Water Body No.: WA-CR-9010
(Segment No.: 26-00-04)

90-e46

WASHINGTON STATE DEPARTMENT OF ECOLOGY
ENVIRONMENTAL INVESTIGATIONS AND LABORATORY SERVICES
TOXICS INVESTIGATIONS/GROUND WATER MONITORING SECTION

TECHNICAL MEMORANDUM

February 8, 1990

TO: John Arnquist, Carl Nuechterlein, and Phil Kauzloric

THROUGH: Bill Yake BY

FROM: Art Johnson AJ

SUBJECT: Results of Screen for Dioxin and Related Compounds in Lake Roosevelt Sportfish

Concerns regarding potential dioxin contamination of fish in Lake Roosevelt, the Columbia River reservoir behind Grand Coulee Dam, were raised to the Water Quality Program (WQP) of the Department of Ecology (Ecology) by the Colville Tribes and the Lake Roosevelt Water Quality Group. In response to these concerns, WQP requested that Environmental Investigations conduct a screening survey for polychlorinated dioxins (referred to as dioxins), and polychlorinated furans (furans) in Lake Roosevelt sportfish. This survey was combined with a previously planned effort to re-evaluate mercury concentrations in edible tissues of walleye (Stizostedion vitreum) and white sturgeon (Acipenser transmontanus), two of the lake's major sportfish species. Preliminary results for mercury, showing continued low levels in both species, were described in an earlier memorandum (Johnson, 1989). The present report provides the results for dioxins and furans. A report giving a more detailed discussion of the mercury in fish issue is planned for later in the year.

The concern about dioxins/furans contamination of Lake Roosevelt fish stemmed from an Environment Canada report (Mah et al., 1989) of elevated concentrations of 2,3,7,8-tetrachlorodibenzodioxin (TCDD) and 2,3,7,8-tetrachlorodibenzofuran (TCDF) in lake whitefish (Coregonus clupeaformis) collected below the Celgar bleached Kraft pulp mill in Castelgar, British Columbia. This mill discharges wastewater to the Columbia River approximately 30 miles above the Canada-U.S. border. Based on results of the Environment Canada study, fish tissue contamination below the Celgar mill ranked among the top four of the 10 inland mills investigated. The Health Protection Branch, Health and Welfare, Ottawa, reviewed the Celgar data and concluded "It would be advisable to restrict consumption of lake whitefish from this area (up to 7 kilometers below the outfall; location of furthest downstream sample) to no more than 40 grams per week" (Kirkpatrick 1989). Although there has been a news release to this effect, no official health advisory was issued.

The B.C. Ministry of Environment is currently analyzing an additional 18 fish samples collected in the vicinity of Celgar last November (Crozier, personal communication). Also, Environment Canada is planning a study later this year of chlorinated organics in the Columbia River, concentrating on the border reach. This study may include analysis of dioxins/furans in fish tissue (Tuominen, personal communication). The Environment Canada study will be coordinated with the Columbia River Integrated Environmental Sampling Program, a cooperative assessment of conditions in the river being undertaken by the Ministry of Environment, B.C. Hydro, and industry.

The Celgar mill has been discharging untreated effluent to the Columbia since it began operation in 1961. The discovery of elevated TCDD and TCDF below Celgar and other B.C. pulp mills brought about new regulations under the Canadian Environmental Protection Act to strictly control the discharge of dioxins and furans. Celgar is presently seeking government approval to double the mill's production. They are proposing to modify their production process to include oxygen delignification and maximum chlorine dioxide substitution of chlorine. The plan provides for wastewater treatment consistent with requirements for U.S. mills. Currently, these improvements are expected to be in place by 1992; discharge limits for chlorinated organics are required to be met by 1994 (Crozier, personal communication). TCDD concentrations in the effluent are expected to be reduced to below detectable levels (10 - 20 parts per quadrillon [ppq]).

METHODS

Figure 1 shows where Ecology collected fish samples in Lake Roosevelt for dioxins/furans analysis. Because this survey constituted the initial screen to determine if these compounds were detectable in Lake Roosevelt fish, the analysis was limited to two walleye and two sturgeon taken from the upper reaches of the lake near Kettle Falls. The walleye were collected July 18, 1989, by electroshocking. These samples were obtained with the help of Tim Peone, Del Brown, and Milo Thatcher of the Upper Columbia United Tribes, Fisheries Research Center, through the courtesy of Dr. Allan Scholz of Eastern Washington University. The ages of the walleye, determined from scale samples read by Tim Peone, were 5+ and 6+ years.

Sturgeon samples consisted of heads from two fish caught in the sport fishery at Marcus Island between June and July 1989. The heads were severed in the field so as to include a portion of the anterior muscle tissue. Gig Lebret of the National Park Service arranged for collection of these samples by Jim Meskan of the Department of Wildlife. Based on examination of otolith and pectoral spine cross-sections by Eric Volk, Washington Department of Fisheries, the age of both sturgeon was estimated to be 12 - 13 years.

All samples were wrapped in aluminum foil in the field and frozen for later dissection at the Ecology Manchester Laboratory. Walleye tissue samples consisted of skinless fillets. Muscle tissue removed from the sturgeon heads did not include skin or tissues exposed in the field.

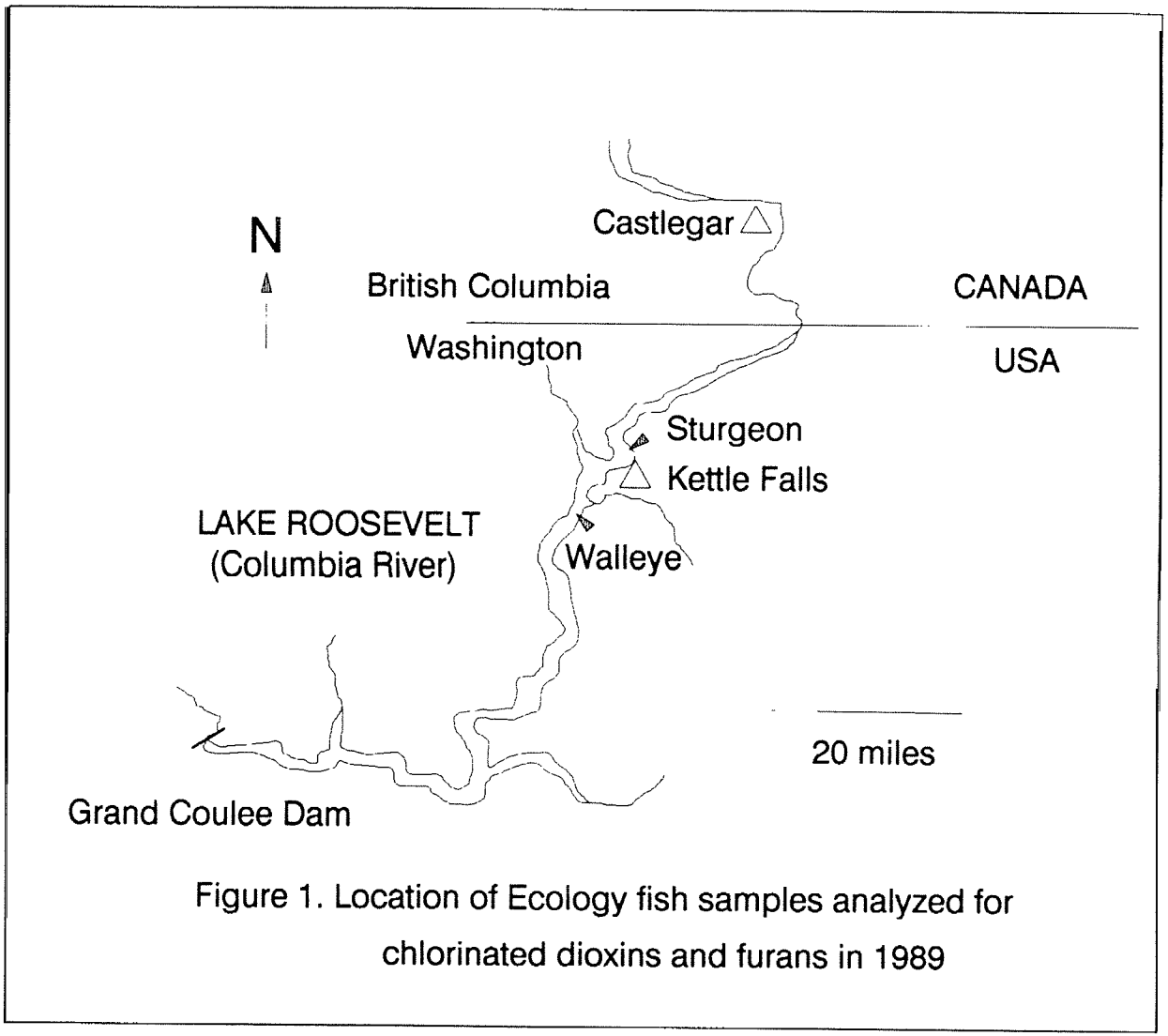

Figure 1. Location of Ecology fish samples analyzed for chlorinated dioxins and furans in 1989

The dissection was done on glass plates using stainless steel instruments. All surfaces coming in contact with the tissue samples were previously washed with Liqui-Nox detergent followed by sequential rinses with de-ionized water, dilute nitric acid, de-ionized water, and pesticide-grade acetone. Sample containers were glass with teflon lid liners. These were obtained from I-Chem, Hayward, California (series 300) and had been specially cleaned for trace organics analysis.

The dioxins/furans analysis was done by Triangle Laboratories, Inc., Research Triangle Park, North Carolina, following a modification of EPA high resolution GC/MS method 8290 (EPA, 1986). The results were reviewed for qualitative and quantitative accuracy by Cliff Kirchmer of the Ecology Quality Assurance Section (see Appendix A). The analysis was concluded to be accurate.

RESULTS

The results are summarized in Table 1. The data are reported in units of parts per trillion (ppt) of wet tissue. Comparable concentrations of TCDD and TCDF were detected in one sturgeon (2.2 - 2.6 ppt TCDD and 221 - 321 ppt TCDF in duplicate analyses) and in one walleye (4.0 ppt TCDD and 326 ppt TCDF). The other sturgeon and walleye had TCDD concentrations at or below detection limits (0.1 ppt) and only trace amounts of TCDF (3.9 and 8.9 ppt, respectively). Other dioxins and furans (penta through octa) were also detected in one or more samples. Concentrations of these congeners was generally 2 ppt or less, except for octachlorodibenzodioxin (OCDD), hexachlorodibenzofuran (HxCDF), and heptachlorodibenzofuran (HpCDF) which were in the approximate range of 3 - 17 ppt in some samples.

The obvious disparity between the levels of TCDD and TCDF in fish of the same species prompted a re-analysis for these two compounds. The results, shown in Table 2, confirmed the results of the original analysis. The reason for these within-species differences is not known, although it could potentially be related to movements of the fish. Both species are known to range widely in Lake Roosevelt and its tributaries (Scholz, personal communication; Setter, personal communication).

DISCUSSION

Table 3 compares the TCDD and TCDF concentrations measured in Lake Roosevelt fish with those reported by Environment Canada for fish in the vicinity of the Celgar pulp mill. The Canadian samples were also skinless fillets, but consisted of a composite of seven fish each. As shown in the table, the elevated TCDD and TCDF concentrations in two of the four Lake Roosevelt samples are approximately one-third to one-half those in the whitefish below the Celgar mill. The low to non-detectable TCDD and TCDF in the other two samples are comparable to fish collected above Celgar. Except for 13.4 ppt of pentachlorodibenzofuran (PeCDF) in one whitefish sample, other dioxins and furans were not detected by Environment Canada. This may be due to higher detection limits (3 - 15 ppt) than achieved in Ecology's survey (0.02 - 0.6 ppt).

Table 1. Chlorinated Dioxins and Furans Analysis of Ecology Muscle Tissue Samples from Lake Roosevelt Sportfish (ng/Kg, wet; ppt)

	Species:	White Sturgeon			Walleye	
	Location:	Marcus Island			off Colville River	
	Date:	3 July 89		11 June 89	18 July 89	
	Length:	58 inches		64 inches	535 mm	458 mm
	Estimated Age:	12-13 years		12-13 years	6+ years	5+ years
	Sample No:	8310	8310(dup)	8305	8293	8298
TEF	Dioxins:					
1	2378-TCDD	2.2	2.6	0.1 U	0.21	4.0
0.5	12378-PeCDD	0.05 U	0.1 U	0.1 U	0.56	0.2 U
0.1	123478-HxCDD	0.03 U	0.05 U	0.1 U	0.08 U	0.1 U
0.1	123678-HxCDD	0.16*	0.18	0.08 U	0.07 U	0.4
0.1	123789-HxCDD	0.04 U	0.06 U	0.1 U	0.1 U	0.1 U
0.01	1234678-HpCDD	0.05 U	0.44	0.2 U	0.61*	0.62
0.001	OCDD	2.7	17.0	0.8 U	8.5	5.7
	Total Dioxins	5.1	20	ND	9.9	11
	Furans:					
0.1	2378-TCDF	221	321	3.9	8.9	326
0.05	12378-PeCDF	0.72	0.96	0.08 U	0.07 U	0.9
0.5	23478-PeCDF	1.3	1.9	0.09 U	0.08 U	2.4
0.1	123478-HxCDF	0.02 U	0.03 U	0.05 U	0.05 U	0.07 U
0.1	123678-HxCDF	0.02 U	0.03 U	0.05 U	0.04 U	0.06 U
0.1	234678-HxCDF	0.02 U	0.04 U	0.07 U	5.0	0.09 U
0.1	123789-HxCDF	0.03 U	0.05 U	0.1 U	0.09 U	0.1 U
0.01	1234678-HpCDF	0.02 U	0.08	0.09 U	0.09*	0.08 U
0.01	1234789-HpCDF	0.04 U	0.04 U	2.9	3.7	0.2 U
0.001	OCDF	0.07 U	0.93*	0.6 U	0.46	0.3 U
	Total Furans	223	325	6.8	18	329
	Total TEQs	25	36	0.4	1.9	38
	% Contributed by 2378-TCDD	9%	7%	0%	11%	11%

TEF = Toxicity Equivalency Factor (Barnes et al, 1989)
TEQs = Toxicity Equivalents
U = Not detected at detection limit shown
ND = Not detected
*Estimated maximum possible concentration

Table 2. Re-analysis of Lake Roosevelt Samples, January 1990
(ng/Kg, wet; ppt)

Sample No.	1st Run 2378-TCDD	2nd Run 2378-TCDD	1st Run 2378-TCDF	2nd Run 2378-TCDF
8293	0.21	1.1 U	8.9	10.3
8298	4.0	3.2	326	242
8305	0.1 U	0.3 U	3.9	4.4
8310	2.2/2.6*	5.9**	221/321*	341

U = Not detected at detection limit shown
*duplicate analysis
**estimated maximum concentration

Table 3. Summary of TCDD/TCDF Concentrations Measured in Muscle Tissue of of Upper Columbia River/Lake Roosevelt Fish (ng/Kg, wet; ppt)

Species	Location	Date	2378-TCDD	2378-TCDF
	Environment Canada Data (Mah et al, 1989)			
Largescale sucker	Columbia R.	16-18 Aug 88	2 U	2.0
Lake whitefish	ab. Celgar	16-18 Aug 88	2 U	45.4
Northern squawfish	Mill		2 U	5.9
Largescale sucker	Columbia R.	16-19 Aug 88	2 U	25.8
Lake whitefish	bw. Celgar	16-19 Aug 88	10.5	908
Lake whitefish	Mill		6.6	647
	Ecology Data			
White sturgeon	Lk. Roosevelt	3 Jul 89	2.2/2.6*	221/321*
White sturgeon	@ Marcus Is.	11 Jun 89	0.1 U	3.9
Walleye	Lk. Roosevelt	18 Jul 89	4.0	326
Walleye	off Colville R.	18 Jul 89	0.21	8.9

U = Not detected at detection limit shown
*duplicate analysis

Figure 2 summarizes the available data on distribution of TCDD and TCDF in Columbia River fish. The lower Columbia data (Lake Wallula to Skamokawa) are unpublished results from the EPA National Bioaccumulation Study (see Appendix B for complete data). EPA's analyses suggest that concentrations typical of fish in the lower parts of the river (both whole fish and fillet samples) are in the approximate range of <1 - 5 ppt for TCDD and 10 - 20 ppt for TCDF.* For a given sampling site, whole fish have consistently had higher concentrations than fillets. The highest levels have been measured in channel catfish muscle (7.9 ppt TCDD) and whole carp (56 ppt TCDD and 321 ppt TCDF) from Lake Wallula (McNary pool). The 300 mile reach of the Columbia River between Lake Roosevelt and Lake Wallula has not been sampled.

Lake Wallula, like the upper Columbia, receives bleached Kraft mill effluent, and, in both instances, this is suspected to be the primary dioxins/furans source. However, sources are not limited to pulp mills using chlorine as a bleaching agent. Other sources include oil refineries, municipal waste incinerators, metal reclamation plants, and motor vehicle exhaust. They are also present as trace impurities in PCBs, commercial herbicides (2,4-D; 2,4,5-T), and chlorinated phenols. Dioxins and furans are persistent, bioaccumulative, widely distributed in the environment, and have been detected in virtually all media.

TCDD is considered the most toxic of the dioxins and furans. It has been shown to be a carcinogen and reproductive toxin in animals. EPA has classed it as a probable human carcinogen based on animal experiments (EPA, 1985). However, results from numerous human studies provide equivocal information of health effects in humans. TCDF, the compound generally present in highest concentrations in Columbia River fish samples, has not been demonstrated to be carcinogenic in laboratory animals (Olsen, et al., 1989).

There is presently no consensus on what levels of dioxins and furans are acceptable in fish used for human consumption, and it is not known at what level these compounds begin to pose a hazard to fish and wildlife. Washington's narrative water quality standard for toxics ("concentrations shall be below those which may adversely affect characteristic water uses, cause acute or chronic conditions to the aquatic biota or adversely affect public health") applies to TCDD. In interpreting this standard, Ecology applied the EPA water quality criteria for TCDD of 0.013 ppq. The corresponding level for TCDD in fish, assuming a bioaccumulation factor from water to tissue of 5,000, is 0.07 ppt (EPA, 1984). This concentration is at the detection limits of commonly employed analytical methods. The effect of adopting this stringent standard is that water quality standards are violated whenever TCDD is detected in fish.

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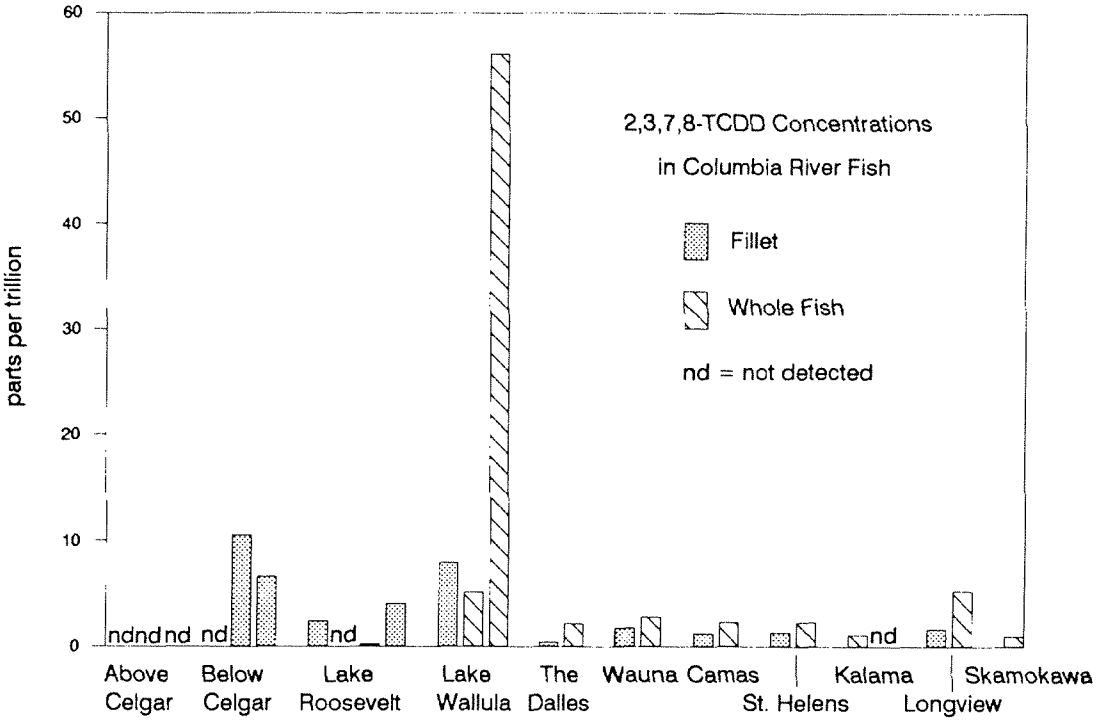
*Results of a recently published, independent survey of TCDD and TCDF in lower Columbia River fish by the Northwest Pulp and Paper Association puts the average levels of TCDD and TCDF at 0.07 - 0.31 ppt and 0.24 - 7.01 ppt, respectively, in migratory salmonids, and 0.09 - 1.68 ppt and 1.76 - 62.4 ppt, respectively, in resident fish (Keenan et al., 1990).


60
50
40
30
20
10
0
parts per trillion
2,3,7,8-TCDD Concentrations
in Columbia River Fish
Fillet
Whole Fish
nd = not detected
ndndnd nd
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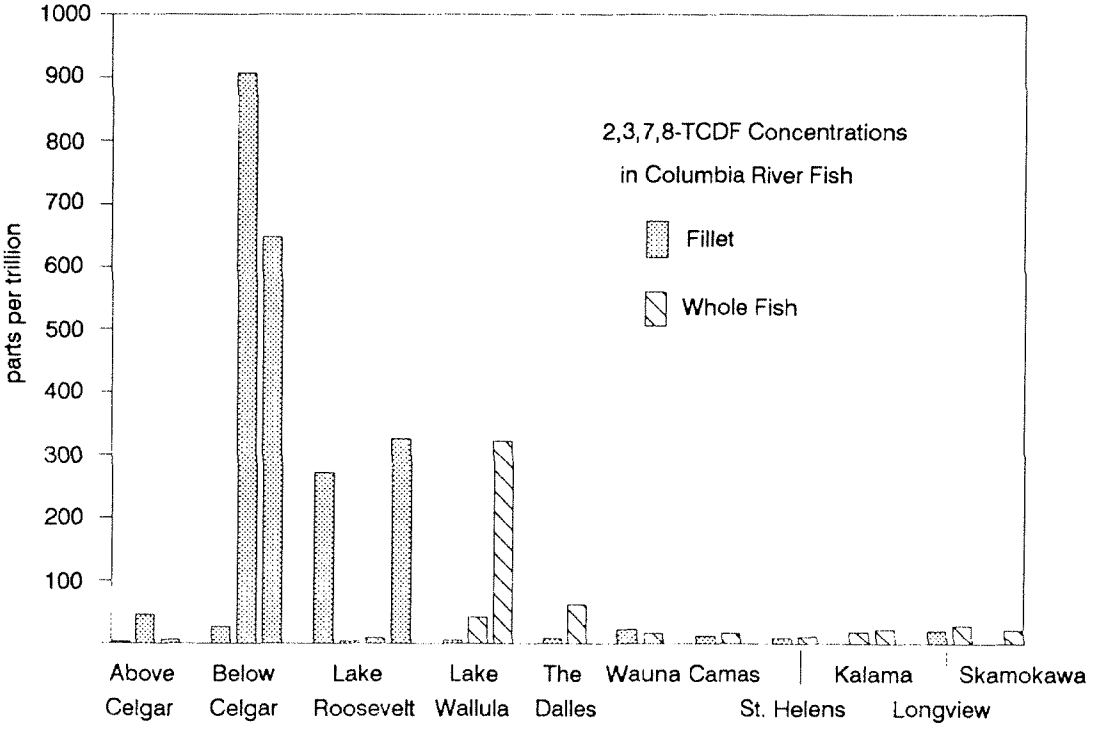
Above Celgar
Below Celgar
Lake Roosevelt
Lake Wallula
The Dalles
Wauna
Camas
St. Helens
Kalama
Longview
Skamokawa


1000
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2,3,7,8-TCDF Concentrations
in Columbia River Fish
Fillet
Whole Fish
Above Celgar
Below Celgar
Lake Roosevelt
Lake Wallula
The Dalles
Wauna
Camas
St. Helens
Kalama
Longview
Skamokawa

Figure 2

EPA's criteria is based on a health risk assessment that this level of TCDD could, theoretically, cause one cancer death per million people. World-wide, however, regulatory agencies, using other assumptions of fish consumption rates, adsorption efficiency, and cancer potency, have different criteria; EPA's is the most stringent. The range of health advisory levels that have been proposed for TCDD in fish is illustrated in Table 4. There is presently no U.S. national or Washington State health advisory level established for TCDD (or other dioxins/furans) in fish.

EPA has recommended an interim procedure for estimating risks associated with exposure to mixtures of dioxins and furans. Toxicity equivalency factors (TEFs) are used to convert concentrations of individual dioxins and furans to equivalent concentrations of 2,3,7,8-TCDD. Each of the dioxins and furans concentrations is multiplied by the appropriate TEF and results summed to give toxicity equivalents (TEQs). Table 1 contains a listing of the most recent TEFs adopted by EPA (Barnes et al., 1989) and shows resulting TEQs for the Lake Roosevelt samples. EPA cautions that the TEF approach is interim, needs additional data, and should ultimately be replaced with a more direct biological assay.

Figure 3 shows results of TEQ calculations using the Columbia River fish data in Appendix B. Examination of the figure shows that, whereas TCDD is a significant contributor (average of 45%) to the toxicity estimate in the majority of lower Columbia samples, other compounds (predominantly TCDF) contribute ninety percent or more of the toxicity estimate in the Celgar and Lake Roosevelt samples. Based on the TEQ approach, the two Lake Roosevelt samples with elevated TCDD and TCDF have a toxicity potential of 30 - 37 ppt, intermediate between those collected below Celgar and those from the lower Columbia River. The other Lake Roosevelt samples have little or no toxicity potential.

RECOMMENDATION

The Washington Department of Health (Health) has reviewed these data and concluded that the Lake Roosevelt sample size is too small, and results too equivocal to consider a health advisory on consumption of Lake Roosevelt fish. In their opinion, there is presently no reason for people fishing the lake to change their fish eating habits. Health has, however, recommended collection of additional samples for dioxins/furans analysis (R. Lorenzana and C. Sagerser, personal communication).

The Toxics Investigations/Ground Water Monitoring Section is presently designing a comprehensive survey of dioxins and furans in Lake Roosevelt fish in consultation with the Departments of Health, Wildlife, Fisheries, and other state and local agencies. Collection of the first samples is anticipated this spring. Target species will include sturgeon, walleye, and rainbow trout; bottom sediments may also be sampled. Preliminary results from analysis of initial samples should be available by June.

Table 4. Sample of Advisories for TCDD in Fish

Regulatory Authority	Advisory Level
U.S. Food and Drug Administration (1981)	>50 ppt - do not consume 25-50 ppt - consume no more than twice monthly <25 - not a concern
Health and Welfare Canada (1986)	20 ppt - tolerance
Michigan (1986)	10 ppt
New York (1987)	10 ppt
Minnesota (1985)	0.6 ppt virtually safe, dose for 1x10E-6 cancer risk

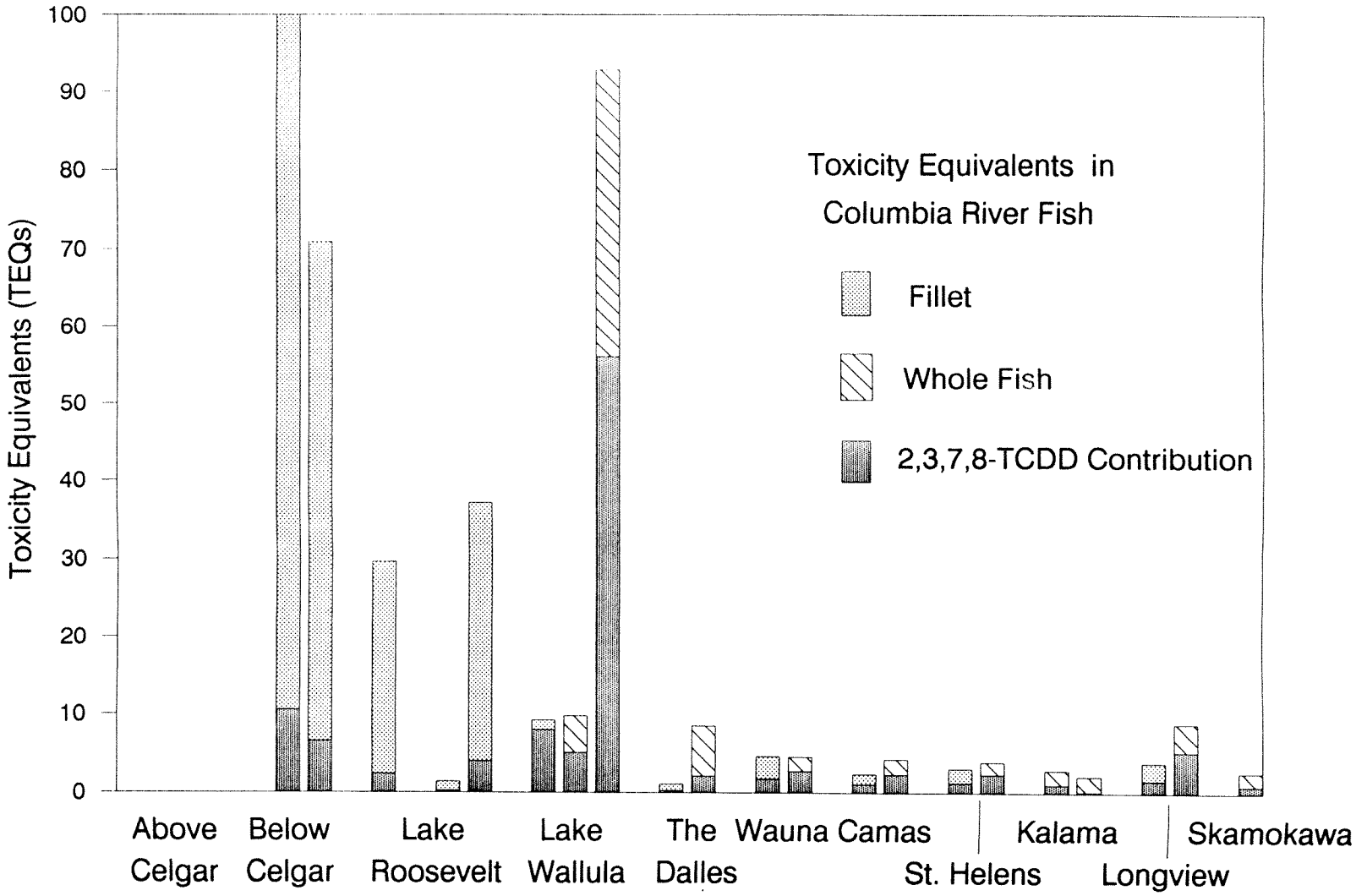
Toxicity Equivalents (TEQs)
100
90
80
70
60
50
40
30
20
10
0
Toxicity Equivalents in
Columbia River Fish
Fillet
Whole Fish
2,3,7,8-TCDD Contribution
Above Celgar
Below Celgar
Lake Roosevelt
Lake Wallula
The Dalles
Wauna
Camas
St. Helens
Kalama
Longview
Skamokawa

Figure 3

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Scholz, A. Personal communication. Eastern Washington University, Cheney, WA.

Tuominen, T.M. Personal communication. Environment Canada, Vancouver, BC.

Appendix A

WASHINGTON STATE DEPARTMENT OF ECOLOGY
ENVIRONMENTAL INVESTIGATIONS AND LABORATORY SERVICES
QUALITY ASSURANCE SECTION

December 15, 1989

TO: Bill Yake

FROM: Cliff Kirchmer

SUBJECT: Review of Lake Roosevelt PCDD/PCDF Data

As requested, I have reviewed the data package from Triangle Labs for the analyses they performed on fish samples from Lake Roosevelt.

Data were reviewed for both qualitative and quantitative accuracy. The laboratory reported high results for 2,3,7,8-TCDF on samples 8298 and 8310. Traces of 2,3,7,8-TCDF were also reported for samples 8293 and 8305. Qualitative identifications meet the criteria for positive identification. The M and M+2 ions are monitored for both 2,3,7,8-TCDF (303.9016 and 305.8987) and the internal standard C-13 labelled 2,3,7,8-TCDF (315.9419 and 317.9389). For example, the attached extracted ion chromatogram for sample 8298 clearly shows the presence of all 4 ions. The retention times for the peaks identified as 2,3,7,8-TCDF were within the expected windows based on retention times for standards. For example, the attached printout for sample 8298 shows that the retention times for unlabeled and labeled 2,3,7,7-TCDF differed by only 2 seconds. The monitored ions for 2,3,7,8-TCDF maximized at the same retention time. The ratios for the M and M+2 ions were within the expected window of 0.65-0.89, and close to the theoretical value of 0.77. The result for sample 8293 was calculated manually and the result found to be valid (there was a small difference, but this was probably due to my assuming a response factor of unity). Results for samples 8293, 8305, and 8310D were confirmed on a second column.

One problem, pointed out by the laboratory, was that the percent recoveries on the matrix spike analyses were much higher than expected. They speculate that this may be due to the fact that they spiked a different sample than the one reported. This problem does not directly affect the results for samples reported as positive for 2,3,7,8-TCDF, although it does indicate a certain lack of quality control somewhere in the process.

A final note is that if you plan on requesting follow-up analyses and you are interested only in 2,3,7,8-TCDF, you could ask for that specific analysis. They would use selected ion monitoring for that ion. The results would probably show some improvement in quantitation as compared with the method for PCDD/PCDF because of the large number of ions that need to be monitored the latter. The reduced complexity of the analysis also makes qualitative identification easier. Attached is a copy of the parameters they analyze. It also includes TCDD/DF, which is simpler (and probably cheaper) than PCDD/PCDF. I can discuss this further with you if you're interested.

CJK:cjk

Appendix B. Summary of Columbia River TCDD/TCDF Data (ng/Kg, wet; ppt)

Location	Species	Tissue	2378-TCDD	2378-TCDF	TEQs
Environment Canada, 1988 Samples (Mah et al, 1989)					
Above Castlegar, BC	Largescale sucker	Muscle	2 U	2.0	0.2
	Lake whitefish	"	2 U	45.4	4.5
	Northern squawfish	"	2 U	5.9	0.6
Below Castlegar, BC	Largescale sucker	Muscle	2 U	25.8	2.6
	Lake whitefish	"	10.5	908	100
	Lake whitefish	"	6.6	647	71
Washington Department of Ecology, 1989 Samples					
Lake Roosevelt	White sturgeon	Muscle	2.2	221	24.4
	White sturgeon (dup)	"	2.6	321	34.9
		duplicate mean =	2.4	271	29.6
	White sturgeon	Muscle	0.1 U	3.9	0.4
	Walleye	"	0.21	8.9	1.4
	Walleye	"	4.0	326	37.0
EPA National Bioaccumulation Program, 1987/1989 Samples					
Lake Wallula	Channel catfish	Muscle	7.92	4.97	9.2
	Largescale sucker	Whole	5.12	41.78	9.71
	Carp	"	56.02	320.69	92.89
The Dalles	White sturgeon	Fillet	0.36	8.02	1.17
	White sturgeon	Whole	2.14	61.58	8.5
Wauna	Northern Squawfish	Fillet	1.73	21.63	4.57
	Largescale sucker	Whole	2.78	16.39	4.55
Camas	Northern Squawfish	Fillet	1.14	11.95	2.46
	Largescale sucker	Whole	2.28	15.95	4.22
St. Helens	Northern Squawfish	Fillet	1.28	9.03	3.07
	Largescale sucker	Whole	2.57	11.38	4.46
	Largescale sucker (dup)	"	2.01	10.27	3.4
		duplicate mean =	2.29	10.83	3.93
Kalama	White sturgeon	Whole	1.06	17.75	2.84
	White sturgeon	"	1.75 U	22.15	2.22
Longview	Northern Squawfish	Fillet	1.48	20.12	3.82
	Northern Squawfish (dup)	"	1.75	20.73	3.85
		duplicate mean =	1.62	20.43	3.84
	Largescale sucker	Whole	5.23	28.34	8.78
Skamokawa	White sturgeon	Whole	2.07 U	22.05	2.21
	White sturgeon (dup)	"	0.88	20.94	3.01
		duplicate mean =	0.96	21.50	2.61

TEQs = Toxicity Equivalents
U = not detected at detection limit shown