Toxaphene

Improved Recognition in Washington Streams, Rivers, and Lakes

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Cover photo: Retrieving an SPMD Passive Sampler from Granger Drain, Yakima Valley (credit: Dianne Kinney)

Toxaphene

Improved Recognition in Washington Streams, Rivers, and Lakes

by

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Waterbody Numbers:

Multiple waterbodies statewide

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Abstract

The occurrence of toxaphene, a banned legacy pesticide, has historically been under-reported in Washington streams, rivers, and lakes because of analytical challenges and a focus on other chemicals. This report reviews several recent water quality and fish tissue studies in Washington that have benefitted from improved recognition of this chlorinated insecticide. Toxaphene should be recognized as a potential water quality concern, particularly in eastern Washington waterbodies, currently known to include the Snake, Yakima, and Walla Walla Rivers.

Acknowledgements

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- A number of individuals gave or arranged for access to sampling sites. We extend our appreciation to Mark Boughman (Star Lake), Elaine Brouillard (Rosa-Sunnyside Board of Joint Control - Sulphur Creek and Granger Drain), Jim Patterson (Silver Lake), Sarah Southam (Lake Lawrence), and Sarah White (Army Corps of Engineers - Lower Granite Dam)
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- Washington State Department of Ecology staff:
    - The toxaphene data reported here is chiefly the work of Myrna Mandjikov of Manchester Environmental Laboratory. We are grateful for her interest, insight, and attention to our samples.
    - Ecology staff provided background information and advice on sampling sites. They included Chad Atkins, Greg Bohn. Jonathan Jennings, Sally Lawrence, and Jim Ross.
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Introduction

Toxaphene, once touted as a replacement for DDT, was the last of the chlorinated pesticides to be banned in the United States. It is a complex mixture of hundreds of chlorinated bornanes, camphenes, and related compounds (Figure 1) and is difficult to analyze. Although toxaphene is one of the EPA priority pollutants, its detection has historically been infrequent in environmental samples.

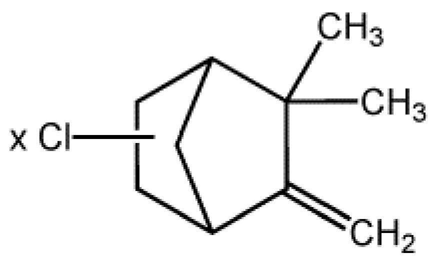

Figure 1. General Structure of Toxaphene (67-69% chlorine by weight).

As of July 2010, 3,355 records of toxaphene being analyzed in fish, water, or sediment had been entered into the Washington State Department of Ecology (Ecology) Environmental Information Management (EIM) database. Overall detection frequency was only 3.3% percent of samples (6.3% for fish, 1.3% for water, and 0.8% for sediment). A pattern of low detection frequency is also seen nationally for toxaphene (Raff and Hites, 2004; Wong et al., 2000).

Discovery of high toxaphene levels in an eastern Washington agricultural stream in 2002 provided an opportunity for improved monitoring of this legacy pesticide by the Ecology Environmental Assessment Program (EAP). The present report reviews the toxaphene concentrations that have been measured in Washington streams, rivers, and lakes over the ensuing 10 years. Further efforts at source investigation and cleanups are recommended for several river basins in eastern Washington.

Uses of Toxaphene

Toxaphene, also known as camphechlor, was introduced in the late 1940s. The primary use in the United States, estimated at 70-90% of total U.S. production, was on insect pests of cotton and soybeans in the southeast (ATSDR, 1996). Less than 1% was used on agriculture in the Midwest and in western states (Von Rumker et al., 1975). Toxaphene does not appear to have been an important pesticide on agricultural crops in Washington, but the true extent of its use here is unknown.

The second largest use, 7-15% of U.S. production, was to control parasites and other insect pests on livestock and poultry (Glassmeyer et al., 1997; ATSDR, 1996; Knipling and Westlake, 1966). There is a circumstantial link between use on animals and water quality concerns in Washington streams, as discussed later in this report.

Toxaphene was also used to eliminate undesirable fish species in lakes and ponds beginning in the mid-1950s (Eisler and Jacknow, 1985). By 1966 it was the chemical of choice in fish eradication programs in Canada and second in the U.S. after rotenone (Lennon et al., 1970). The practice was especially prominent in the northern states.

Records provided by the Washington Department of Fish and Wildlife (WDFW) show that 94 Washington lakes were treated with toxaphene or a combination of toxaphene and rotenone between 1954 and 1969. There were 111 toxaphene treatments overall. Four lakes were also treated with pentachlorophenol. WDFW stopped using toxaphene and pentachlorophenol after 1969 because the persistent residues killed planted trout (Hisata, 2002).

For many years toxaphene was the most heavily used insecticide in the U.S. EPA cancelled most uses as a pesticide or pesticide ingredient in 1982. All registered uses of toxaphene were banned in 1990 and existing stocks were not allowed to be sold or used in the U.S. after March 1, 1990.

Water Quality Criteria

Washington State's water quality criteria for toxaphene are shown in Table 1. Toxaphene is unique among the chemicals regulated by Washington's water quality standards in having an aquatic life criterion that is lower (more restrictive) than its human health criterion (0.2 ng/L vs. 0.73 ng/L, parts per trillion). Furthermore, toxaphene's chronic criterion is the lowest among Washington's 32 aquatic life criteria chemicals.

Table 1. Washington State Water Quality Criteria for Toxaphene
(ng/L; parts per trillion).

Aquatic Life (WAC 173-201A)		Human Health (EPA National Toxics Rule)
Freshwater Chronic	Freshwater Acute	Water & Fish Consumption
0.20	730	0.73

The chronic criterion is a 4-day average concentration not to be exceeded more than once every three years on average. The acute criterion is for a 1-hour average concentration not to be exceeded more than once every three years on average. The human health criterion is for a 10^{-6} excess lifetime cancer risk (1 in 1,000,000) as promulgated in the EPA National Toxics Rule (NTR).

To assess the human health risk from chemical contaminants in edible fish tissue, Ecology uses values derived from the NTR human health water quality criteria and EPA bioconcentration factors (BCFs). The BCF predicts the chemical concentration in fish that would be expected to result from a given concentration in the water column. For a 10^{-6} cancer risk where both water and fish are consumed, the fish tissue criterion for toxaphene is 9.6 ug/Kg (parts per billion, wet weight; BCF = 13,100). In essence, the fish tissue criterion is the human health water quality criterion expressed in tissue form.

The edible fish tissue criterion for toxaphene is shown in Table 2 and compared with the seven toxic chemicals most frequently reported in Washington State freshwater fish (Seiders and Deligeannis, 2009). In terms of relative health risk, toxaphene ranks in the mid-range (45^{th} percentile) of the 84 chemicals for which Ecology has adopted human health criteria. Compared to the contaminants most frequently accumulated by local sport fish species, toxaphene is more toxic than mercury[1] or DDT; roughly similar in toxicity to chlordane, hexachlorobenzene, and PCBs; and less toxic than dieldrin or dioxin.

[1] Unlike the other chemicals listed in Table 2, mercury is not a carcinogen but adversely affects the nervous system. The fish tissue mercury criterion is based on a reference dose that is likely to be without an appreciable risk of deleterious health effects during a lifetime (EPA 2001).

Table 2. Human Health Criteria for Edible Fish Tissue: Toxaphene vs. Toxic Chemicals Most Frequently Reported in Washington State Freshwater Fish.
(ug/Kg, wet weight).

Chemical	Human Health Criteria
TCDD (dioxin)	0.000065
Dieldrin	0.65
Total PCBs	5.3
Hexachlorobenzene	6.5
Chlordane	8.0
Toxaphene	**9.6**
DDT	32
Mercury	300

Recent Ecology Studies Detecting Toxaphene

Initial Recognition in the Walla Walla Drainage

In 2002-03 Ecology conducted a water quality study to develop a Total Maximum Daily Load (TMDL) for chlorinated pesticides and PCBs in the Walla Walla River (Johnson et al., 2004). Section 303(d) of the Clean Water Act requires states to prepare a list every two years of waterbodies that do not meet water quality standards. The Act requires that a TMDL be developed for every waterbody and pollutant on the list. A TMDL determines the loading capacity of the waterbody and allocates that pollutant load among sources. Toxaphene was not among the original chemicals of concern for the Walla Walla TMDL.

Passive Sampling

This study marked the department's first use of a passive water sampling technique employing a Semipermeable Membrane Device (SPMD, Figure 2). An SPMD is composed of a thin-walled, layflat polyethylene tube filled with a neutral lipid material, triolein. When placed in water, dissolved lipophilic (fat soluble) compounds such as chlorinated pesticides diffuse through the membrane and are concentrated over time.

Figure 2. Standard SPMD Membrane Mounted on a Stainless Steel Spider Carrier.

SPMDs offer several advantages over traditional water samples. The large chemical residues accumulated in the membranes give a strong analyte signal, which translates into parts per trillion detection limits or lower. Because SPMDs measure the long-term average concentration of a chemical, random fluctuations are smoothed and representativeness of the data improved.

Studies have shown that chemical concentrations derived from SPMDs are comparable to other more complex, low-level sampling methods such as solid-phase and liquid-liquid extraction, generally agreeing within a factor of 2 (Ellis et al., 1995; Rantalainen et al., 1998; Hyne et al., 2004). Details of SPMD theory, construction, and applications can be found at wwwaux.cerc.cr.usgs.gov/spmd/index.htm and in Huckins et al. (2006).

Methods used by Ecology to convert the chemical residues accumulated by SPMDs into estimated water column concentrations are described in the EAP SOP (Seiders et al., 2011) and in studies referenced in the present report. Because the concentrations are calculated values based on known uptake properties of the membranes, as opposed to directly measured concentrations, the results are considered estimates.

Walla Walla Findings

SPMDs were used in the Walla Walla TMDL study to identify pesticide and PCB sources to the river. They were deployed quarterly for approximately one month each at four mainstem stations and on six tributaries (Figure 3).

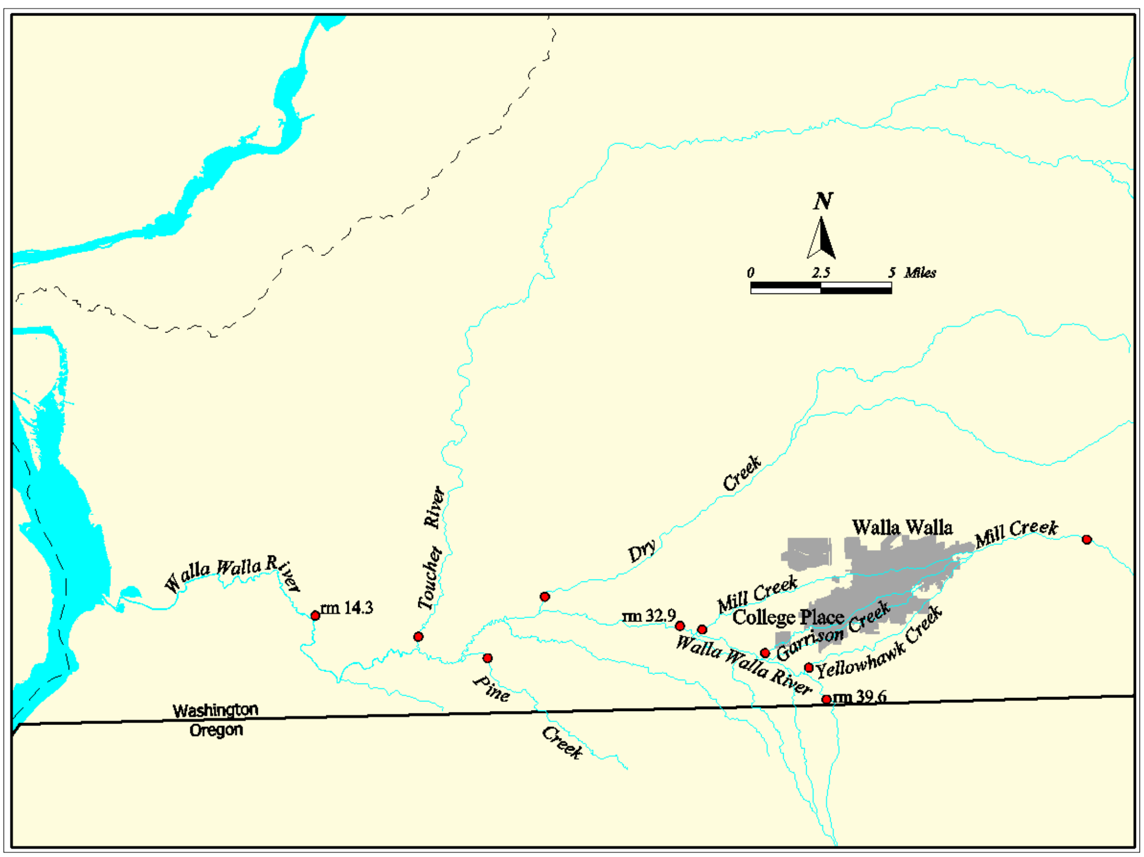

Figure 3. Sites Where SPMDs were Deployed in the Walla Walla River Drainage during 2002-03.

The extracts from the Walla Walla SPMDs were analyzed by the Ecology Manchester Environmental Laboratory using gas chromatography with electron capture detection (GC/ECD). GC/ECD is the most commonly employed technique for analyzing chlorinated pesticides and PCBs in environmental samples. In the past, Manchester had difficulty identifying low-level toxaphene residues due to background noise and weathering effects that change the composition over time. PCBs, which are ubiquitous in the environment, also cause interferences.

The concentration effect of the SPMDs, however, revealed large, distinct peaks resembling degraded toxaphene in Pine Creek. Figure 4 shows the chromatogram from the initial Pine Creek SPMD retrieved in June 2002.

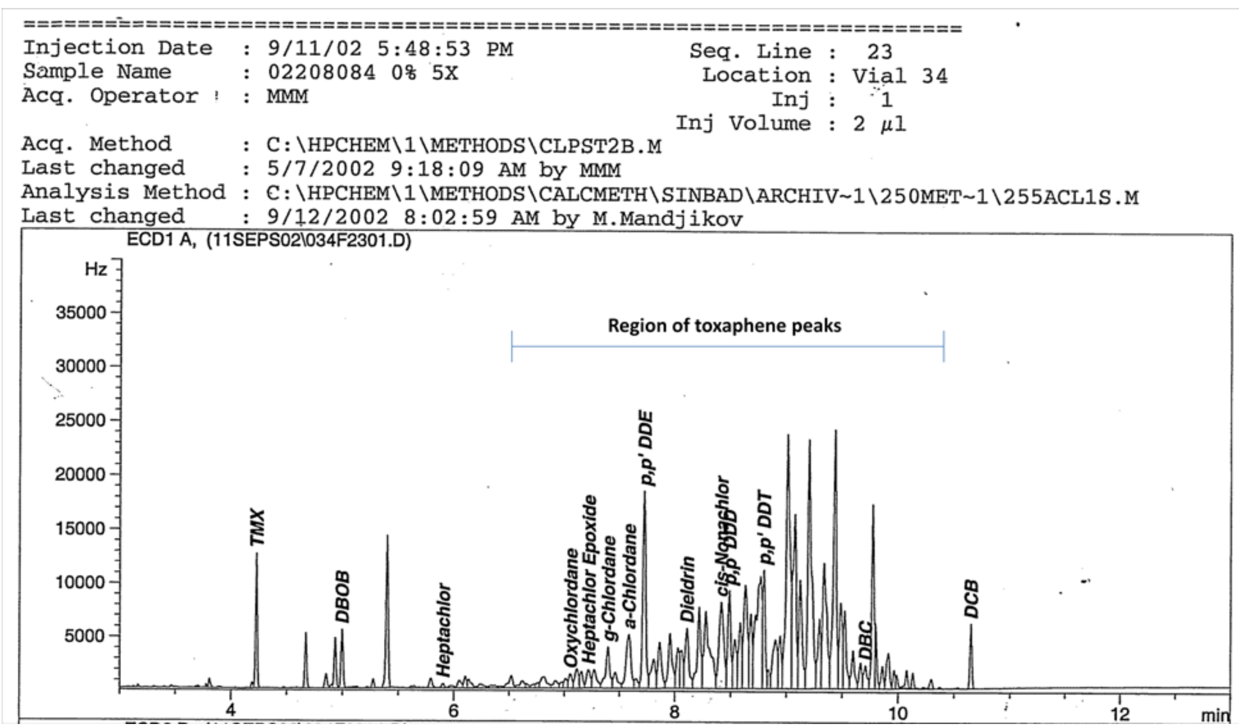

Figure 4. GC/ECD Chromatogram from Pine Creek SPMD Deployed May-June 2002.

In this figure, the pesticide compounds marked g-chlordane, a-chlordane, p,p'-DDE, p,p'-DDD, and p'p'-DDT are single peaks. Toxaphene makes up the rest of the pattern starting around 6.5 minutes and ending around 10.5 minutes (retention time). Other pesticides labeled within this range (e.g., dieldrin, heptachlor epoxide) are false positives on toxaphene peaks.

Once the pattern became clear, Manchester was able to quantify toxaphene in other Walla Walla samples. Samples closest to Pine Creek showed a definite pattern match, with the same peaks being present but reduced in height, being taken further from the source. Toxaphene peaks could be determined because they matched the specific retention times and were proportional.

The toxaphene concentration in Pine Creek during May-June 2002 (early part of irrigation season) was estimated at 40 ng/L, exceeding the 0.2 ng/L chronic aquatic life criterion by more than two orders of magnitude (Table 3). Concentrations decreased to approximately 2 ng/L by the fall – still, however, substantially above criteria. Elevated toxaphene levels were also measured in the mainstem Walla Walla River downstream of Pine Creek.

The near-detection limit levels observed elsewhere in the Walla Walla drainage during this period suggested a unique source of toxaphene within Pine Creek, as opposed to widespread use on crops. When these data were presented to a citizen advisory group in Walla Walla, several members implicated an animal feedlot on Pine Creek as a possible source of contamination. This claim has never been substantiated.

Table 3. Toxaphene Concentrations in the Walla Walla River and Tributaries during 2002-03.
(total toxaphene in parts per trillion; N=4)

Sampling Site	River Mile	Toxaphene (ng/L)	
		Mean	Maximum
Upper Walla Walla River @ Peppers Bridge	39.6	<0.3	<0.3
Yellowhawk Creek @ Old Milton Highway	37.9	0.7	1.6
Garrison Creek @ Mission Rd.	36.1	0.7	1.8
Upper Mill Creek @ Seven Mile Rd.	- -	<0.3	<0.3
Lower Mill Creek @ Mission Rd.	33.6	0.4	0.6
Middle Walla Walla River @ Detour Rd.	32.9	0.5	0.6
Dry Creek @ Highway 12 Bridge	27.2	0.4	0.5
Pine Creek @ Sand Pit Rd.	**23.4**	**13**	**40**
Touchet River @ Highway 12 Bridge	21.6	0.4	0.7
Lower Walla Walla River bw. Cummins Bridge	**14.3**	**3.2**	**8.3**
Chronic aquatic life water quality criterion = 0.2 ng/L			

Fish samples collected from the Walla Walla River for the same study also showed exposure to toxaphene (Table 4). Concentrations in fish tended to be higher in the lower river downstream of Pine Creek. The 9.6 ug/Kg human health criterion was exceeded in all samples, by up to a factor of 6 in channel catfish and carp fillets. Movements of fish may have obscured differences in toxaphene levels known to exist between the upper and lower river.

At low levels, toxaphene is difficult to distinguish from background noise in environmental samples. With the benefit of experience gained in pattern recognition during the Walla Walla project, Manchester was positioned to detect and quantify toxaphene in several similar EAP studies that followed, described below.

Table 4. Average Toxaphene Concentrations in Composite Fish Tissue Samples Collected from the Walla Walla River during July-September 2002.
(parts per billion, wet weight; N = 1- 4)

Species	Tissue	Toxaphene (ug/Kg)	
		Upper River*	Lower River†
Channel catfish	Fillet	NA	58
Common carp	"	17	56
Northern pikeminnow	"	20	27
Bridgelip sucker	"	16	20
Smallmouth bass	"	26	10
Northern pikeminnow	Whole body	36	50
Bridgelip sucker	"	27	36
Human health edible fish tissue criterion = 9.6 ug/Kg			

*Dry Creek to Yellowhawk Creek.
†below Touchet River.
NA = not analyzed.

Detection in Mainstem of Lower Columbia River

A second water quality study that relied on SPMDs was conducted for the Lower Columbia River in 2003-04 (Johnson and Norton, 2005). The objective was to monitor 303(d) listed pesticides, PCBs, and polyaromatic hydrocarbons (PAH) that had exceeded human health criteria in fish or water samples from the river. SPMDs were deployed in the spring, winter, and fall at five sites on the mainstem between Bonneville Dam and Longview, and at the mouths of eight major tributaries, including the Willamette River.

Toxaphene was not a target compound for this project. However, for the SPMDs deployed in the spring, Manchester again saw patterns indicative of toxaphene, but only discernable in the mainstem Columbia River. This suggests that the major sources were upstream of Bonneville Dam, possibly associated with the early part of the irrigation season when pesticide levels are typically highest in eastern Washington irrigation returns.

Concentration estimates for the mainstem Columbia River are shown in Table 5.

Table 5. Toxaphene Concentrations in the Mainstem Lower Columbia River during May – June 2004.
(dissolved toxaphene in parts per trillion; N=1)

Site	River Mile	Toxaphene (ng/L)
Above Bonneville Dam	147	0.26
Below Bonneville Dam	142	0.24
Above Willamette River	103	0.16
Above Kalama River	75	*
Below Longview	54	0.21
Chronic aquatic life water quality criterion = 0.2 ng/L		

*sample lost in laboratory accident.

Widespread Contamination of the Yakima River Basin

A 2006-08 study in support of a chlorinated pesticide and PCB TMDL for the Yakima River basin afforded an opportunity to analyze for toxaphene in one of the most intensively irrigated and agriculturally diverse areas in the U.S. (Johnson et al., 2007, 2010). Although not among the pesticides that had been identified as needing a TMDL, toxaphene was readily detectable in a number of samples.

SPMDs were deployed in the upper and lower Yakima River and selected tributaries and irrigation returns in 2007, specifically to give improved detection capability for PCBs and toxaphene. Except for the Ellensburg area (Wilson Creek drainage) irrigated agriculture is primarily concentrated in the lower Yakima Valley. The locations of the lower river deployment sites are shown in Figure 5. Monitoring was conducted in May-June during the early part of the irrigation season and again in October-November after the end of the irrigation.

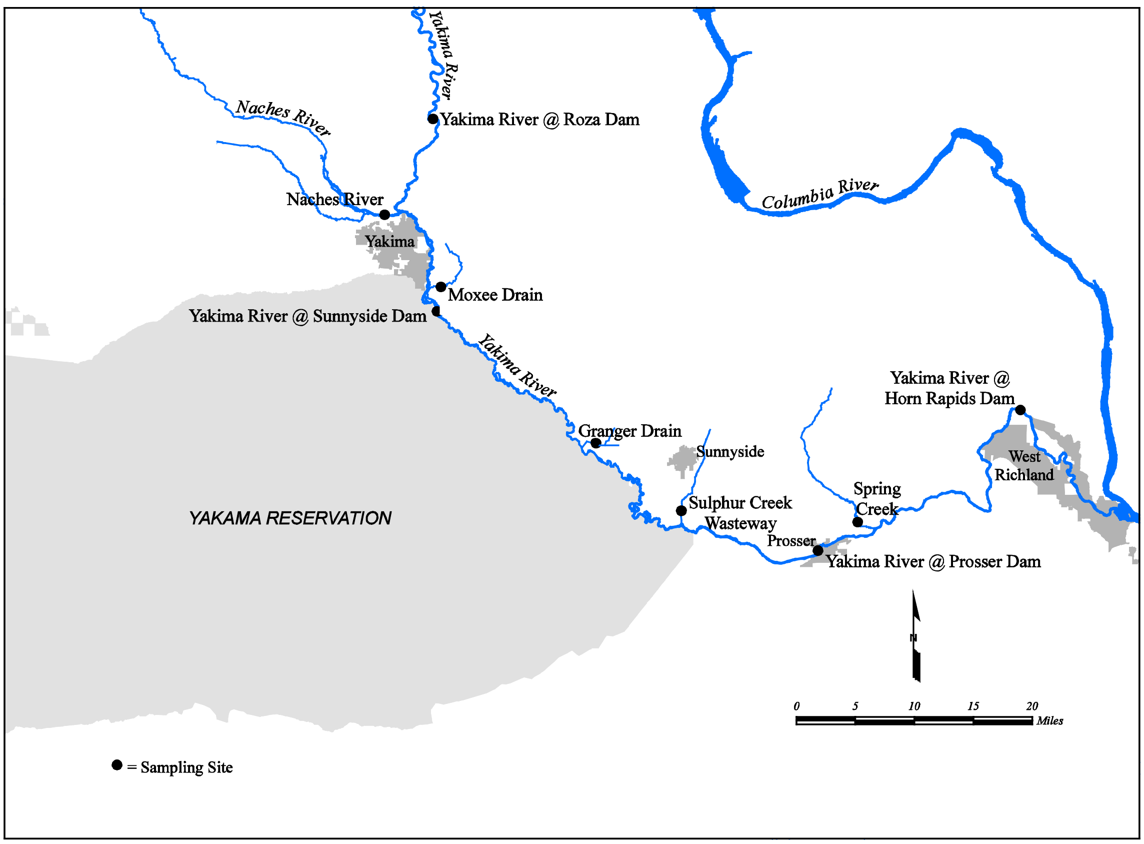

Figure 5. Location of SPMDs Deployed in the Lower Yakima River during 2007.

The toxaphene results are summarized in Figure 6. In this figure, the sampling sites are arranged in downstream order, left to right. The mainstem SPMDs were installed at diversion dams (labeled Easton, Town, Roza Dam, Sunnyside, Prosser, and Horn Rapids in Figure 6).

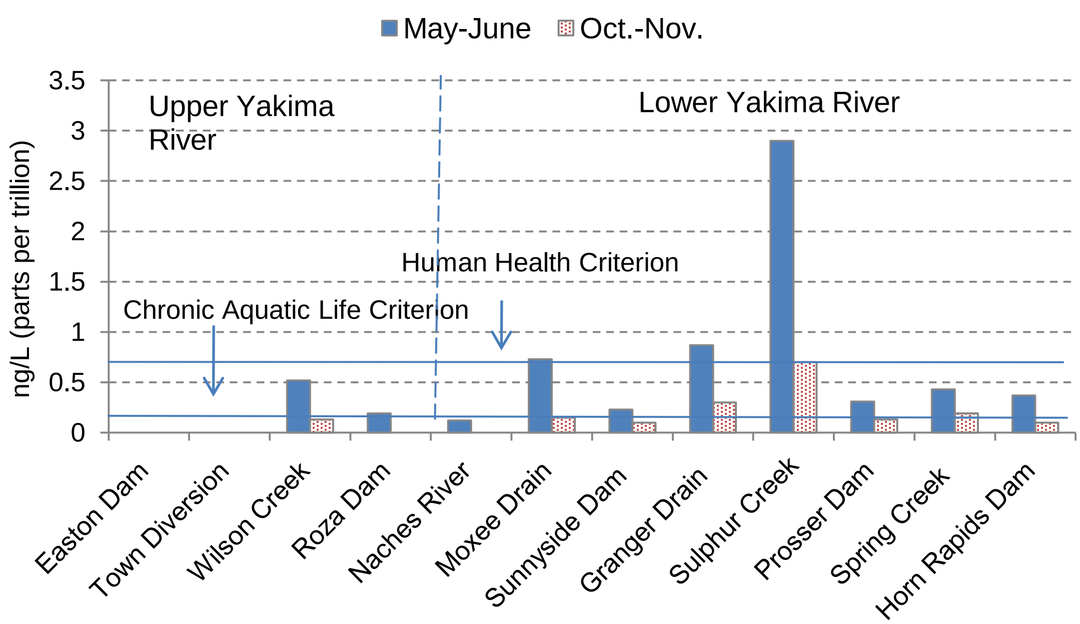

Figure 6. Toxaphene Concentrations in the Mainstem Yakima River, Tributaries, and Irrigation Returns During and After the 2007 Irrigation Season.

(total toxaphene, non-detects plotted at zero)

Toxaphene levels gradually increased in the mainstem Yakima River moving downstream from Easton, at river mile (RM) 202 to Horn Rapids Dam (RM 18) near the confluence with the Columbia. During the irrigation season, toxaphene was below detection limits down to at least Ellensburg (Town Diversion site, RM 147).

Wilson Creek, which enters the Yakima at river mile 147 just below Ellensburg, had an elevated toxaphene concentration of 0.52 ng/L. Further downstream, mainstem levels rose to 0.19 ng/L at Roza Dam (RM 128), and 0.23 ng/L by Sunnyside Dam (RM 104) just below the city of Yakima, at or slightly above the chronic aquatic life criterion. Between Sunnyside Dam and Horn Rapids Dam, toxaphene nearly doubled to 0.37 ng/L. After the end of the irrigation season (October-November samples), toxaphene continued to exhibit increased concentrations in the lower river, but remained within criteria.

Toxaphene substantially exceeded the chronic aquatic life criterion in Wilson Creek, Moxee Drain, Granger Drain, and especially Sulphur Creek Wasteway. This occurred primarily during the irrigation season, when the 0.2 ng/L criterion was exceeded by factors of 2- 4 in Wilson Creek, Moxee Drain, and Granger Drain, and by more than a factor of 10 in Sulphur Creek Wasteway (2.9 ng/L). Concentrations decreased markedly in all the four drains after the end of irrigation, but remained elevated in Sulphur Creek, exceeding the criterion by a factor of 3. Toxaphene was at or above the human health criterion (0.73 ng/L) in two of these returns.

Fish tissue samples collected from the Yakima mainstem showed a strong trend toward increasing toxaphene concentrations moving downstream, consistent with the location of identified sources (Figure 7). Several species exceeded human health criteria in the lower river, including mountain whitefish, largescale suckers, northern pike minnow, and carp. Concentrations in fillets ranged from 11 – 56 ug/Kg, similar to earlier findings for lower Walla Walla River fish.

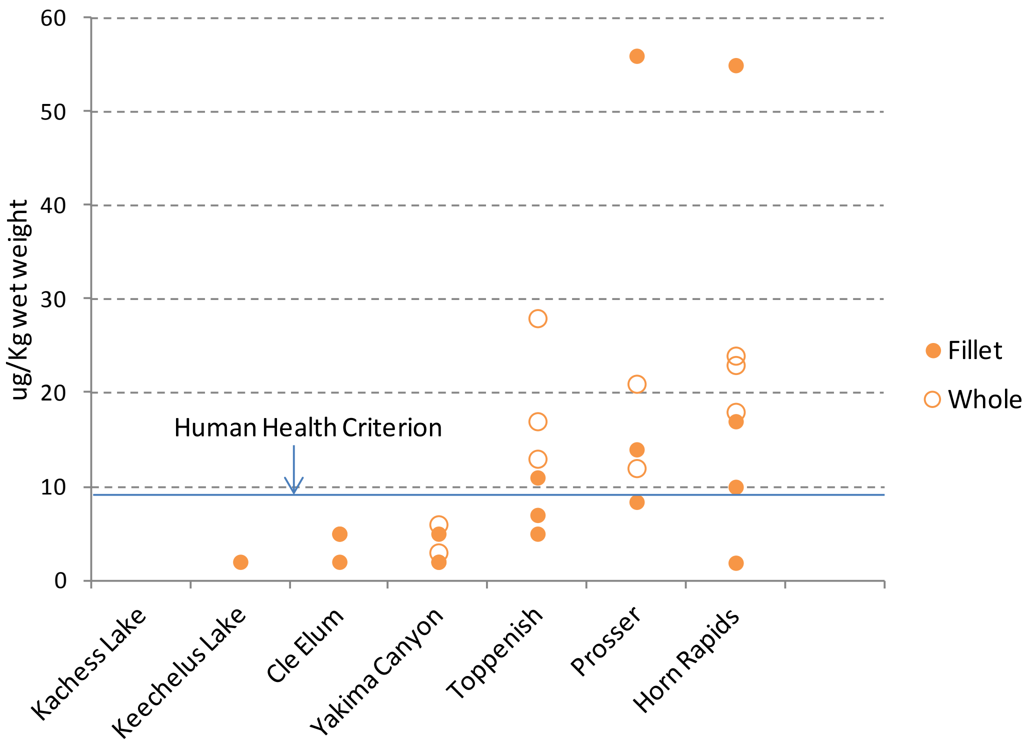

Figure 7. Mean Toxaphene Concentrations in Composite Fish Tissue Samples Collected from the Yakima River in 2006.

(Johnson et al., 2007; Kachess and Keechelus Lakes are storage reservoirs in the upper river.)

The link between elevated levels of toxaphene in the Yakima River drainage and its use on livestock is, again, circumstantial. Sulphur Creek Wasteway and Granger Drain both have an unusually high concentration of dairies and feedlots in their watersheds; 5% of the land is confined animal feeding operations, not including pasture. In 2002, there were 24 dairies and two large feedlots within the Sulphur Creek watershed and 23 dairies in the Granger watershed (Zuroske, 2004). Many of these may have been in existence prior to 1984 when EPA cancelled most uses of toxaphene in agriculture.

A large cattle feedlot (Schaake) was historically located in Ellensburg along Wilson Creek. Its runoff discharged to Wilson Creek via Tjossem Ditch. Potential sources to Moxee Drain are less obvious. A medium-sized seasonal sheep operation (40-50 years old) is located upstream, in additional to the many hobby farms common to the Yakima area.

Increased Focus by Washington State Toxics Monitoring Program

Fish Tissue

Ecology's Washington State Toxics Monitoring Program (WSTMP) has an exploratory component that analyzes toxic chemicals in resident freshwater fish statewide www.ecy.wa.gov/programs/eap/toxics/wstmp.htm. This is a screening-level effort primarily devoted to identifying areas of concern for follow-up actions. The bulk of the 303(d) water quality limited listings for chemicals exceeding human health criteria for fish consumers come from this program. www.ecy.wa.gov/programs/wq/303d/index.html.

Since its inception in 2001, WSTMP has screened over 300 fish fillet samples from more than 100 waterbodies statewide. From 2001 through 2007 and again in 2010, reporting limits for toxaphene were typically in the vicinity of 20 ug/Kg range or higher and detection frequency was only 5% of samples or less. In 2008 and 2009, a 5 ug/Kg reporting limit was more consistently achieved and detection frequency for toxaphene increased to 47% and 46% of samples, respectively.

Up through 2010, WSTMP has identified 11 waterbodies where toxaphene has exceeded the 9.6 ug/Kg human health criterion in one or more fish fillet samples (Table 6). Snake River fish stand out here. For the four species analyzed, the criterion was not being met throughout 140 miles of the Snake, from Clarkston at the state line to Ice Harbor Dam, six miles above the Columbia River confluence. The highest concentrations were found in channel catfish and carp, exceeding the toxaphene criterion by factors of approximately 5–10 (Figure 8).

The source of contamination in the Snake is assumed to be agriculture-related but has not been determined. A recent limited effort at screening several Snake River tributaries for toxaphene is described later in this report.

Most of the other rivers and lakes listed in Table 6 as exceeding criteria appear to have a similar level of contamination in the approximate range of 10-20 ug/Kg. Some of these are minimally impacted by local human activities. This suggests that the toxaphene residues in these fish may be largely due to long-range global atmospheric transport, as has been observed for toxaphene in other studies (Raff and Hites, 2004 and references therein).

Table 6. Waterbodies Where One or More WSTMP Fish Fillet Samples Have Exceeded Human Health Criteria for Toxaphene.
(parts per billion, wet weight)

Waterbody	Year Sampled	Species	Toxaphene (ug/Kg)
Queets River	2004	CHNK	9.7
Snake River ab. Ice Harbor Dam	2004	CAT	19
Meridian Lake	2006	LMB, KOK	11 - 15
Vancouver Lake	2006	LMB	28
Fish Lake	2008	BNT, LMB	13 - 20
Goodwin Lake	2008	RBT	13
Merrill Lake	2008	CTT	9.9
Stevens Lake	2008	KOK	11
Klickitat River	2008	MWF	16
Snoqualmie River	2008	MWF	10 - 13
Snake River @ Clarkston	2009	CCP	74
Snake River above Lower Granite Dam	2009	CAT, CCP, NPM, MFW	11 - 91
Snake River nr. Central Ferry	2009	CAT, CCP	32 - 63
Snake River nr. Lyon's Ferry	2009	CAT, CCP	12 - 40
Snake River ab. Lower Monumental Dam	2009	CAT, NPM	16 - 91
Snake River ab. Ice Harbor Dam	2009	CAT, NPM	13 - 72
Lake Ozette	2009	SOCK	15
Human health edible fish tissue criterion = 9.6 ug/Kg			

Source: Seiders and Deligeannis (2009); Seiders et al. (2007, 2008); Coots (2007);
WSTMP unpublished 2009 data.

Fish species:
BNT = brown trout
CAT = channel catfish
CCP = common carp
CHNK = Chinook salmon
CTT = cutthroat trout
KOK = kokanee
LMB = largemouth bass
MWF = mountain whitefish
NPM = northern pike minnow
RBT = rainbow trout
SOCK = sockeye salmon

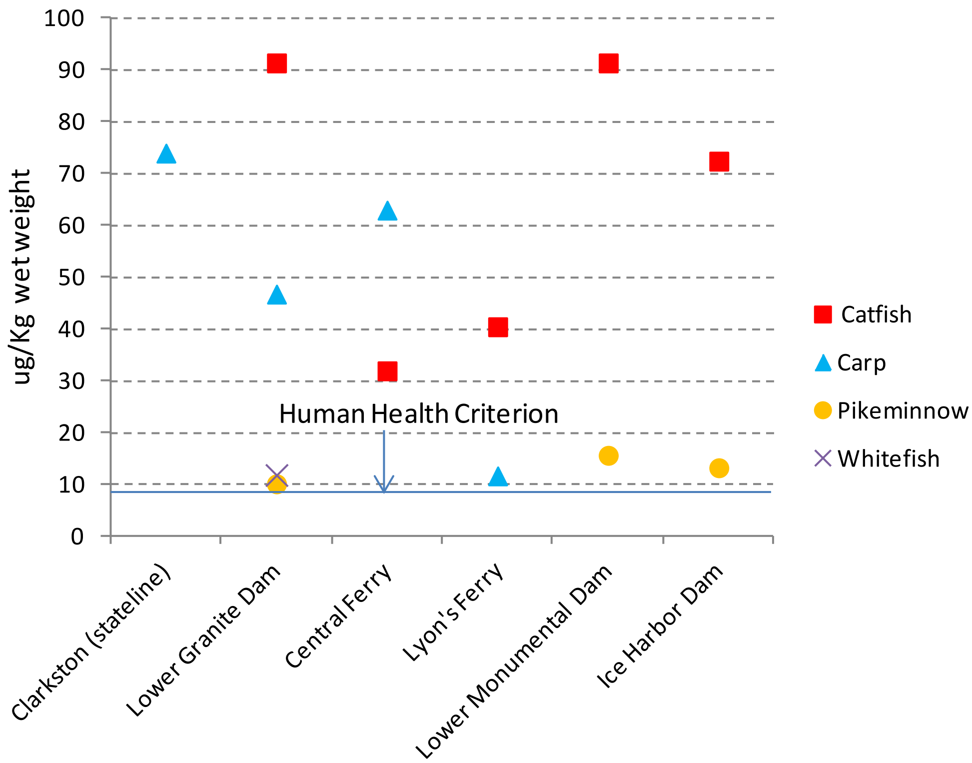

Figure 8. Toxaphene Concentrations in Snake River Fish Fillet Samples Collected in 2009. *(unpublished WSTMP data)*

Trend Monitoring

In 2007, WSTMP initiated a water quality-based trend monitoring program for persistent, bioaccumulative, and toxic chemicals (PBTs). SPMDs were deployed at 12 major Washington rivers and lakes in the spring and fall. Target chemicals included chlorinated pesticides, PCBs, polybrominated diphenyl ethers (PBDEs), and PAHs. Results are reported in Sandvik (2009, 2010) and Sandvik and Seiders (2011).

Monitoring sites include the lower Walla Walla, Yakima, and Columbia Rivers, each with a history of toxaphene detection. The toxaphene data obtained so far are summarized in Table 7 and compared to results from earlier SPMD deployments at these same sites, previously discussed.

Table 7. Trend Data for Toxaphene in the Lower Walla Walla, Yakima, and Columbia Rivers, 2007-09.
(total concentrations from Sandvik (2009, 2010) and Sandvik and Seiders (2011)

Monitoring Site	Toxaphene (ng/L)					
	2007 (spring)	2007 (fall)	2008 (spring)	2008 (fall)	2009 (spring)	Previous samples
Walla Walla River (RM 9)	1.2	0.55	1.2	0.55	1.1	1.7-8.3 (2002-2003*)
Yakima River @ Horn Rapids Dam (RM 18)	0.20	0.19	<0.080	0.13	0.20	0.10-0.37 (2007†)
Columbia River near Clatskanie (RM 54)	0.097	<0.091	<0.084	<0.092	<0.10	0.21 (2004**)
Chronic aquatic life water quality criterion = 0.2 ng/L						

*Johnson et al. (2004).
†Johnson et al. (2010).
**Johnson and Norton (2005).

Toxaphene levels in the lower Walla Walla have shown a consistent pattern of elevated concentrations in the spring compared to fall, first observed in 2002-03. Although a similar type of seasonality was seen for the Yakima River in the 2007 TMDL study, it has not been evident in the trend data so far. Toxaphene has generally escaped detection in the lower Columbia River. For all three rivers, the level of contamination observed in the WSTMP trend monitoring program is comparable to or, for the Columbia, slightly lower than findings from previous studies.

Expanded Investigation of Agricultural Streams and Treated Lakes

With the detection of toxaphene becoming almost routine, EAP initiated an expanded investigation of agricultural streams and treated lakes in 2010 using SPMDs. Details on the design of the study, sampling methods, and chemical analysis can be found in the Quality Assurance Project Plan (Johnson, 2010). These results have not previously been reported.

Case narratives establishing the quality of these data have been prepared by the Ecology Manchester Laboratory and are available for the author on request. Overall, no significant shortcomings were identified in the analytical results. As with other SPMD data reviewed in the present report, all toxaphene concentrations should be considered estimates.

Streams

SPMDs were deployed in 15 streams and irrigation returns and one river in agricultural land use areas that include livestock operations (Table 8, Figure 9). Toxaphene had previously been detected using SPMDs at four of these waterbodies – Wilson Creek, Granger Drain, Sulphur Creek, and Pine Creek. The remaining creeks and returns had not been sampled before. Greater effort was devoted to eastern Washington due to already known instances of significant toxaphene contamination, with half of those sites being in the Snake River basin, including one on the Snake River mainstem.

Table 8. Streams and Rivers where SPMDs were Deployed during Fall 2010 (western Washington) or Spring 2011 (eastern Washington).

Waterbody	Tributary To	Vicinity Of	County
Western Washington			
Fishtrap Creek	Nooksack River	Lynden	Whatcom
Bertrand Creek	Nooksack River	Lynden	Whatcom
Big Ditch	Skagit River	Mt. Vernon	Skagit
Newaukum Creek	Green River	Enumclaw	King
South Fork	Chehalis River	Boistfort	Lewis
Eastern Washington			
Wilson Creek	Yakima River	Ellensburg	Kittitas
Granger Drain	Yakima River	Granger	Yakima
Sulphur Creek	Yakima River	Sunnyside	Yakima
Pine Creek	Walla Walla River	Touchet	Walla Walla
Deadman Creek	Snake River	Central Ferry	Garfield
Snake River	Columbia River	Lower Granite Dam	Whitman
Steptoe Canyon	Snake River	Clarkston	Whitman
Asotin Creek	Snake River	Clarkston	Asotin
Cow Creek	Palouse River	Hooper	Adams
Crab Creek	Columbia River	Davenport	Lincoln

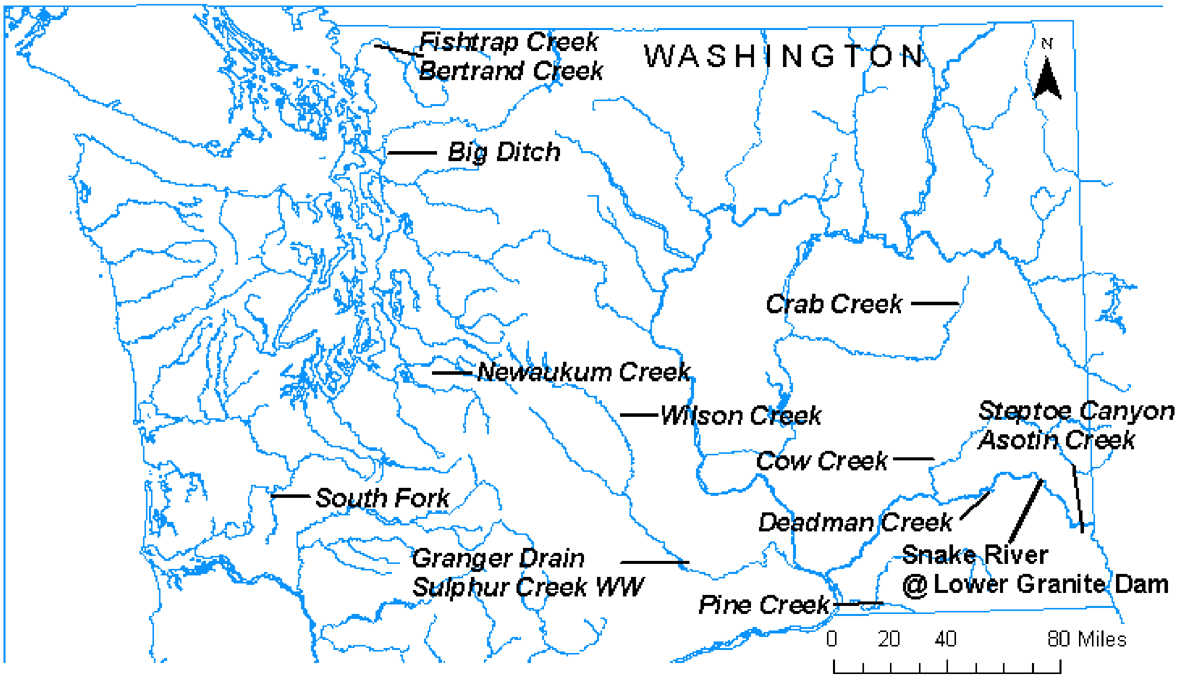

Figure 9. Locations of Streams and Rivers where SPMDs were Deployed.

A USGS review of legacy and current-use pesticides in Pacific Northwest surface waters concluded that detections in the west and in urban areas were dominated by rainfall runoff in the winter and spring, and on the eastside by the irrigation season in the spring and summer (Anderson et al., 2005).Therefore, toxaphene monitoring for this study focused on these periods. For western Washington, SPMDs were deployed in early October during the onset of the rainy season. Deployments for eastern Washington took place in April when runoff typically peaks due to snowmelt and onset of the irrigation season. The SPMDs were left in place for approximately one month.

Results from analyzing the SPMD extracts are summarized in Table 9. The presence of significant levels of toxaphene was confirmed in Wilson Creek Granger Drain, Sulphur Creek, and Pine Creek. Concentrations were similar to earlier findings discussed elsewhere in this report.

Table 9. Results from Analyzing Toxaphene in Agricultural Streams during 2010-2011.
(dissolved concentration in parts per trillion)

Waterbody	Sample No.	Sampling Period	Toxaphene (ng/L)
Western Washington (2010)			
Fishtrap Creek	1012023-07	10/5 - 11/3	< 0.06
Bertrand Creek	1012023-08	10/5 - 11/3	< 0.05
Big Ditch	1012023-14	10/14 - 11/3	< 0.08
Newaukum Creek	1012023-10	10/5 - 11/3	< 0.06
South Fork	1012023-12	10/7 - 11/4	< 0.10
Eastern Washington (2011)			
Wilson Creek	1105050-10	4/12 - 5/11	**0.27**
Granger Drain	1105050-09	4/12 - 5/10	**0.51**
Sulphur Creek Wasteway	1105050-07	4/12 - 5/10	**0.65**
Pine Creek	1105050-01	4/11 - 5/9	**1.4**
Deadman Creek	1105050-02	4/11 - 5/9	**0.75**
Snake River @ Lower Granite Dam	1105050-03	4/11 - 5/9	< 0.17
Steptoe Canyon	1105050-04	4/11 - 5/9	< 0.26
Asotin Creek	1105050-05	4/12 - 5/9	< 0.07
Cow Creek	1105050-06	4/12 - 5/9	< 0.09
Crab Creek	1105050-11	4/14/- 5/10	< 0.12
Chronic aquatic life water quality criterion = 0.2 ng/L			

Toxaphene was only detected in one additional stream, Deadman Creek, a right bank tributary to the Snake River located at Central Ferry. The concentration, 0.75 ng/L, was in the same range as in lower Yakima River irrigation returns and exceeded the aquatic life criterion. None of the four other monitoring sites in the Snake River drainage had detectable amounts of toxaphene.

Failure to detect toxaphene in the mainstem Snake River at Lower Granite Dam runs counter to the high levels seen in fish samples collected near this location in 2009 (Table 6). Due to security concerns at the dam, the deployment site for this particular SPMD was outside the main channel and therefore potentially not representative of water quality conditions in the river.

Historical accumulations of toxaphene in Snake River bottom sediments are a potentially important pathway to fish that has not been evaluated. A recent and large sediment quality study by the Army Corps of Engineers (USACE, 2011) analyzed sediment samples collected over a 150 mile reach of the Snake River from the Columbia River confluence to Idaho border. Toxaphene was not among the pesticides analyzed.

Lakes

Eight lakes were sampled that had been treated with toxaphene between 1957 and 1969 (Table 10, Figure 10). Lakes with characteristics that should favor toxaphene persistence were selected from among the 94 lakes treated from 1954 to 1969, based on examination of records provided by WDFW. Loss rates of toxaphene are typically higher in shallow, eutrophic waterbodies. Long-term persistence is generally associated with deep, soft-water lakes, the type of waterbody where trout fisheries are often important (Hughes and Lee, 1973). Toxaphene may persist for over three decades in the sediments of lakes where it was applied (Miskimmin et al., 1995; Donald et al., 1998).

Factors weighted in selecting lakes for sampling therefore included the following characteristics:

- treated most recently
- treated twice
- deep or located in cooler climates

Table 10. Lakes where SPMDs were Deployed during Fall 2010.

Lake	County	Year Treated	Acres	Maximum Depth	Location
South Puget Sound					
Lawrence	Thurston	63	339	26	6 mi. S of Yelm
Star	King	62	34	50	3 mi. SW of Kent
Quincy Wildlife Area					
Burke	Grant	66	73	33	7.5 mi. SW of Quincy
H	Grant	62 / 67	7	17	6.7 mi. SW of Quincy
Columbia National Wildlife Refuge					
Canal	Grant	59 / 68	76	120	6.5 mi. N of Othello
Upper Goose*	Grant	60 / 69	112	95	9.2 mi N of Othello
Lyle	Adams	59 / 69	22	15	5.3 mi. N of Othello
Spokane Area					
Silver	Spokane	59 / 67	559	80	1.1 mi. E of Medical Lake

*sampler lost.

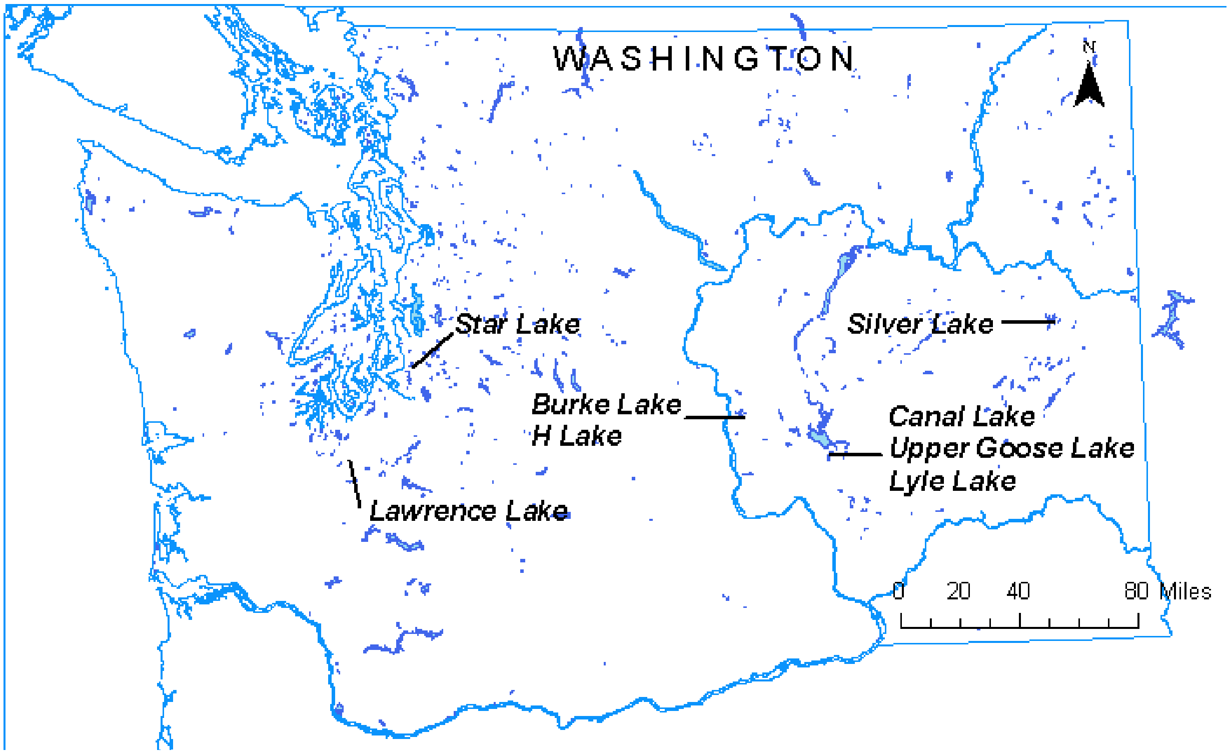

Figure 10. Locations of Lakes where SPMDs were Deployed

To minimize the field effort, lakes were selected that were close to one another within four geographic areas: South Puget Sound, Quincy Wildlife Area, Columbia National Wildlife Refuge, and Spokane area. Where possible, a shallow lake was included for comparison with the deeper lakes in three of the areas. The Upper Goose Lake sampler was lost.

In an effort to obtain results that were representative of each lake as a whole, the SPMDs were deployed in late September after the fall overturn. Formation of the summer thermocline restricts exchange between surface and bottom water which could act to bring about relatively higher toxaphene concentrations near the bottom during the summer months. The samplers were located toward the outflow end of the lake (or center of lake if no outlet) and suspended in the water column. Each deployment lasted about one month.

Results of the SPMD deployments in the toxaphene-treated lakes are summarized in Table 11. Toxaphene was not detected in any of these lakes at detection limits of 0.05 to 0.20 ng/L.

Table 11. Results from Analyzing Toxaphene in SPMDs Deployed in Seven Washington Lakes Historically Treated with Toxaphene.
(dissolved concentrations).

Waterbody	Sample No.	Sampling Period (2010)	Toxaphene (ng/L)
South Puget Sound			
Lawrence Lake	1012023-11	10/7 - 11/9	<0.05
Star Lake	1012023-13	10/13 - 11/9	<0.20
Quincy Wildlife Area			
Burke Lake	1012023-02	9/27 - 10/26	<0.10
H Lake	1012023-03	9/27 - 10/26	<0.08
Columbia National Wildlife Refuge			
Canal Lake	1012023-01	9/27 - 10/26	<0.16
Lyle Lake	1012023-05	9/27 - 10/26	<0.06
Spokane Area			
Silver Lake	1012023-06	9/28 - 10/25	<0.08
Chronic aquatic life water quality criterion = 0.2 ng/L			

Although toxaphene was not detected in the SPMDs deployed in treated lakes or western Washington agricultural streams, trace amounts of other commonly observed chlorinated pesticides or breakdown products were tentatively identified (Table 12). This demonstrates that the samplers were functioning properly as low-level monitors for chemicals similar to toxaphene.

Table 12. Other Chlorinated Pesticides or Breakdown Products Tentatively Identified in SPMD Extracts from Toxaphene Investigation of Streams and Lakes in 2010.

Waterbody	Sample No.	Compounds Tentatively Identified
Fishtrap Creek	1012023-07	DDT, DDE, DDD, endosulfan sulfate, chlordane, pentachloroanisole
Bertrand Creek	1012023-08	DDT, DDE, DDD, endosulfan sulfate, chlordane, pentachloroanisole
Big Ditch	1012023-14	Pentachloroanisole
Newaukum Creek	1012023-10	Pentachlorobenzene
South Fork	1012023-12	Pentachloroanisole
Lawrence Lake	1012023-11	Chlordane, hexachlorobenzene, pentachloroanisole
Star Lake	1012023-13	DDE, pentachloroanisole

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Historical Analyses for Toxaphene

Prior to 2002, the detection frequency of toxaphene in water or fish tissue samples from Washington streams, rivers, and lakes approached zero. Past analytical efforts are reviewed below.

Water Samples

High toxaphene levels are generally required for detection in water samples without a preconcentration technique, SPMDs being a recent example. The only detections in whole water samples in Ecology's EIM database are from the previously mentioned Yakima River TMDL (Johnson et al., 2010). Concentrations estimated at 3.5–11 ng/L (total) were detected in grab samples from Sulphur Creek Wasteway and Moxee Drain on two occasions, each during the 2007 irrigation season. In the same study, a high concentration of 31 ng/L was found in a stormwater runoff sample collected in the City of Yakima during a rain event in March 2008.

USGS deployed SPMDs during 1997 low-flow conditions and 1998 high-flow conditions at nine mainstem sites and seven tributary sites on the Columbia River (McCarthy and Gale, 1999). The study spanned approximately 700 river miles from Northport (RM 735) just below the British Columbia border to Bradwood (RM 39). Although chlorinated pesticides were analyzed, no results are reported for toxaphene.

Fish Fillets

Going back to 1983, Ecology, EPA, and other agencies have conducted a number of large statewide or national surveys to identify toxic chemicals of concern in edible fish tissue. Because of analytical challenges and a focus on other more widely-used pesticides and PCBs, toxaphene has almost always been under-reported in these studies.

EPA conducted two national surveys that included freshwater fish samples from Washington State. The National Study of Chemical Residues in Fish sampled multiple Columbia River sites and the Yakima, Puyallup, and Snohomish River in the late 1980s (EPA, 1992). The National Study of Chemical Residues in Lake Fish Tissue sampled 14 Washington lakes and reservoirs in 1999-2003 (EPA, 2009). The former did not analyze or did not report toxaphene and the latter suffered from an elevated quantitation limit (100 ug/Kg).

Fish in the Columbia River and its tributaries were the focus of the 1991-1995 Bi-State Program (Tetra Tech, 1996) and the 1996-98 Columbia River Basin Fish Contaminant Survey (EPA, 2002) The Bi-State sampled downstream of Bonneville Dam. The more recent EPA study sampled as far upstream as the Hanford Reach and included fish from the Yakima and Snake Rivers, but it did not collect from the Walla River. Here again, reporting limits were not optimized for toxaphene (150-300 ug/Kg and 48-120 ug/kg, respectively).

The earliest statewide edible fish tissue studies by Ecology either do not report toxaphene (Hopkins et al., 1985) or had elevated detection limits (Johnson and Norton, 1990 reported 150 ug/Kg; Hopkins, 1991: 160 ug/Kg; Serdar et al., 1994: 346-550 ug/Kg). Between 1992 and 1995, the Ecology Washington State Pesticide Monitoring Program (WSPMP), a predecessor to the WSTMP, screened 42 fish fillet samples from 22 waterbodies including the Yakima, Snake, Walla Walla, and Lower Columbia (Davis and Johnson, 1993; Davis et al., 1995, 1998). Toxaphene was not detected in any of these samples at or above 110-1,095 ug/Kg. Fish tissue studies that focused on specific Washington waterbodies have also generally failed to detect toxaphene, although not always due to poor detection limits (e.g., WDOH, 2004).

Whole Fish

The lone exception to this pattern has been whole fish samples. The U.S. Fish and Wildlife Service had a National Contaminant Monitoring Program in the 1970s and 80s that included stations on the Yakima, Snake, and Columbia Rivers (Schmitt et al., 1990, 1999). Nationally, this program saw that toxaphene concentrations in whole freshwater fish "plateaued in 1970-76 after a period of steady increase through the 1970s", during which time DDT residues were decreasing. From 1980 to 1986, whole fish from the Yakima, Snake, and Columbia stations showed toxaphene concentrations in the range of 200 – 1,000 ug/Kg. The U.S. Geological Survey (USGS) BEST Program (Biomonitoring of Environmental Status and Trends) revisited these sites in 1997-1998 and only detected toxaphene in two whole largescale sucker samples from the Columbia River near Vancouver (50 ug/Kg). The BEST reporting limit for toxaphene was 30 ug/Kg (Hinck et al., 2004).

In 1986, the Yakima River basin was selected as one of four surface-water pilot studies for the USGS National Water-Quality Assessment (NAWQA) Program. Rinella et al. (1992) reports substantial toxaphene concentrations of 310 – 1,200 ug/Kg in whole fish collected in 1989 and 1990 from several locations on the lower Yakima River and within irrigation returns. Although primarily focused on edible tissue, WSPMP also analyzed a few whole fish samples statewide; 230-250 ug/Kg toxaphene was detected in three whole largescale sucker samples from the Yakima River near Granger in 1995 (Davis et al., 1998). A later USGS NAWQA study of the Central Columbia Plateau employed detection limits of 200 ug/Kg and could not see evidence of toxaphene in whole fish (Munn and Gruber, 1997).

Because the ecological significance of finding toxaphene in whole fish was unclear and human health not clearly implicated, these results did not trigger follow-up actions. The DDT and dieldrin problem in the lower Yakima overshadowed other toxics of possible concern (e.g., Joy and Patterson, 1997; Fuhrer et al., 2004). A summary of the historical fish tissue data for the Yakima River can be found in Johnson et al. (2007).

303(d) Listings

Washington currently has only three 303(d) water quality limited listings for toxaphene, based on the WSTMP fish fillet samples from Meridian Lake, Vancouver Lake, and the lower Snake River in 2004 and 2006 (see Table 6). Although Queets River Chinook salmon marginally exceeded the criterion in 2004, the river was not listed because this is a migratory species. TMDLs - that now include toxaphene - are currently underway in the Walla Walla River and Yakima River, which removes them from the 303(d) list.

Ecology will update the 303(d) list in 2012 and include the toxaphene data from the WSTMP 2008-10 fish samples. SPMD data are not used for 303(d) listing purposes.

Conclusions

Recent improvements in analyzing toxaphene have shown exceedances of aquatic life and human health water quality criteria in certain Washington freshwater environments. Eastern Washington rivers and streams in agriculture basins appear to be the areas particularly affected, with a circumstantial link to toxaphene's historical use for parasite control on livestock. A data gap exists on toxaphene sources to the Snake River.

A recent investigation of lakes treated with toxaphene in the mid-to-late 1960s to eradicate undesirable fish species failed to detect evidence of its persistence in the water column. However, toxaphene is detectable in fish from a number of Washington lakes (treated and untreated) and rivers. Long-range global atmospheric transport may be an important source in those instances where the level of contamination is relatively low.

Recommendations

As a result of this study, the following recommendations are made:

- An investigation should be conducted to identify toxaphene sources to Snake River fish. One source, Deadman Creek, has already been identified. This effort should assess the relative importance of bottom sediments as a reservoir of contamination.

- Feedlots, dairies, and other livestock operations with runoff to Sulphur Creek Wasteway, Granger Drain, Moxee Drain, and Wilson Creek should be evaluated as potential sources of toxaphene to the Yakima River. Other tributaries, irrigation returns, and storm drains to the lower Yakima River should be monitored for toxaphene using low-level methods.

- Identify and eliminate the source of toxaphene to Pine Creek in the Walla Walla basin.

- Future surface water studies in Washington that attempt to identify chemicals of potential concern for aquatic life or fish consumers should analyze toxaphene at aquatic life or human health criteria levels, as appropriate.

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Appendix.
Glossary, Acronyms, and Abbreviations

Glossary

Clean Water Act: A federal act passed in 1972 that contains provisions to restore and maintain the quality of the nation's waters. Section 303(d) of the Clean Water Act establishes the TMDL program.

Stormwater: The portion of precipitation that does not naturally percolate into the ground or evaporate but instead runs off roads, pavement, and roofs during rainfall or snowmelt. Stormwater can also come from hard or saturated grass surfaces such as lawns, pastures, playfields, and from gravel roads and parking lots.

Total Maximum Daily Load (TMDL): Water cleanup plan. A distribution of a substance in a waterbody designed to protect it from not meeting (exceeding) water quality standards. A TMDL is equal to the sum of all of the following: (1) individual wasteload allocations for point sources, (2) the load allocations for nonpoint sources, (3) the contribution of natural sources, and (4) a Margin of Safety to allow for uncertainty in the wasteload determination. A reserve for future growth is also generally provided.

303(d) list: Section 303(d) of the federal Clean Water Act requires Washington State to periodically prepare a list of all surface waters in the state for which beneficial uses of the water – such as for drinking, recreation, aquatic habitat, and industrial use – are impaired by pollutants. These are water quality-limited estuaries, lakes, and streams that fall short of state surface water quality standards and are not expected to improve within the next two years.

Acronyms and Abbreviations

DDT	dichlorodiphenyl trichloroethylene
EAP	Environmental Assessment Program
Ecology	Washington State Department of Ecology
EIM	Environmental Information Management database
EPA	U.S. Environmental Protection Agency
N	number of samples
NTR	National Toxics Rule
PCB	polychlorinated biphenyls
RM	River mile
SPMD	Semipermeable Membrane Device
TMDL	(See Glossary above)
USGS	U.S. Geological Survey
WAC	Washington Administrative Code
WDFW	Washington Department of Fish and Wildlife
WDOH	Washington State Department of Health
WSPMP	Washington State Pesticide Monitoring Program
WSTMP	Washington State Toxics Monitoring Program

Units of Measurement

ng/L	nanograms per liter (parts per trillion)
ug/Kg	micrograms per kilogram (parts per billion)