which toxicity has been observed in the laboratory. The EPA criteria are intended to "protect most, but not necessarily all, aquatic life and its uses" (EPA, 1980a).

The EPA criteria for chronic exposure to t-DDT and dieldrin are based on data for residues in aquatic organisms. EPA did not find sufficient data to derive chronic criteria for DDE or DDD. EPA's chronic criterion of 0.001 ug/L t-DDT was calculated by taking 150 ug/Kg, the lowest of the concentrations (primarily DDE) observed in the diet of brown pelicans during the period 1969 - 1974 when reduced eggshell thickness caused low hatching success, dividing by 17,870, a geometric mean of lipid-normalized bioaccumulation factors, and dividing by a lipid value of 8 percent for pelican diets. The criterion of 0.001 ug/L t-DDT in water, therefore, is intended to keep t-DDT below 150 ug/Kg, the "lowest maximum permissible tissue concentration" based on reduced productivity of a sensitive species.

As discussed previously, the case for eggshell thinning at a dietary level of 150 ug/Kg is weak. The EPA chronic criterion of 0.001 ug/L for t-DDT is, therefore, conservative. However, other independently derived t-DDT criteria (in addition to the NAS criteria discussed below) call for similar low levels in water. Macek (1985) concluded a t-DDT concentration of 0.0037 ug/L would not cause significant adverse impact to aquatic organisms. The American Fisheries Society (Thurston, et al., 1979) recommended an even lower criterion of 0.0001 ug/L to protect against the potential for sublethal effects on fish.

The EPA chronic criterion for dieldrin is 0.0019 ug/L. It was calculated in a manner similar to that for the t-DDT criterion, but used the FDA action level of 300 ug/Kg as a maximum permissible fish tissue concentration. Residue data were not available for endosulfan. The EPA chronic endosulfan criterion of 0.056 ug/L is the result of dividing the acute criterion of 0.22 ug/L by an acute:chronic ratio of 3.9 obtained from laboratory data.

The NAS criteria for DDT, DDD (no DDE criteria are given), dieldrin, endosulfan, and other pesticides were derived by multiplying the lowest reported 96-hour LC50 by an "application factor" (safety factor) of 0.01. These criteria represent the "recommended maximum concentrations ... in whole (unfiltered) water sampled at any time and any place" (NAS, 1973). Both the EPA chronic and NAS approaches result in similar criteria for DDT and dieldrin; the NAS endosulfan criterion is much lower than EPA's.

Table 24 gives the EPA and NAS criteria for organochlorines detected in water from the Yakima River and tributaries and shows where concentrations exceeded criteria. Violations of criteria to protect aquatic life under conditions of acute exposure were not observed. In most instances, water concentrations of DDT, DDE, DDD, dieldrin, and endosulfan were two or more orders of magnitude lower than the acute criteria. The endosulfan concentration of 0.16 ug/L recorded for Birchfield Drain on June 24 approached EPA's 0.22 ug/L acute criterion.

The EPA acute criterion of 1,050 ug/L for DDE is probably too high. More recent studies (Johnson and Finley, 1980) have shown LC50s for DDE on the order of 32 ug/L for rainbow trout, 96 ug/L for Atlantic salmon, and 240 ug/L for bluegill--still many times above concentrations observed in the present study.

Numerous violations of the EPA chronic exposure criterion and NAS maximum recommended concentrations for t-DDT, DDT, dieldrin, and endosulfan, however, did occur. The most frequent and severe violations were observed in Birchfield Drain (t-DDT, p,p'-DDT, p,p'-DDT, p,p'-DDT, dieldrin). Violations of less magnitude were observed once each for t-DDT, p,p'-DDT, or dieldrin in Spring/Snipes Creek, the Yakima River at Kiona, Toppenish Creek, and Satus Creek. The diazinon concentration measured in Birchfield Drain on August 5 (see Table 23 footnote) exceeds the LC50 reported for some freshwater invertebrates (Johnson and Finley, 1980). Ecology has previously documented elevated diazinon in Birchfield Drain (Johnson and Newman, 1983). Table 25 summarizes the water quality criteria violations documented in the present study.

Table 25. Frequency of water quality criteria violations for organochlorine pesticides observed during Ecology's 1985 survey of the Yakima River and tributaries. (Number of samples above criteria shown; 3 samples collected at each site except 5 samples at Birchfield Orain.)

	EPA Water Quality Criteria						Recomme Concent	ration
Location	t- DDT	Diel- drin	tndo- sulfan		DDT	DDD	Diel- drin	Endo- sulfan
Birchfield Drain [†]	3	0	1		3	1	0	3
Sulphur Creek	2	2	0		2	0	2	0
Granger Drain	2	2	0		2	0	2	0
Spring/Snipes Creek	1	1	0		1	0	1	0
Yakima R. @ Kiona	1	1	0		0	0	1	0
Wilson Creek	0	1	0		0	0	1	0
Toppenish Creek	0	1	0		0	0	0	0
Satus Creek	0	1	0		0	0	0	0
Marion Drain	0	0	0		0	0	0	0
Wide Hollow Creek	0	0	0		0	0	0	0
Ahtanum Creek	0	0	0		0	0	0	0
Naches River	0	0	0		0	0	0	0
Yakima R. @ Parker	0	0	0		0	0	0	0
Yakima R. @ Cle Ełum	0	0	0		0	0	0	0

 $^{^\}dagger$ Diazinon, an organophosphorus pesticide, exceeded the NAS recmmended maximum concentration in Birchfield Orain on one occasion (see Table 23 footnote).

Table 24. Instances during Ecology's 1985 survey of the Yakima River and tributaries where organochlorine pesticide concentrations in water exceeded EPA or NAS criteria for protection of freshwater aquatic life.

						ality Crite	ria			NAS	Recomm Concer	ended M ntrations	
		Concen-	t-DDT	DDD	DDE	Dield		Endosu				Diel-	Endo-
Location	Compound	tration (ug/L)	0.001 1.1 (chronic) (acute)	O.6 (acute)	1,050 (acute)	0.0019 (chronic)	2.5 (acute)	0.056 (chronic)	0.22 (acute)	DDT 0.002	DDD 0.006	drin 0.005	sulfar 0.003
					June	24							
Wilson Creek	dieldrin (repl)	0.006J 0.007J				X X						X X	
Birchfield Drain	p,p'-DDT t-DDT endosulfan	0.016 0.029J 0.16	X X					X		Х			Х
Granger Drain	p,p'-DDT t-DDT dieldrin	0.03 0.03 0.02	X X			X				X		Х	
Toppenish Creek	dieldrin	0.004J				Х							
Satus Creek	dieldrin	0.003J				Χ							
Sulphur Creek	p,p'-DDT t-DDT dieldrin	0.03 0.06 0.02	X X			X				Х		X	
Spring/Snipes Creek	p,p'-DDT t-DDT	0.011J 0.02J	X X							X			
Yakima River @ Kiona "	t-DDT (repl) dieldrîn (repl)	0.006J 0.007J 0.006J 0.005J	X X			X X						X	
					July	2							
Birchfield	p,p'-DDT	0.04	X							Х			
Drain #	p,p'-DDD t-DDT endosulfan	0.01 0.07 0.03	Х								Х		Х
					Augu	ıst 5tt							
Granger Drain	p,p'-DDT	0.02	X							Х			
и	t-DDT dieldrin (repl)	0.04 0.014J 0.016J	X			X X						X X	
Sulphur Creek	p,p'-DDT t-DDT dieldrin	0.01 0.02 0.012J	X X			X				Х		X	
11	(repl)	0.014J				X						x	
Spring/Snipes Creek	dieldrin	0.008J			Λ	X						X	
Birchfield Drain	p,p'-DDT t-DDT endosulfan	0.02 0.03 0.02	X X		<u>Augt</u>	<u>ıst 21</u>				Х			X

J = estimated concentration.

[†]U.S. Environmental Protection Agency, 1980. Water Quality Criteria Documents: Availability. Federal Register, Vol. 45, No. 231. *National Academy of Sciences/National Academy of Engineering, 1973. Water Quality Criteria - 1972. EPA.R3.73.033. 594 pp.

^{††}Diazinon, an organophosphorus pesticide, was detected at 1.7 ug/L in Birchfield Drain on August 5 which exceeds the NAS recommended maximum concentration of 0.009 ug/L.

The above criteria do not address adverse effects on aquatic life from exposure to combinations of organochlorine pesticides. There are limited data on this subject. Macek (1975) exposed rainbow trout and bluegills to DDT and dieldrin both singly and in combination for a 96-hour period. The action of combined DDT and dieldrin was additive; i.e., mortality was approximately equal to the sum of mortalities observed for DDT exposure and dieldrin exposure.

The implications of the water quality criteria violations observed in the Yakima tributaries with the highest organochlorine concentrations are: (1) sensitive aquatic species may be adversely affected either through direct toxicity or impaired reproduction; (2) birds feeding on fish from these tributaries may have below-normal reproductive rates; and (3) fish in these tributaries may not meet FDA standards for human consumption. For example, bioaccumulation factors for t-DDT in freshwater fish are on the order of 104 - 10^6 (EPA, 1980c). At a water concentration of 0.06 ug/L t-DDT (as measured in Sulphur Creek on June 24--see Table 15) and a bioaccumulation factor of 10^5 , fish might be expected to have 6,000 ug/Kg t-DDT. (Interestingly, t-DDT concentrations in whole fish collected below the mouth of one of the abovementioned tributaries, Birchfield Drain, have reached 5,100 uq/Kq--see Table As previously shown, a t-DDT concentration of 6,000 ug/Kg is sufficient to cause mortality in salmonid eggs and fry, exceeds both the NAS recommended maximum and FDA action level criteria, and is sufficient to prevent maintenance of stable populations in sensitive fish-eating bird species. that some of these tributaries have been degraded to the point that they are no longer important habitats does not diminish the potential significance of the existing contamination.

Concentrations of DDT compounds and dieldrin in the main stem of the Yakima River (and in the Naches River) appeared to be, at worst, not appreciably above the 0.001 - 0.005 ug/L range of the EPA and NAS water quality criteria, although the method detection limit prevented quantification at this extremely low level. On this basis, there does not appear to be evidence of a toxicity problem due to organochlorine concentrations in Yakima River water.

No criteria or toxicity data are available to evaluate the potential for adverse effects on benthic invertebrates due to the concentrations of organochlorines measured in bed sediments during the present study.

Recent or Historical Sources?

Information on the eight major organochlorine compounds detected in Yakima River fish, water, or sediment during this study is summarized in Table 26 showing alternative names, structures, and present use status.

Figure 6 traces the degradation of DDT to DDE, DDD, and other compounds. DDE is more persistent than either DDT or DDD and, therefore, should predominate in environmental samples away from sources. Technical DDT has a p,p'-DDT: o,p'-DDT ratio of approximately 5:1 (Gunther and Gunther, 1971; Brookes, 1979). Because the o,p' isomer has been reported to be less stable than the p,p' isomer, its presence has also been used to indicate the age of DDT mixtures in environmental samples.

Table 26. Information on organochlorines detected in Yakima River fish, water, or sediment samples collected by Ecology in 1985

CAS Number	Common Name	Trade Names [†]	Chemical Name [†]	Structure*	Status ^{††}	Reference ^{††}
50-29-3	DOT	Anofex Chloro-phenothane Genitox	1,1,1-trichloro-2,2-bis(p-chlorophenyl)ethane (principal component)	a - ç- a	Cancel led all products except: 1. USPHS and other Health Service officials for control of vector diseases. 2. USDA or military for health quarantine. 3. In prescription drugs for body lice control.	PR Notice 71-1 January 15, 1971 and 37 FR 13369 July 7, 1972
72-54-8	DDD (TDE)	Rothane	1,1-dichloro-2,2- bis(p-chlorophenyl)ethane	CICCI CI	Cancelled all products.	PR notice 71-5 March 18, 1971
72-55-9	DDE	(DDT metabolite)	1,1-dichloro-2,2- bis(p-chlorophenyl)ethylene	CI CCI,	(DDT metabolite, not regulated)	
60-57-1	Dieldrin	HEOD Octalox Panoram	hexachloroepoxyoctahydro- endo, exo-dimethanonaphthale (85%; related compounds 15%)	CI H H H H	Most uses cancelled except: 1. Subsurface ground insertion for termite control. 2. Dipping non-food roots and tips. 3. Moth-proofing by manufacturing process in closed system.	PR notice 71-4 March 18, 1971 and 37 FR 372146 October 18, 1974
309-00-2	Aldrin	Aldrosol Compound 118 Seedrin	hexachlorohexahydro-endo, exo-dimethanonaphthalene (95%; related compounds 5%)		See dieldrin	See dieldrin
115-29-7	Endosulfan	Insectophene Malix Thiodan	hexachlorohexahydromethano- 2,4,3-benzodioxathiepin 3-oxide	0 Cl CC	Registered; no restrictions.	
72-20-8	Endrin	Compound 269 Hexadrin Mendrin	1,2,3,4,10,10-hexachloro-6,7 epoxy-1,4,4a,5,6,7,8,8a- octahydro-1,4-endo-endo- 5,8-dimethanonaphthalene	7 - Cl ₂ Cl H H H H ₂	Multiple cancellations and restrictions. (see text)	Unnumbered PR no- tice May 20, 1964 44 FR 43632 July 25, 1979
1336-36-3	PCB-1260	Aroclor Clopen Phenoclor	pentachlorobiphenyl (approx. 60%)	4 2 2 3 4 4 5 6 6 5 4	Elimination (all use as active or inactive ingredients)	PR Notice 70-25 October 29, 1970

[†]EPA, 1979b. Acceptable Common Names and Chemical Names for the ingredient Statement on Pesticide Labels. EPA-54019-77-017. 247 pp. *Matsumura, F., 1985. Toxicology of insecticides. 2nd Ed., Plenum Press, N.Y. 598 pp.

^{††}EPA, 1985b. Suspended and Cancelled and Restricted Pesticides. 3rd rev. 29 pp.

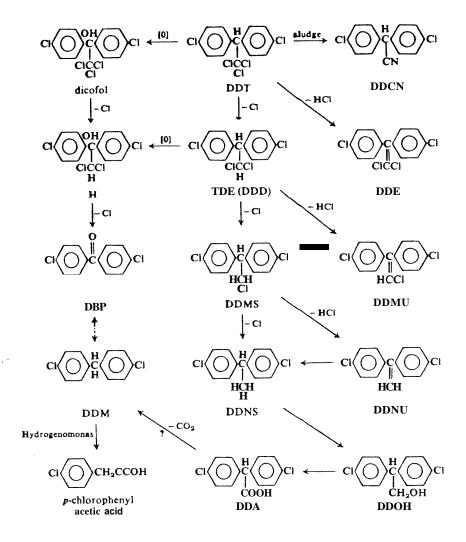


Figure 6. Microbial and environmental degradation of DDT (from Menzer and Nelson, 1980).

The predominant DDT compound in Yakima River fish is p,p'-DDE. On the average, p,p'-DDE constituted 84 percent of t-DDT in whole fish from the lower river at Buena and Kiona. Nationally, p,p'-DDE in whole fish averages 70 percent of t-DDT (Schmitt, et al., 1985). In the one instance where a DDT compound was detectable in water from the main stem of the Yakima River (Kiona), p,p'-DDE was the compound identified. These findings are consistent with a historical source.

However, analysis of water from Yakima River tributaries showed p,p'-DDT was present in concentrations equal to or greater than p,p'-DDE. The o,p'-isomer of DDT was at concentrations too low to quantify in water, but was quantified in sediment. Table 27 shows the relative percentages of DDT compounds and the p,p'-DDT:o,p'-DDT ratios found.

Table 27. Composition of t-DDT in bed sediment samples collected by Ecology from the Yakima River and tributaries on September 24 and 25, 1985.

		Perce	ent as		
	p,p'- DDE	p,p'- DDT	o,p'- DDT	p,p'- DDD	p,p'-DDT:o,p'-DDT
Yakima R. @ Cle Elum	100	0	0	0	not detected
Wilson Cr.	56	25	7.5	11	3.3:1
Yakima R. @ Rosa Dam	27	42	5.8	26	7.2: 1
Birchfield Drain	33	45	16	8.3	2.9: 1
Yakima R. @ Wapato Dam	70	12	2.2	16	5.3:1
Granger Drain	30	56	3.5	10	16: 1
Marion Drain	60	18	2.8	19	6.6: 1
Sulphur Cr.	55	25	3.6	16	7.0:1
11 ' 11	36	55	1.7	7.3	31: 1
Yakima R. @ Prosser Dam	62	17	4.2	17	4.1:1
11 11 11 11	63	16	6.8	18	2.4:1
Spring/Snipes Cr.	42	39	8.4	10	4.7: 1
Yakima R. @ Horn Rapids Dam	76	8.6	1.8	13	4.8:1

With the exception of one replicate sample at Sulphur Creek, sediment from tributaries known to be sources of DDT compounds had a percentage of p,p'-DDT equal to or greater than p,p'-DDE. The sample from the Yakima River behind Rosa Dam also had a high percentage of p,p'-DDT. p,p'-DDT:0,p'-DDT ratios in most of these samples were generally similar to the 5:1 ratio in technical DDT.

While some of these findings could be construed as evidence of recent DDT application in the Yakima basin, there is more support for the conclusion that the source is primarily historical. The occurrence of elevated p,p'-DDT in the absence of recent DDT use is not unique to the Yakima River. This phenomenon has been observed in fish from other Washington State rivers (Hopkins, et al., 1985); Puget Sound sediment (Konasewich, et al., 1982); Wisconsin streams (Zabik, 1985); New Mexico and Texas soils (Gillette, 1985; Hitch, 1985); and California fish, sediment, and soils (Riseborough, et al., 1984a,b,c; Agee, 1985, 1986; Mischke, et al., 1985).

Intensive investigation of this problem in California has led to the conclusion that DDT is being "preserved" in agricultural soils statewide (Agee, 1986; Mischke, et al., 1985). DDT's half-life in these soils appears to be at the upper end of the 4 - 30 year range reported in the literature (Edwards, 1973). Evidence for illegal use in California has not been found (Mischke, et al., 1985). Furthermore, the common occurrence of p,p'-DDT:o,p'-DDT ratios resembling technical DDT in agricultural soils, drain sediments, and suspended particulates was concluded to be the result of unexpected persistence of the o,p' isomer (Risebrough et al., 1984b). The scarcity of reports of o,p'-DDT in the literature was thought to be due to difficulty in distinguishing it from interfering compounds in the packed columns used in the past (Risebrough, et al., 1984b). Evidence of o,p'-DDT persistence after a 1965 forest application of DDT in Oregon (Tarrant, et al., 1972), in soil (Lichtenstein, et al., 1971), and in sea lion tissues (Riseborough, et al., 1984b) was cited in support of this conclusion.

Release of DDT to California surface waters appeared to be due to agricultural practices and rainfall runoff events. Once soils were disturbed and washed into drains, conversion of DDT mixtures resembling ≥ 60 percent technical DDT to mixtures resembling ≤ 45 percent technical DDT appeared to take place quickly; i.e., several months. Erosion control practices were recommended to reduce the problem (Agee, 1986).

While illegal use of DDT in the Yakima drainage cannot be completely ruled out, four lines of reasoning provide support for the conclusion that present-day levels of DDT contamination in the Yakima River are primarily the result of historical application coupled with long half-life and that t-DDT concentrations in the river have decreased since DDT was banned. These are as follows:

- 1. Illegal field use has not been detected WSDA and FDA routinely check pesticide applicators and monitor Pesticide residues on crops. Since the ban on DDT in 1972, no instance of illegal field use has been found in the Yakima drainage (Brown, 1986).
- DDT inputs to the Yakima River are not localized If DDT were used in 2. the Yakima basin, the practice would almost certainly be limited to a very few sites because (1) it is illegal and users face loss of crops (and associated revenues) if DDT residues are found by WSDA or FDA. (Brown, 1986), (2) it is not readily available, and (3) less persistent, more effective pesticides are now available. The results from Ecology's 1985 investigation, however, indicate widespread input of DDT to the Yakima River. Sediment samples from separate tributaries had comparable isomer distributions of DDT compounds. The one drainage which was intensively surveyed had a similar level of contamination along the length of the main drain and in each of the three major The distribution of DDT compounds in fish and sediment from laterals. the river showed readily detectable residues above tributaries identified as current sources.
- 3. Current levels of DDT compounds in Yakima River fish are not extremely high The preceding discussion of the significance of DDT compounds in Yakima River fish tissues (main stem, not tributaries) demonstrated

that levels are generally below those indicative of adverse impacts on aquatic life and wildlife. Concentrations of t-DDT in Yakima fish also do not appear extreme when viewed in a national perspective. The USFWS analyzed organochlorine pesticides and PCBs in whole fish from 106 rivers and lakes nation-wide in 1980-81 (Schmitt, 1985). The data on those contaminants identified in fish tissue during the present study are summarized in Table 28. Whole fish from the USFWS Yakima River station at Granger (r.m. 82.8) averaged 380 ug/Kg t-DDT, only slightly above the national mean of 290 ug/Kg. Thirty-one of the stations sampled (27 percent) had a mean t-DDT concentration higher than the Yakima. Results for dieldrin, endrin, and PCBs also show Yakima River fish to be at or below the national average. The USFWS did no analyses for endosulfan.

Table 28. USFWS data on organochlorine levels in freshwater fishes (whole fish) in 1980-81 showing relative contamination in the Yakima River (ug/Kg, wet).

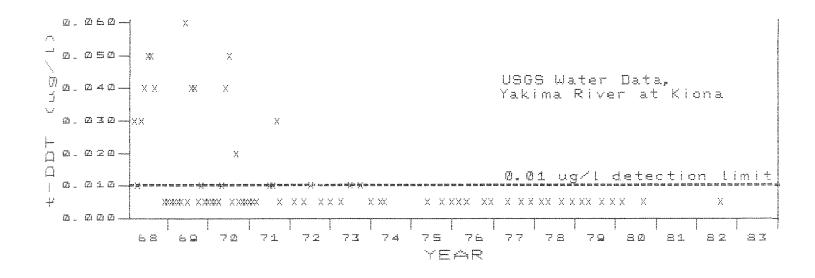
Com- pound	U.S. Maximum	U.S Mean (106 sta- tions)	Yakima R. @ Granger (mean of 3 samples)	Number of Stations Greater Than Yakima River
t-DDT	6,500 - Yazoo River Redwood, Mississippi	290	380	31 (27%)
Dieldrin	720 - Manoa Stream Honolulu, Hawaii	40	1 0u	280 (69%)
Endrin	300 - Mississippi River Memphis, Tennessee	10u	10u	225 (22%)
PCBs	11,300 - Hudson River Poughkeepsie, New Yor	530 k	100	77 (66%)

Source: Schmitt (1985).

u = Not detected at detection limit shown.

4. t-DDT levels in the Yakima River are decreasing, not increasing - The historical record on contamination by DDT in the Yakima River has been reconstructed in Figure 7. The long-term record on t-DDT in water is from samples collected at Kiona between 1968 and 1982 by the U.S. Geological Survey (USGS). This is the same station sampled during the present study. The longest continuous record on DDT compounds in Yakima River fish is the one shown for USFWS whole fish samples collected since 1970 at Granger (both of these data sets are in Appendices V and VI).

During the period 1968-1970, concentrations of t-DDT in water from the lower Yakima River were as high as 0.04 - 0.06 ug/L (similar to concentrations now observed in some Yakima trībutaries). A gradual decline in river concentrations occurred between 1970 and 1974. A marked annual



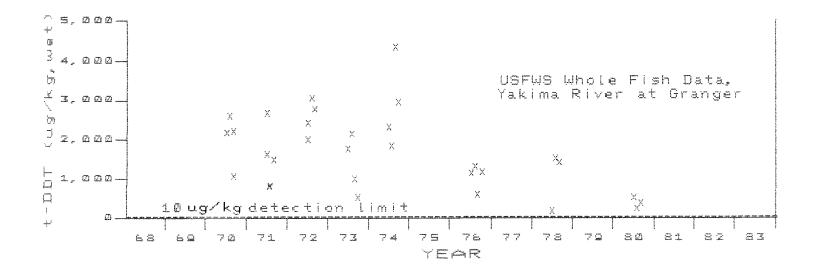


Figure 7. Historical record on t-DDT in water and fish from the Yakima River. (Water data from Schultz [1973] and USGS Water Data Reports for water years 1968-1982; fish data from Schmitt, et al. [1981 1983, 1985]. Non-detections plotted at one-half the detection limit.)

cycle is evident during 1968 - 1973 (more easily seen in Appendix V) in that DDT compounds were detectable only during spring through fall.DDT compounds remained undetectable (0.01 ug/L detection limit) in all USGS samples collected between 1974 and 1982. USGS discontinued pesticides analysis after 1982. As discussed earlier, only one of four water samples collected at Kiona during the present study showed detectable DDT compounds. P,p'-DDE was detected at an estimated concentration of 0.006 - 0.007 ug/L, approximately one-tenth of peak t-DDT concentrations observed fifteen years ago.

The decline in DDT residues shown for Yakima River fish at Granger follows that in water. Mean concentrations of t-DDT in whole fish between 1970 ~ 1974 ranged from 1,400 to 2,600 ug/Kg. Concentrations declined to a median of 390 ug/Kg in 1980, comparable to the median of 490 ug/Kg measured in whole fish eleven miles upstream at Buena during the present study. Analysis of covariance was used to test the significance of observed changes in lipid-normalized t-DDT concentrations between 1974 and 1980. The trend toward decreasing t-DDT concentrations in fish was significant at the p=0.05 level (Schmitt, et al., 1985).

The miticide dicofol (1,1-bis[chlorophenyl]-2,2,2-trichloroethanol; trade name "Kelthane") has been suggested as a current source of DDT (Graham, 1984). Dicofol is manufactured from DDT by dehydrochlorination to form DDE, chlorination to form Cl-DDT (extra chlorine DDT), and hydrolysis replacing the extra chlorine by a hydroxyl (-0H) group. Impurities can include o,p' and p,p' isomers of DDT, DDE, DDD, and Cl-DDT. DDE and Cl-DDT are the predominant impurities. The only U.S. manufacturer, Rohm and Haas, reported that DDT-related impurities averaged 5.75 percent in 1984. This was reduced to 2.5 percent in 1985. The company has pledged to further reduce DDT-related impurities to 0.1 percent by January 1, 1987 (Ellenberger, 1985). Dicofol itself readily metabolizes and degrades to dichlorobenzylic acid and dichlorobenzophenone; DDT, DDE, or DDD are not breakdown products (Gillette, 1985).

The major use of dicofol is control of mites in citrus and cotton. The states of California and Florida have investigated dicofol as a possible contributor to environmental levels of DDT compounds (Riseborough, et al., 1984; Mischke, et al., 1985; Agee, 1985; Pickerell, et al., 1984). In spite of large-scale application (annually approximately 1.5 million pounds in California and 200,000 pounds in Florida), the unanimous conclusion was that the level of impurities was insignificant compared to residues from historical application.

While data on the amount of dicofol used in Washington is not available, **it** is not used extensively (Maxwell, 1986). In the Yakima basin, application is approved for hops (new plantings only), cherries, and pears. Over the past ten years, there has been little or no use o dicofol because target pests had become resistant. There has been a slight increase in use over the past two years, but resistance to dicofol continues to limit applications (Brown, 1986).

Inasmuch as dicofol has a low level of DDT-related impurities; does not break down to give DDT, DDE, or DDD; and is used in small amounts in the Yakima basin, it is unlikely that its use has contributed significantly to contamination by DDT compounds in the Yakima River.

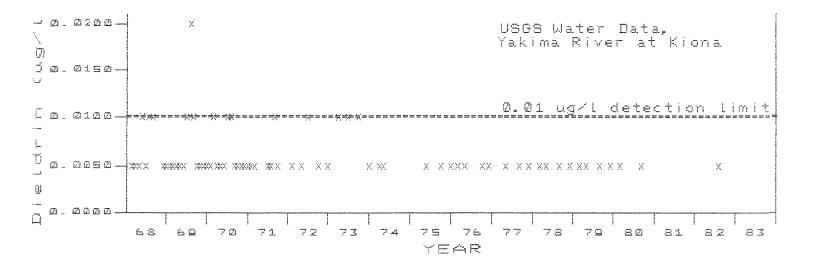
Results from the present study show that dieldrin (severely restricted use) and PCB-1260 (new use eliminated) are widespread low-level contaminants of the Yakima system. Dieldrin is one of the most commonly detected organochlorine pesticides in U.S. surface waters (Gilliom, 1985); polychlorinated biphenyls are among the most widely distributed pollutants on earth (Konasewich, et al., 1982). With the exception of aldrin's detection in Spring/Snipes Creek, there was no evidence of fresh inputs of either compound to the river.

USGS and USFWS historical data are also available on dieldrin in Yakima River water and fish. This information is plotted in Figure 8. The decrease in dieldrin concentrations parallels that observed for DDT compounds. An annual cycle in occurrence is also evident for dieldrin.

Aldrin was withdrawn from agricultural use about 12 years ago. The previously mentioned detection of a high aldrin concentration in Spring/Snipes Creek sediment is unusual because aldrin is rapidly metabolized to its epoxide, dieldrin, in the environment. Aldrin's half-life in soil has been calculated at 0.3 year, with 95 percent disappearance ranging between 1 to 6 years and averaging 3 years (Edwards, 1973). Present-day allowed uses are subsurface insertion for termite control, dipping non-food roots, and mothproofing by manufacturing process in closed systems (EPA, 1985b). The presence of aldrin in sediment could be the result of illegal use, improper disposal of a container(~),or perhaps, disturbance of subsurface soils containing high concentrations from historical application.

The use of the two remaining compounds, endosulfan and endrin, is still allowed in agriculture. Endosulfan is applied to a variety of crops in the Yakima basin, including grapes, potatoes, tree fruits, corn, small grains, alfalfa, and grass (Willet, 1985). Its occurrence in the water samples collected in Birchfield Drain during the present study is probably due to its use as a cover spray on orchards (Faulconer, 1986).

The use of endrin, a rodenticide, has been severely curtailed in Washington. Prior to 1983, there were no legal restrictions on its application. Endrin application now must be for crisis situations only and approved by WSDA (Toohey, 1986). In 1984, one approved application of 30 gallons occurred, this in the Wapato area (Faulconer, 1986). No applications occurred in the Yakima basin in 1985. The fact that endrin (or its metabolite endrin aldehyde) was detected at low levels in only three of the 46 fish tissue samples analyzed in the present study and was never detected in water or sediment, suggests that residues from past use do not pose a threat to the Yakima River aquatic environment.



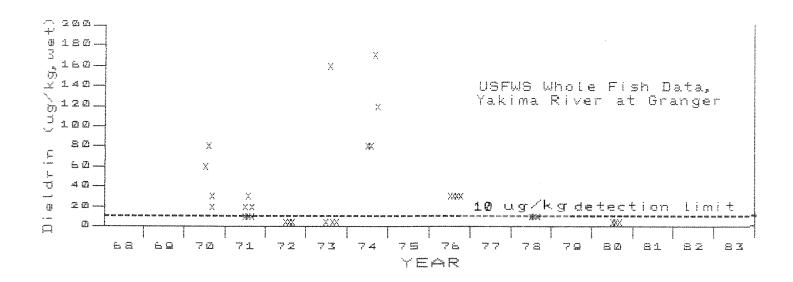


Figure 8. Historical record on dieldrin in water and fish from the Yakima River. (Water data from Schultz [1973] and USGS Water Data Reports for water years 1968-1982; fish data from Schmitt, et al. [1981, 1983, 1985]. Non-detections plotted at one-half the detection limit.)

RECOMMENDATIONS

1. Results of the present study and USGS historical data show that transport of the organochlorine pesticides DDT (and metabolites) and dieldrin to the Yakima River occurs primarily during the irrigation season. Tributaries to the Yakima identified in the present study as predominant sources were Sulfur Creek, Birchfield Drain, Granger Drain, and Spring/Snipes Creek. Concentrations measured in these tributaries may be high enough to pose a threat to aquatic life and make them unfit sources of fish for consumption by humans and wildlife.

DDT compounds and dieldrin have low water solubilities (<1 ppm) and tend to be associated with particulate matter suspended in the water column. Eighty percent or more of the DDT and dieldrin in agricultural runoff is associated with the particulate phase (Wauchope, 1978). For this reason, erosion control is an effective means of reducing the transport of organochlorines from fields to irrigation returns and subsequently to the Yakima River.

Ecology should work with the Soil Conservation Service, soil conservation districts, irrigation districts, and farmers to design, fund, and implement a plan to reduce erosion, focusing initially on the **tri**butary drainages identified above. This would reduce oranochlorine loads to the Yakima and generate concomitant water quality improvements: reductions in suspended solids, turbidity, and phosphates.

- 2. Concentrations of two currently used pesticides, endosulfan and diazinon, approached or exceeded acutely toxic levels in Birchfield Drain. Endosulfan was detected in Yakima River fish and sediment downstream of Birchfield Drain. Diazinon has been previously detected in the discharge from this drain. Ecology should work with Department of Agriculture inspectors, applicators, and farmers to reduce or eliminate the discharge of these compounds, focusing initially on the Birchfield drainage area.
- 3. Additional bed sediment samples should be collected in Spring/Snipes Creek to verify the presence of high aldrin concentrations (or high levels of its metabolite dieldrin).
- 4. The potential for sublethal adverse effects from DDT compounds and dieldrin on fish from the lower Yakima River should be evaluated. One promising approach for evaluating these effects is the histopathological examination of resident fish (especially bottom-living species) to determine the types and incidence of abnormalities (i.e., lesions). (Ecology is planning this type of survey at Kiona in July 1986.)
- 5. The toxicity of bed sediments was not evaluated in the present study. Reduction of benthic invertebrate populations could have a negative impact on salmonid and other fisheries resources. Selected sediment samples should be screened for toxicity by bioassay.

6. The Columbia River pool behind McNary Dam (Lake Wallula) has been the ultimate sink for Yakima River sediment since the 1940s. Results of the only relevant study known to have been done showed that DDE and PCBs in Columbia River white sturgeon peak in Lake Wallula. Two of the three muscle samples collected there exceeded FDA criteria for t-DDT and PCBs (Bosley and Gately, 1981). For these reasons it is recommended that Lake Wallula be included as one of the stations in Ecology's 1987 BWMP fish collections, with subsequent tissue analysis for a range of pesticides and PCBs.

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APPENDICES

Appendix I. Cleaning procedures for containers used in Ecology's 1985 study of DDT and other contaminants in the Yakima River.

Organochlorines

1. Rinse with pesticide-grade methylene chloride, and drain.

Herbicides, Acid/Base-Neutrals

- 1. Wash containers, closures, and teflon liners in hot tap water with laboratory-grade, non-phosphate detergent.
- 2. Rinse three times with tap water.
- 3. Rinse with 1:1 nitric acid.
- 4. Rinse three times with ASTM Type 1 de-ionized water.
- 5. Rinse with pesticide-grade methylene chloride.
- 6. Oven dry.
- 7. Remove containers, closures, and teflon liners from oven.
- 8. Place teflon liners in closures and place closures on containers. Attendant to wear gloves and containers not to be removed from preparation room until sealed.

Volatiles, Total Organic Carbon

- 1. Wash vials, septa, and closures in hot tap water with laboratory-grade, non-phosphate detergent.
- 2. Rinse three times with tap water.
- 3. Rinse three times with ASTM Type 1 de-ionized water.
- 4. Oven dry vials, septa, and closures.
- 5. Remove vials, septa, and closures from oven.
- 6. Place septa in closures, teflon sidedown, and placeonvials. Attendant to wear gloves and vials not to be removed from preparation room until sealed.

Mercury

Bottles

- **1.** Wash in hot, soapy water.
- 2. Rinse with high-purity water.
- 3. Rinse with concentrated HNO3.
- 4. Rinse several times with high-purity water.
- 5. Bake in oven at 450°C for 8 hours.

Caps and Liners

- 1. Remove teflon liners.
- 2. Wash both caps and liners in hot, soapy water.
- 3. Rinse with high-purity water.
- 4. Rinse teflon liners with concentrated HNO3.
- 5. Rinse with high-purity water.

Other Metals

- 1. Fill with 1 + 1 HCl (AR grade).
- 2.. Allow to stand one week at room temperature.
- 3. Empty and rinsewith distilled water.
- 4. Fill with $1 + 1 \text{ HNO}_3$ (AR grade).
- 5. Allow to stand one week at room temperature.
- 6. Empty and rinse with distilled water.
- 7. Fill with de-ionized water.
- 8. Allow to stand several weeks or until needed, changing water periodically to ensure continued cleaning.

Appendix II. CNFRL methodology for organochlorine pesticides/PCBs analysis of fish tissue samples.

1.0 Introduction

The 27 organochlorine contaminants (including organochlorine pesticides, other organochlorine contaminants, and PCB Aroclor mixtures) that are to be measured are listed in Attachment A. Attachment B summarizes steps used to separate and measure these residues in tissue (plant and animal) samples. Organic contaminants will be extracted from tissue with a solvent and separated from lipids by gel permeation chromatography. Residue extracts will be fractionated into resolvable mixtures by adsorption chromatography (Florisiland Silica gel). Component mixtures will then be analyzed by gas chromatography, and at our request, some (as much as 10 percent for sample batches) of the sample set will be confirmed by GCMS.

2.0 Sample Preparation

2.1 Sample Homogenation

Samples should be homogenized so that replicate aliquot element concentrations are within ±10 percent standard deviation.

2.2 Moisture Determination

Percent moisture content in homogenized tissue samples will be determined with a C.W. Brabender Moisture and Volatiles tester or equivalent.

2.3 Extraction of Tissue

A modified Hesselberg and Johnson (1972) method or equivalent will be used for extraction. Ground tissue (10.0 g) will be mixed with anhydrous Na₂SO₄ (40 g) and placed in a glass extraction column over a pledget of glass wool and Na₂SO₄ (1-2 cm-layer). Another layer of Na₂SO₄ will be placed on top and methylene chloride (200 mL) will be added to the column fo extraction. The solvent flow rate will then be adjusted to 3-5 mL/min by manipulating the stopcock at the lower top of the tissue and methylene chloride (MeCl₂) end of the column. The eluate from the column extraction will be collected in a 250 mL double reservoir flask (or equivalent) designed for efficient solvent removal (May and Stalling, 1979). After extraction is complete, MeCl₂ will be stripped from the lipid by rotoevaporation at ambient temperature (to minimize losses of volatile constituents). After solvent removal, the double reservoir vessel walls will be rinsed with methylene chloride:cyclohexane (1:1) and the lipid concentrate diluted to 10 mL in the calibrated small reservoir of the collection flask. Measured portions of the "lipid" extract will be used for GPC fractionation and lipid determination.

2.4 Lipid Analysis

An aliquot of the "lipid" extract used for GPC will be transferred to an accurately weighed vial. The solvent will be slowly evaporated overnight at 90°C (using a heating block), and the vial will then be reweighed for lipid determination.

2.5 Gel Permeation Chromatography (GPC)

Lipid materials will be removed from organic residues by size fractionation using an automated gel permeation system (GPC Autoprep 1001, equipped with a column of SX-3 Biobeads, 200/400 mesh) (Stalling, et al., 1972; Anoop and Tucker, 1977; or equivalent). The GPC column must be characterized before use to establish the relative elution patterns of lipid materials and neutral pesticides. Neutral residues will be eluted into double reservoir flasks (May and Stalling, 1979; or equivalent) with In methylene chloride:cyclohexane, concentrated by rotoevaporation, and brought up to final volume with hexane.

2.6 Florisil Fractionation

Florisil fractionation will be done by the method of Ribick, et al. (1981) or equivalent. Florisi (Fisher, 60-100 mesh or equivalent) will be activated at 135°C until columns are preapred. Five-gram columns will be dry-packed and immediately rinsed with 20 mL hexane, and then the concentrated CPC eluates (in hexane) will be transfered to the columns. (Florisi columns must be periodically characterized to establish relative elution patterns of non-polar and polar compounds; oven temperature, room humidity, and batch-to-batch solvent variations may greatly affect the elution behavior.) Non-polar and moderately polar residues will be eluted with 40 mL of five percent diethyl ether in hexane (5 percent fraction). More polar compounds, including dieldrin, endrin, and phthalates, will typically be eluted with 38 mL or 40 percent diethyl ether in petroleum ether (40 percent fraction).

Both fractions will be collected in 125 mL double reservoir flasks or equivalent. One to two milliliters of isooctane will be added to each flask to prevent loss of volatile pesticides, and ethers will be removed by rotoevaporation. The 40 percent fraction will be brought to 5 mL with isooctane for gas chromatographic analysis, and the 5 percent faction (or a measured aliquot) will be transferred to silica gel for further fractionation.

2.7 Silica Gel Fractionation

Silica gel fractionation will be done by the method of Ribick, et al. (1981) or equivalent. Silica gel (Silica Gel 60, 70-230 mesh

ASTM, E. Merck or equivalent) will be activated at 135°C until column preparation. Five-gram silica gel columns will be dry-packed just before use and immediately rinsed with 20 mL hexane. Relative elution patterns must be characterized periodically, as for Florisil. The five-percent fractions (or a measured aliquot) will be transferred to the columns. The PCB fraction (including Aroclor mixtures, aldrin, heptachlor, HCB, mirex, and 80-90 percent of the DDE) will be eluted with 38 mL 0.5 percent benzene in hexane. The pesticide fraction (Pesticide Fraction, including the remaining DDE and other residues) will be eluted with 35 mL 25-percent diethyl ether in hexane. Both eluates will be concentrated to 1-2 mL by rotoevaporation (after the addition of isoocane), diluted to 5 mL with isooctane, and transferred to culture tubes for gas chromatographic analysis.

- 3.0 Sample Analysis--Gas chromatography with Electron Capture Fraction
- 3.1 Packed Column Gas Chromatographic Analysis
- 3.1.1 Forty Percent Fraction from Florisil and Pesticide Fraction from Silica Gel-Column:

Typical conditions (U.S. EPA Method 608 or equivalent) for gas chromatographic analysis are:

GC column: 6 foot x 2 mm I.D. glass containing 1.5 percent

SP 2250+1.95 percent SP 2401 on Supelcoport

(100/120 mesh) or equivalent

Detector: 63Ni electron capture detector equipped with a

linearizer

Flow conditions: 30 mL/min of ultrapure N_2

Column temperature: isothermal at 180-200°C

Injector temperature: 220°C

Detector temperature: 350°C

Identification and quantification of contaminant residues in these fractions are based on retention times and peak magnitude (area or height) of external standard compounds.

3.1.2 PCB Fraction from Silica Gel Column:

Typical conditions (Webb and McCall, 1973, or equivalent) for gas chromatographic anatysis are:

Column: 6 foot x 2 mm I.D. glass containing 3 percent SE-30

on 801100 mesh GAS Chrom 0 (or equivalent)

Detector: 63Ni electron capture detector equipped with a

linearizer

Flow conditions: 30 mL/min of ultrapure N₂

Column temperature: isothermal at 190-200°C

Injector temperature: 220°C

Detector temperature: 350°C

Packed column identification and quantitation of PCB Aroclor residues are based on relative retention times vs. p,p'-DDE and peak magnitude (area or height) and response factors for external standard Aroclor 1242, 1248, 1254, and 1250 compounds as described by Webb and McCall (1973) or equivalent.

- 3.2 High-Resolution Capillary Gas Chromatographic Analysis
- 3.2.1 Typical conditions (Ribick, et al., 1982) for gas chromatographic analysis are:
- 3.2.1.1 Forty percent fraction from florisil and pesticide fractions from silica gel--

Column: 24M OV 17 fused silica capillary, 0.25 mm I.D., 0.25 uM film thickness or equivalent.

3.2.1.2 PCB fraction from silica gel--

Column: 29M Se30 fused silica capillary, 0.25 mm I.D., 0.25 uM film thickness or equivalent.

It is strongly recommended that the bonded or immobilized versions of these above-mentioned coatings be used.

Both capillary injection systems are split/splitless types, operated in the splitless mode with 1.25 min or 1.0 min, split vent delay for OV 17" or SE 30, respectively. Septum purge flow rate is 4 mL/min.

Splitvent flow rate is 20 mL min.

Column flow rates as linear velocity:

OV 1/ - 100 cm/sec, H_2 SE 30 - 85 cm/sec, H_2

Column temperature program: 70°C/0 min/5°C/min/220°C/9 min.

Injector temperature: 220°C

Detector temperature: 300°C

Det flow: N₂ make-up + H₂

Carrier gas flow:

0V 17--25 mL/min SE 38-25 mL/min

Detector: 63Ni electron capture with linearizer

3.2.2 Capillary Quantitation of 40 Percent Fraction and Pesticide Fraction

Identification and quantitation of contaminant residues and toxaphene components are based on relative retention times vs. aldrin and peak magnitude (area or height) of external standard compounds. Confirmation analyses with alternate liquid phases (e.g., DE-5, DB1701) may be requested on some of the samples. Further confirmation and/or identification of unknown compounds may be requested for up to 10 percent of the samples in some batches by high-resolution capillary GCMS.

3.2.3 Capillary GC Quantitation of PCBs

Capillary column identification of PCB Aroclor residues is based on their relative retention versus aldrin or p,p'-DDE (aldrin or p,p'-DDE is added to each sample prior to capillary analysis).

3.2.3.1 Quantitation of Aroclors 1242, 1249, 1254, and 1260

For quantitation of Aroclors, 10 to 15 chromatographic peaks are selected as representative of each aroclor region of the chromatogram. The relative retention times of peaks are stored in three data calculations methods: 48, 54, and 60. Using Basic Program ARO (or equivalent), the three Aroclor standards, their asociated calculation methods, and an appropriate reference peak (aldrin or p,p'-DDE), the Basic Language program solves a system of three linear equations in concentration to obtain R1248, R1254, R1260, R'1249, . . R"1260. The three equations are:

- 1. A1248 = R1248 \times [C1248] + R1254 \times [C1254] + R1260 \times [C1260]
- 2. A1254 = R'1248x[C1248] + R'1254x[C1254] + R1260x[C1260]
- 3. A1260 = R"1248x[C1248] + R"1254x[C1254] + R"1260x[C1280]

where A1248 is the total area for all components (peaks) in the 1248 region taken from a standard or sample chromatogram, A1254 is the total area for all components in the 1254 region taken from a standard to sample chromatogram, A1260 is the total area for all components in the 1260 region taken from a standard or sample chromatogram.

R1248 = A1248/concentration of 1248 standard, R1254 = A1248/concentration of 1254 standard, R1260 = A1248/concentration of 1260 standard, R'1248 = A1254/concentration of 1248 standard, etc. C1248 is the concentration of PCB 1248; C1254 is the concentration

C1248 is the concentration of PCB 1248; C1254 is the concentration of PCB 1254; and C1260 is the concentration of PCB 1260.

Using the nine R-values (R1249 to R"1260) calculated from the three standards' chromatograms, the Basic program then calculates the relative concentrations (amounts) of Aorclor 1248, Aroclor 1254, and Aroclor 1260 components in the PCB fraction samples. When Aroclor 1242-type components appear to be present, a more rigorous calculation may be made by including Aroclor 1242 standards in this calculation. In this case, an Aroclor 1242 standard may be substituted for Aroclor 1248 standard in the calculation.

This method of calculation assumes an "average" R-value (i.e., R1248 is assumed to be the same for all "1248-like" components in the 1248 region regardless of the number of chlorine atoms attached to the biphenyl molecule) and therefore does not assign specific R-values for individual isomer components. Although the resolution achieved by the SE30 capillary column is probably adequate for most isomer

assignments, the calibration of the individual R-values may not be cost-effective for some studies. Because these "average" R-values are used, the PCB concentration is usually underestimated when 1242 is present due to the higher incidence of 2 and 3 chlorine substituted biphenyls. In the same context, the PCB concentration of an environmental sample will also be underestimated if the lower chlorine-substituted isomers within a given Aroclor region are at the highest concentration.

3.2.3.2 Quantitation of Individual PCB Isomers

The contractor must also be able to identify individual PCB isomers, the corresponding total PCB concentration and Aroclor mixture proportions valid retention indices versus added n-alkyl trichloroacetates (Schwartz and Petty, 1983; Schwartz, et al., 1984), and SIMCA pattern recognition techniques (Dunn, et al., 1984).

4.0 Quality Control

4.1 Blank

An organic method blank extracted with the fish samples will be subjected to cleanup by florisil and silica gel. Any interferences found will then be traced to the problematic solvent/column system.

4.2 Determination of Elution Profile

Optimum elution volumes must be experimentally determined. It is advisable to chromatograph a set of standards prior to each batch of samples to monitor any changes in elution patterns. The data obtained in analysis of individual eluates for determination of the elution profile will be included in the final report.

4.3 Sample Spikes

Spikes will be added to uncontaminated tissue at the beginning of the column extraction step. Recoveries of these compounds will reflect losses throughout the entire cleanup/determination process. Recoveries should be within ±2 standard deviations of the established mean. If a recovery is out of control, the possible cause will be investigated and corrected or the data will be flagged as possibly biased.

5.0 Reporting

The final report will include: all sample residue concentration data; quality control recovery results; methods and instrumental parameters writeup; and when isomer-specific determination methods have been requested; the retention indices or equivalent data used to identify individual PCB components. Additionally, all raw data including preparation of standards, sample worksheets, and raw chromatographic data will be provided in its original form.

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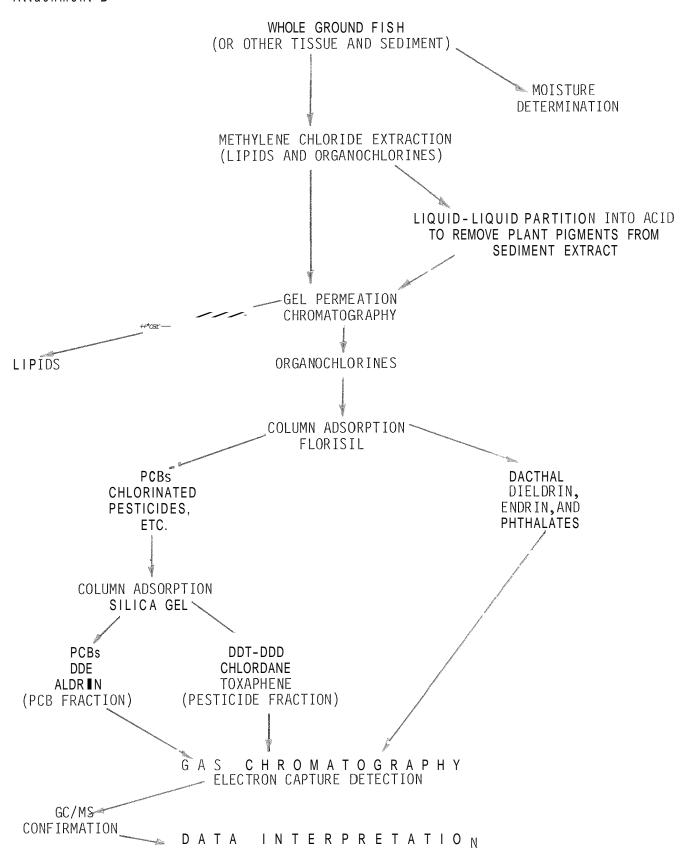
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Attachment A. Organic compounds analyzed by CNFRL Methodology

Dacthal Dieldrin cis-Chlordane trans-Chlordane p,p'-DDT p,p'-DDE p,p'-DDD Endrin Endrin aldehyde Heptachlor	Endosulfan I/II Alpha-BHC Beta-BHC Gamma-BHC Delta-BHC Methoxychlor Mirex cis-Nonachlor trans-Nonachlor Oxychlordane	o,p'-DDT o,p'-DDE o,p'-DDD Pentachloroanisole PCB-1242 PCB-1248 PCB-1254 PCB-1260
Heptachlor epoxide	Tox aphene	

Attachment B



Results of organochlorine analysis on bed sediment samples collected by Ecology in Birchfield Drain on March 25, 1985 (ug/Kg, dry; detected compounds underlined). Appendix III.

	Sample	Per- cent Drv	Percent Total Organic	- 1 Q . Q	- , d · 0	-10.0	1	1	1	Percellance of commence and com	- hule
Sample Loction	Number	Weight	Carbon	DDE	DDE	DDT	TO TO			t-DDT	BHC
Birchfield Drain above Rosa Canal	7012	74.3	0.7	10	1u	21	Jn	17	1n	32	7
Birchfield Drain below Rosa Canal	7013	77.2	1.0	ای	n T	12	٦ ۲	17	<u>n</u>		7
Birchfield Drain at Faucher Road	7014	77.5	0.2	41	Ę	mΙ	Ju	11	1n	∞I	17
Birchfield Drain at Beauchene Road	7015	6.97	0.7	4	Ju	12	1n	1u	Ju	61	1u
Hubbard Ditch "	7016 7017	60.5 59.3	2.0	65 40	mml	12 41	ח ח	22	크급	102 93	lu lu
Union Gap Canal	7018	96.4	0.4	10	17	61	11	က	lu	24	Ju
Birchfield Drain above Postma Confluence	7019	76.03	8.0	13	Ju	91	Ju	۳I	1n	22	Ju
	7024	73.7	1.0	13	17		2	13	Ju	36	Ju
Postma Ditch at Birchfield Road	7023	8.69	0.2	14	7	26	7	18	4	71	Ju
Postma Ditch at Birchfield Confluence)21	65.7	8.0	14	17	41	1n	41	1n	23	1n
Birchfield Drain Mouth	7022	62.3	0.9	18	17	4	1u	4	1u	27	1u

NOTE: See Table 5 Part A for detection limits of organochlorines analyzed but not detected, $u = Not\ detected\ at\ detection\ limit\ shown.$ J = Estimated concentration.

Appendix IV. Results of organochlorine analysis on water samples collected by Ecology in Birchfield Drain on July 2 and August 21, 1985 (ug/L; detected compounds underlined).

Sample Loction	Sample Number	Flow (cfs)	Sp. Cond. (umhos/cm)	Total Susp. Solids (mg/L)	Total Organic Carbon (mg/L)	p,p'-DDE	p,p'-DDT	p,p'-DDD	t-DDT	Endo- sulfan
				Jul	y 2					
Birchfield Drain above Rosa Canal	7296	3.1	278	210	5.3	0.01u	0.02u	0.02u		0.02
Birchfield Drain @ Beauchene Road	7298	not gaged	323	500	8.0	0.03	0.02	0.02u	0.05	0.01
H	7299		325	470	6.9	0.03	0.02	0.02u	0.05	0.02
Hubbard Ditch	7300	26	192	190	4.1	0.01	0.02u	0.02u	0.01	0.01
Union Gap Canal	7302	38 est.	83	7	3.2	0.02	0_04	0.02u	0.06	0.05
Birchfield Drain @ Birchfield Road	7303	64	249	380	5.1	0.02	0.04	0.01	0.07	0.03
Postma Ditch @ Birchfield Road	7304	5	382	13	4.2	0.01u	0.02u	0.02u		0.01u
				Aug	ust 21					
Birchfield Drain above Rosa Canal	8040	8.6	186	36	4.6	0.02u	0.04u	0.04u		0.01u
Birchfield Drain below Rosa Canal	8041	19	237	73	4.1	0.01u	0.02u	0.02u		0.01u
Birchfield Drain @ Faucher Road	8042	35	232	160	3.9	0.01u	0.02u	0.02u		0.01u
Birchfield Drain @ Beauchene Road	8043	not 9 aged	226	230	8.0	0.01u	0.02u	0.02u		0.01u
н	8044		226	250	6.9	0.01u	0.02u	0.02u		0.01u
Hubbard Ditch	8045	13	209	78	6.2	0.01u	0.02u	0.02u		0.01u
Union Gap Canal	8046 8047	49.3	68 69	15 19	2.2 2.2	0.01u 0.01u	0.02u 0.02u	0.02u 0.02u		0.01u 0.01u
Birchfield Drain @ Birchfield Road	8048	106	209	140	7.2	0.01u	0.02u	0.02u		0.01u
Postma Ditch @ Birchfield Road	8049	6.2	349 °	52	5.4	0.01u	0.02u	0.02u		0.01u
Birchfield Drain Mouth	8050	112	233	160	4.7	0.01	0.02	0.02u	0.03	0.02

NOTE: See Table 5 Part A for detection limits of organochlorines analyzed but not detected. u = Not detected at detection limit shown.

Appendix V. USGS data on DDT compounds and dieldrin in Yakima River water collected at Kiona (station 12510500) 1968 - 1982 (ug/L; detected compounds underlined).

Date	p,p'-DDE	p,p'-DDT	p,p'-DDD	t-DDT	Dieldrin
02/24/68 03/28/68 04/24/68 05/29/68 06/27/68 07/18/68 08/26/68 11/25/68 12/16/68	0.01 0.01u 0.01u 0.01 0.02 0.01 0.01 0.01u 0.01u	0.01 0.01 0.03 0.03 0.02 0.02 0.02 0.01 0.01u	0.01 0.01u 0.01u 0.01u 0.01 0.02 0.01 0.01u 0.01u	0.03 0.01 0.03 0.04 0.05 0.05 0.04	0.01u 0.01u 0.01u 0.01 0.01u 0.01 0.01u 0.01u 0.01u 0.01u
01/20/69 02/25/69 03/26/69 04/09/69 05/19/69 06/27/69 07/30/69 08/31/69 09/09/69 10/17/69 11/25/69 12/30/69	0.01u 0.01u 0.01u 0.01u 0.01 0.01 0.01u 0.01u 0.01u 0.01u 0.01u 0.01u 0.01u 0.01u	0.01u 0.01u 0.01u 0.01u 0.04 0.04 0.03 0.02 0.01u 0.01 0.01u 0.01u	0.01u 0.01u 0.01u 0.01u 0.01 0.01u 0.01 0.01	0.06 0.05 0.04 0.04 0.01	0.01u 0.01u 0.01u 0.01u 0.01u 0.01 0.02 0.01 0.01u 0.01u 0.01u 0.01u
01/27/70 02/16/70 03/19/70 04/28/70 05/25/70 06/22/70 07/20/70 08/24/70 09/30/70 10/15/70 11/24/70 12/29/70	0.01u 0.01u 0.01u 0.01u 0.01 0.01 0.01u 0.01u 0.01u 0.01u 0.01u 0.01u 0.01u	0.01u 0.01u 0.01u 0.01 0.02 0.02 0.01u 0.01u 0.01u 0.01u 0.01u 0.01u 0.01u 0.01u	0.01u 0.01u 0.01u 0.01u 0.01 0.02 0.01u 0.01 0.01u 0.01u 0.01u 0.01u 0.01u	0.01 0.04 0.05 0.02	0.01u 0.01 0.01u 0.01u 0.01u 0.01 0.01 0
01/22/71 02/03/71 06/21/71 07/16/71 08/06/71 09/23/71	0.01u 0.01u 0.01u 0.01u 0.02 0.01u	0.01u 0.01u 0.01 0.01 0.01 0.01u	0.01u 0.01u 0.01u 0.01u 0.01u 0.01u	0.01 0.01 0.03	0.01u 0.01u 0.01u 0.01u 0.01 0.01
01/19/72 04/03/72 06/30/72 09/08/72 12/18/72	0.01u 0.01u <u>0.01</u> 0.01u	0.01u 0.01u 0.01u 0.01u	0.01u 0.01u 0.01u 0.01u 0.01u	n n1	0.01u 0.01u 0.01

Appendix V - continued.

Date	p,p'-DDE	p,p'-DDT	p,p'-DDD	t-DDT	Dieldrin
03/30/73 06/01/73 09/21/73 12/27/73	0.01u 0.01u 0.01u 0.01u	0.01u 0.01 0.01 0.01u	0.01u 0.01u 0.01u 0.01u	0.01 0.01	0.01 0.01 0.01 0.01u
03/12/74	0.01u	0.01u	0.01u	100 100	0.01u
04/08/74	0.01u	0.01u	0.01u	100 100	0.01u
05/22/75	0.01u	0.01u	0.01u	0.01u	0.01u
09/16/75	0.01u	0.01u	0.01u	0.01u	0.01u
12/17/75	0.01u	0.01u	0.01u	0.01u	0.01u
02/13/76	0.01u	0.01u	0.01u	0.01u	0.01u
04/11/76	0.01u	0.01u	0.01u	0.01u	0.01u
09/21/76	0.01u	0.01u	0.01u	0.01u	0.01u
11/22/76	0.01u	0.01u	0.01u	0.01u	0.01u
04/02/77	0.01u	0.01u	0.01u	0.01u	0.01u
08/03/77	0.01u	0.01u	0.01u	0.01u	0.01u
11/08/77	0.01u	0.01u	0.01u	0.01u	0.01u
02/08/78	0.01u	0.01u	0.01u	0.01u	0.01u
04/17/78	0.01u	0.01u	0.01u	0.01u	0.01u
08/08/78	0.01u	0.01u	0.01u	0.01u	0.01u
11/06/78	0.01u	0.01u	0.01u	0.01u	0.01u
02/20/79	0.01u	0.01u	0.01u	0.01u	0.01u
04/07/79	0.01u	0.01u	0.01u	0.01u	0.01u
08/06/79	0.01u	0.01u	0.01u	0.01u	0.01u
11/06/79	0.01u	0.01u	0.01u	0.01u	0.01u
02/03/80 08/11/80	0.01u 0.01u	0.01u 0.01u	0 0111	0.01u 0.01u	Λ Λ1 ₁₁
07/12/82	0.01u	0.01u	0.01u	0.01u	0.01u

Source: Schulz (1973) and USGS $\underline{\text{Water Data Reports}}$ for water years u = Not detected at detection limit shown.

Appendix VI. USFWS data on DDT compounds and dieldrin in whole fish samples collected from the Yakima River at Granger, 1970-1980 (ug/Kg, wet).

				Commence of the second		5×-0	
Species	Date	% Lipid	p,p'-DDE	p,p'-DDT	p,p'-DDD	t-DDT	Dieldrin
Bridgelip sucker	1980	3.8 2.0	320 200	140 10u	70 30	530 230	10u 10u
Black crappie	11	2.5	350	10u	40	390	10u
White crappie Carp	1978	3.1 4.7	10 1240	50 10u	110 280	170 1520	10 10
11	1 E	5.3	1190	10	230	1430	10
Largescale sucker	1976	6.9 6.4	810 1100	40 10u	280 220	1130 1320	30 30
II	11	4.5	400	20	190	610	30
Black crappie	H	3.0	780	100	300	1180	30
Largescale sucker	1974	5.1	1100	540	690	2330	80
Smallmouth bass Carp	'' II	4.8 4.9	1140 3100	310 440	390 800	1840 4340	80 170
lt .	11	4.9	2200	190	550	2940	120
Largescale sucker	1973	6.0	880	510	390	1780	10u
Largemouth bass		12.0 4.2	1000 770	670 110	470 140	2140 1020	160 10u
Carp	н	5.4	520	10u	10u	520	10u
Largescale sucker	1972	4.3	920	640	440	2000	10u
Northern squawfish	H	4.2 3.8	1100 2700	780 10	550 340	2430 3050	10u 10u
Carp	11	6.4	2200	140	420	2760	10u
Largescale sucker	1971	4.9	1120	1040	520	2680	10
Black crappie		$\frac{5.4}{1.9}$	710 490	550 180	380 140	1640 810	20 10
н	11	2.3	550	160	140	850	30
Carp	11 11	4.4 5.0	1070 1020	170 80	250 380	1490 1480	10 20
	4 A						
Bridgelip sucker Largemouth bass	1970 "	7.9 3.9	660 1660	1030 420	480 510	2170 2590	60 80
Carp	II	5.6	890	150	30	1070	30
<u> </u>	11	5.0	1700	170	350	2220	20

Sources: Schmitt, et al. (1981, 1983, 1985).

Note: USFWS data older than 1970 were not used because of reported interferences in the analysis (Schmitt, 1986).

u = Not detected at detection limit shown.