

# Verifying 303(d) DDT/DDE and Dieldrin Listings for the Upper Yakima River

#### Abstract

Two reaches of the upper Yakima River are currently on the 1998 303(d) List for Washington State as being water quality limited for fish tissue: the Yakima Canyon for dieldrin and p,p'-DDE, and the Cle Elum area for p,p'-DDE. The listing for dieldrin was based on a single excursion in an anadromous species of fish collected in 1985. The purpose of this 1999 study was to determine if the listing is still appropriate.

During October 1999, fish were collected at two sites: above Cle Elum and at Wymer in the Yakima Canyon. Predatory and bottom fish species were collected at each site. Composite fish muscle tissue samples were analyzed for dieldrin, DDT, and DDT breakdown products. Fish contaminant levels were compared to Washington State human health screening guidelines established under the National Toxics Rule. Samples from the Wymer site exceeded the screening levels for dieldrin, as did the rainbow trout composite sample for total DDT. One whitefish composite sample of four fish from Cle Elum exceeded the screening level for dieldrin.

Based on this information, it is recommended that the upper Yakima River (WRIA 39) remain on the 303(d) List as being water quality limited for fish tissue, specifically for dieldrin (WRIA 39-1010, 39-1060) and total DDT (WRIA 39-1010).

#### Introduction

The upper Yakima River begins at river mile 116.3, where the Naches River enters the Yakima River, and continues upstream to its headwaters above river mile 214.5. Two reaches of the upper Yakima River are on the 1998 303(d) listing for Washington State as being water quality limited for fish tissue: the Yakima Canyon (Wymer and Roza Dam) for dieldrin and p,p'-DDE, and the Cle Elum area for p,p'-DDE (Table 1). This listing is based on data collected by the Washington State Department of Ecology (Johnson et al., 1986).

There is some question as to how appropriate it is to list the upper Yakima River on the 303(d) List for dieldrin. Although dieldrin was found in water samples in tributaries to the upper basin of the Yakima River (unpublished 1999 TMDL study), the 1998 listing is based upon one individual fish sample of an anadromous species collected in 1985. In addition, it is not clear if

Location	Waterbody Number	Species		Date	<i>p,p</i> '- DDE	Dieldrin
Cle Elum	WA-39-1030	Mountain Whitefish		8/19/85	90	<20
		Rainbow Trout	3	8/19/85	<20	<20
		Crayfish	10	8/19/85	<20	<20
Wymer	WA-39-1010	Mountain Whitefish	3	8/19/85	150	<20
		Bridgelip Sucker	2	8/19/85	60	<20
		Northern Pike Minnow/ "squawfish"	2	8/19/85	190	<20
		Rainbow Trout	3	9/23/85	30	<20
		Crayfish	16	9/12/85	<20	<20
Roza Dam	WA-39-1010	Spring Chinook	2	5/21/85	390	<20
		Spring Chinook	1	5/21/85	50	20
		Spring Chinook	1	7/23/85	20	<20
		Spring Chinook	1	7/23/85	30	<20
NTR criteria used for 303(d) listing						0.65
Manchester Laboratory detection limit					3.6	?

Table 1. Data used to list the upper Yakima River on the 303(d) List for pesticides in fish (muscle fillets). (Adopted from Johnson et al., 1986) [µg/kg, wet weight: ppb].

n = number of individuals in composite sample

bold = detected values

the laboratory reporting limit was sufficiently low to make comparisons to the standard (Johnson et al., 1986). The target detection limit for dieldrin was  $20 \,\mu\text{g/kg}$ . There is also a question about whether dieldrin is present after 14 years.

The purpose of this study was to verify the 303(d) listings for dieldrin and DDT/DDE in fish tissue. Samples were collected at two stations (Table 2, Figures 1, 2). The sampling sites were planned to coincide where fish had been previously collected (Johnson et al., 1986; Rinella et al., 1992). But due to water conditions and boat access restrictions, the 1999 Cle Elum site was located above the Cle Elum River and the town of Cle Elum. The 1999 Cle Elum sampling site was located in a different reach (WRIA 39-1060) than the site from the 1985 study (WRIA 39-1030). The dividing line is the Cle Elum River.

Target species were based on previous collections and availability. A predator fish and a bottom fish were selected at each site (Table 2). Common fish species present at the Cle Elum site are rainbow trout and mountain whitefish. In the Yakima Canyon (vicinity of Umtanum) mountain whitefish, largescale sucker, bridgelip sucker, and rainbow trout are commonly found (Cuffney et al., 1996).



# Figure 1. Upper Yakima River study area.



Figure 2. Yakima River fish sampling locations at Cle Elum and Wymer.

Table 2. Upper Yakima River 303(d) verification sampling location and fish species collected during October 1999.

Location / Waterbody number	Latitude*	Longitude*	Species	Method
Yakima River, Wymer WRIA 39-1010	ymer 10 46 48.832 120 27.032 Bridgelip Sucker Rainbow Trout		Electroshocking	
Yakima River, Cle Elum WRIA 39-1060	47 10.576	121 02.409	Mountain Whitefish Rainbow Trout	Electroshocking

\*Deg./Min. NAD 83

WRIA - Water resource inventory area

# Sampling Methods

#### Fish Collection and Processing

Sample collection and processing followed U.S. Environmental Protection Agency (EPA) recommendations for fish sampling and analysis (EPA 1993). A scientific collection permit was obtained from the Washington State Department of Fish and Wildlife (#99-297).

Fish were collected using a Smith Root Model SR16 electroshocking boat. The goal was to collect enough fish to meet the minimum criteria for consideration on the 303(d) List. A listing requires an excursion in a composite of at least five fish, or at least two excursions in single–fish samples. The criteria are calculated for human health impacts based on EPA bioconcentration factors established under the National Toxics Rule (40 CFR Part 131).

An attempt was made to collect fish of approximately the same size for each species. Collection site locations were recorded with a global positioning system. The collecting site extended from the location given (latitude, longitude) to approximately 0.8 km (0.5 mile) upstream.

Fish weight and length were recorded in the field (Appendix A). Each fish was wrapped in aluminum foil (dull side in contact with the fish). The collected fish were then placed on ice and transported to the laboratory intact.

#### Composite Tissue Samples

Each composite sample consisted of muscle fillets from five to ten individual fish. One whitefish sample contained only four fish. Fillets were prepared by removing the scales, and resecting the entire skin-on fillets on both sides from the gill arch to the caudal peduncle. Fillets included dark muscle along the lateral line and belly flap fat. Care was taken to avoid rupturing or including contents of the gut cavity with the fillet.

Preparation of composite tissue samples were consistent with EPA recommendations for chemical contaminants in fish (EPA, 1993). Tissues were homogenized with three passes through a Kitchen Aide® food grinder. Ground tissue was thoroughly mixed following each pass through the grinder. Fillets were homogenized individually, then equal weight aliquots of the homogenized fillet were composited.

All equipment used for tissue preparation was thoroughly washed with Liquinox® detergent, then rinsed in hot water, de-ionized water, pesticide-grade acetone, and pesticide-grade hexane. This decontamination procedure was repeated between processing of each composite sample. Fully homogenized tissues were stored frozen ( $-20^{\circ}$ C) in 8-oz. glass jars with Teflon lid liners certified for trace organic analysis.

# **Analytical Methods**

All tissue samples were analyzed at the EPA/Ecology Manchester Laboratory using EPA SW-846 Method 8081, Organochlorine Pesticides by Gas Chromatography. Percent lipids were gravimetrically determined (EPA 608.5).

# **Quality Control Procedures**

A replicate sample, split sample, and a matrix-spiked sample were analyzed. There was a large difference in the results of the replicate sample (Table 3). This gives an indication of the variability of whitefish composite samples. The relative percent difference of the replicate sample ranged from 14% to 146%. There was good agreement between results of the sample and its split. This gives an indication of the variability within a composite sample. The relative percent difference ranged from 0.0% to 21%. Matrix spike recovery was within acceptable limits, and no analytes were detected in the laboratory method blank.

The laboratory case narrative for the analytical work is found in Appendix B.

# **Results and Discussion**

Appendix A contains data on individual fish used in composite samples. Table 3 provides a summary of the analytical results. Complete analytical results are available upon request.

There was one excursion of the recommended Washington State human health screening levels at the upper Yakima River site above Cle Elum. The replicate whitefish sample exceeded the screening level for dieldrin (0.65  $\mu$ g/kg) in fish muscle tissue. This composite was composed of only four fish instead of the recommended minimum of five fish for determining 303(d) listing. Although the whitefish replicate (2) did not meet the minimum composite number of fish recommended for 303(d) listing, neither did any of the composite samples collected in 1985 for the initial 303(d) listing (Johnson et al., 1996). The other Cle Elum whitefish sample had a

Location/fish		0/ linida	Dialdrin	4,4'-	4,4'-	4,4'-	Total
		% lipids	Dielarin	DDE	DDD	DDT	DDT
Cle Elum site							
Whitefish (1)	5	3.9	0.24	J 2.8	0.41	0.48	3.7
Whitefish replicate (2)	4	4.5	1.3	18	1.1	2.7	21
Rainbow trout	5	1.8	0.38	1.2	0.17 J	0.21 J	1.6
Wymer site							
Bridgelip sucker	7	2.7	1.5	14	2.0	4.6	21
Rainbow trout	9	2.7	2.1	28	1.4	2.7	32
*RB trout split	9	*2.3	*1.7	*28	*1.3	*2.5	*32
RB trout average		2.5	1.9	28	1.4	2.6	32
Lab method blank	-	-	0.48	U 0.48	U 0.48 U	U 0.48 U	
WA screening level (tissue)	-	-	0.65	32	45	32	32

Table 3. Yakima River 303(d) verification results and Washington screening levels ( $\mu$ g/kg wet weight) of composite fish tissue samples.

# - number of fish used in composite sample.

U - analyte was not detected at or above the reported value.

J - analyte was positively identified. The associated numerical result is an estimate.

\*- split sample.

Values in **bold** are above screening guidelines.

detectable level of dieldrin, but at less than one third of the screening level. The dieldrin concentration in the rainbow trout composite sample also did not exceed the screening level for dieldrin.

DDT, DDE, and DDD were detected in all three fish composite samples at the Cle Elum site. Total DDT and metabolite values for the three samples collected at the Cle Elum site were below NTR screening levels.

DDT metabolites and dieldrin were detected in all samples collected at the downstream site at Wymer. The trout composite fish sample collected from the Yakima River at the Wymer site equaled the screening levels for total DDT, and both the trout and bridgelip sucker sample exceeded the screening levels for dieldrin. Lower levels of dieldrin and DDE in the bridgelip sucker sample, compared to the trout sample, may be related to diet. Suckers feed lower on the food chain than predatory fish such as trout, so bioconcentration of contaminants is probably lower in the trout. The bridgelip sucker is believed to feed primarily on algae scraped from rocks and, to some extent, on aquatic insect larvae and crustaceans (Wydoski and Whitney 1979).

Due to the difficulty of obtaining fish, the replicate composite sample of whitefish (whitefish 2) collected at the Cle Elum site was not ideal. The size and number of fish used in the two whitefish composite samples were different. One of the samples consisted of five fish while the

other contained four fish. The average weight of the fish in the samples was 190 mg and 277 mg (Appendix A). The larger size of fish in sample 2 may account for the difference in the contaminant levels observed. One fish was twice the weight of any other. Composite sample whitefish 2 had higher levels of all analytes. Larger fish tend to be older fish, and thus have had a longer time to bioaccumulate contaminants.

Fish collected at Cle Elum generally had lower levels of contaminants than fish collected at Wymer. This is consistent with the results from the 1985 study (Johnson et al 1986). Above the Cle Elum site the watershed is primarily forested, while the watershed below Cle Elum and above Wymer is primarily devoted to agriculture.

It is difficult to adequately compare the results from fish collected in this study with those collected in 1985 (Johnson et al., 1986). The Cle Elum collection site was different. Fish collected in 1999 were obtained approximately 4 kilometers (2.4 miles) upstream of the 1985 study, above the Cle Elum River and the town of Cle Elum. The detection limits for dieldrin, DDT, and DDT metabolites in the 1985 study were above the levels of analytes detected in the 1999 fish samples. The detection limits for the Johnson et al., 1985 study varied between 20 and 40  $\mu$ g/kg.

DDE was the only DDT metabolite detected at or above the detection limits in the 1985 study. Levels of 4,4'-DDE in fish fillets collected in 1999 were lower than from fish collected in 1985 (Table 4). Four of the six composite samples collected at the two upper Yakima sites and analyzed in 1985 exceeded the screening guidelines for 4,4'-DDE ( $32 \mu g/kg$ ), while none of the samples collected in 1999 exceeded the guidelines.

1985 study (Johnson et al., 1986)			1999 study			
Location/fish	#	4,4'-DDE	Location/fish		4,4'-DDE	
Cle Elum site			Cle Elum site			
Whitefish	3	90	Whitefish (1)	5	2.8	
-	-	-	Whitefish replicate (2)	4	18	
Rainbow trout	3	<20 u	Rainbow trout	5	1.2	
Wymer site			Wymer site			
Bridgelip sucker	2	60	Bridgelip sucker	7	14	
Whitefish	3	150	Rainbow trout	9	28	
-	-	-	*RB trout split	9	*28	
Rainbow trout	3	30				
Northern pike minnow	2	190				

Table 4. Comparison of DDE levels in fish fillets collected in 1985 and 1999 from the Yakima River ( $\mu g/kg$ ).

# - number of fish used in composite sample.

\* - split sample.

Values in **bold** exceeded recommended guideline.

## Conclusion

This study provides evidence that dieldrin, DDT, and DDT breakdown products (DDE, DDD) are still present in fish from the upper Yakima River. Although levels in these fish tissue composites were lower than what has previously been detected, levels in fish tissue continue to exceed the Washington State human health screening levels.

#### Recommendations

- It is recommended that the upper Yakima River (WRIA 39) remain on the 303(d) List as water quality limited for fish tissue, specifically in the Yakima Canyon reach (WRIA 39-1010) for dieldrin and total DDT, and above Cle Elum (WRIA 39-1060) for dieldrin. Samples were not collected from the reach immediately below Cle Elum.
- The 303(d) listing for DDE is no longer warranted.
- Future fish tissue monitoring for dieldrin, DDT, and its breakdown products is recommended. The lower main stem of the Yakima River is scheduled for a pesticide study in 2007, according to the report *A Suspended Sediment and DDT Total Maximum Load Evaluation Report for the Yakima River* (Joy and Patterson, 1997). Fish from the upper Yakima River should also be included in the pesticide sampling effort.

#### References

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## Contacts

Dale Norton	Washington State Department of Ecology
	Environmental Assessment Program
	Watershed Ecology Section
	(360) 407-6765

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Appendix A

Fish Data

			Total	Wet
			Length	Weight
Composite Sample (lab number)	#	Sex	(mm)	(mg)
Cle Elum				
Whitefish 1 (420861)	2	F	290	216
	5	М	240	140
	7	F	225	100
	6	F	305	277
	9	М	294	218
Average			271	190
Whitefish 2 (4208064)	1	F	298	231
	3	F	253	137
	4	F	376	565
	8	F	257	175
Average			296	277
Rainbow Trout (4208065)	1	F	330	356
	2	F	428	732
	3	F	247	142
	4	?	467	1262
	5	F	285	220
Average			351	542
Wymer		-		-
Rainbow Trout (4208062)	1	F	343	370
(split sample = $4208063$ )	2	М	403	675
	3	Μ	430	799
	4	Μ	236	154
	5	F	372	571
	6	F	400	605
	7	Μ	386	574
	8	F	317	329
	9	F	270	227
Average			351	478
Bridgelip Sucker (4208060)	1	F	429	981
	2	F	461	991
	3	F	434	1007
	4	?	435	1154
	5	F	419	886
	6	F	416	868
	7	F	386	669
Average			426	937

Appendix A. Yakima River composite fish (fillet) samples, corresponding lab number, and individual length and weight measurements.

#### Appendix B

Case narratives for laboratory analyses

Appendix B is available only in printed copies of this report. See page 10 for ordering information.