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Transboundary Metal Pollution of the Columbia River (Franklin D. Roosevelt Lake)

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Franklin D. Roosevelt Lake in northeast Washington State is the Columbia River reservoir formed by Grand Coulee Dam in 1941. With a full pool length of 151 miles and an average depth of 118 feet, it is the sixth largest reservoir in the United States. The lake and much of its shoreline constitute the Coulee Dam National Recreation Area which is managed by the National Park Service.

Historically, the Washington Department of Ecology (WDOE) has received sporatic water quality alerts from the U.S. Geological Survey warning that cadmium and mercury concentrations at their monitoring station in Northport on the upper reaches of the lake exceeded U.S. Environmental Protection Agency (USEPA) water quality criteria. Independent measurements of high metals concentrations in samples of the lake's fish collected during WDOE's Basic Water Monitoring Program (Hopkins et al. 1985), and the U.S. Fish and Wildlife Service (USFWS) National Contaminant Biomonitoring Program (Lowe et al. 1985) prompted WDOE to survey the lake in 1986 to determine the extent of contamination. This paper presents data from analysis of Lake Roosevelt bottom sediments. Complete survey results can be found in Johnson et al. (1988).

MATERIALS AND METHODS

Figure 1 shows the study area. Sediment samples were collected with a 0.1m^2 stainless steel van Veen grab along a longitudinal transect from the international border to Grand Coulee Dam and at the mouths of the Colville, Kettle, Spokane, and Sanpoil Rivers, the lake's major tributaries. A 2-inch gravity core with plexiglass liner was used to obtain a sediment core off Frenchman Point Rocks near Kettle Falls, approximately 50 miles below the border. The British Columbia (BC) Ministry of Environment provided sediments collected with an Ekman grab from Lower Arrow Lake, the Columbia River reservoir above Lake Roosevelt.

Surface sediment samples (top 2-cm layer only) were transferred in the field to stainless steel beakers and homogenized by stirring with stainless steel spoons. Subsamples were taken for analysis

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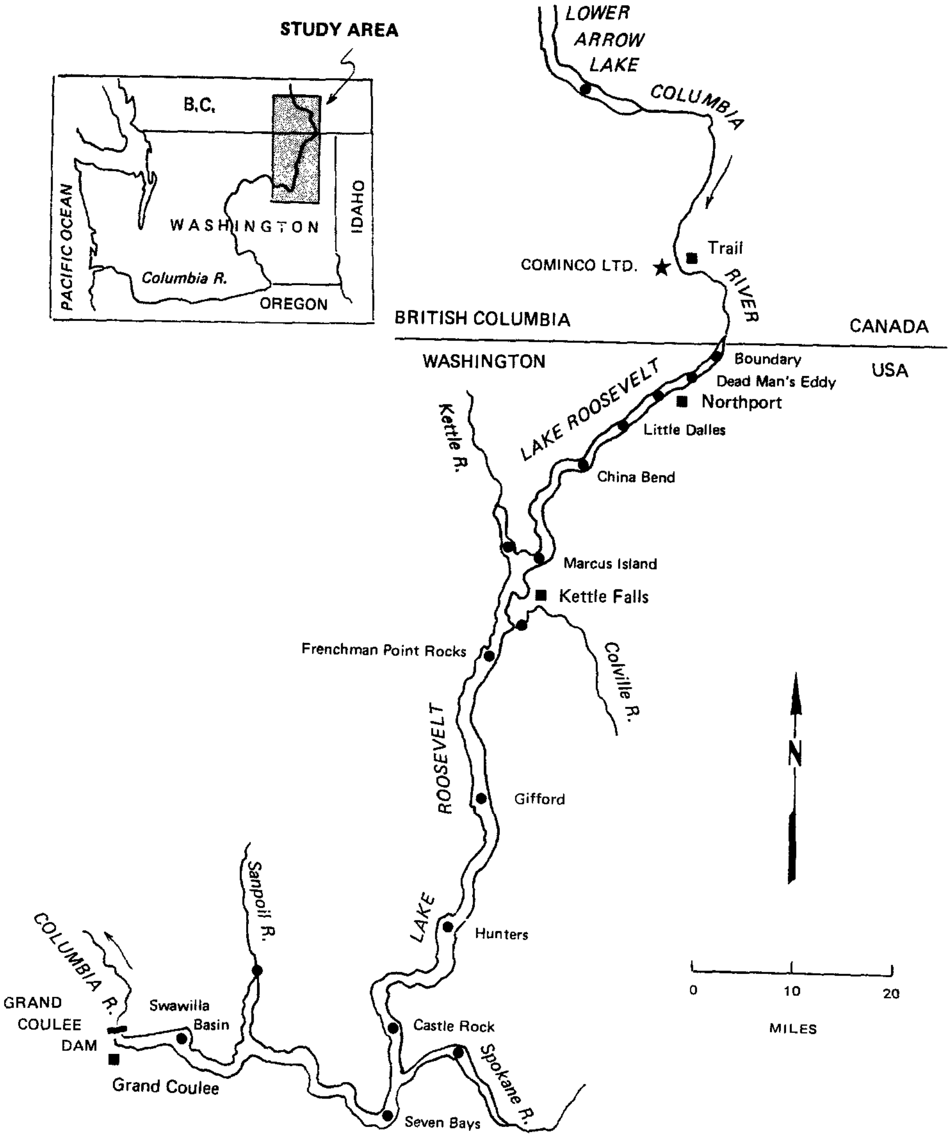

Figure 1. Sampling locations

of zinc, copper, lead, arsenic, cadmium, mercury, grain size, and total organic carbon. The core was extruded from the liner and cut into 5 cm increments. The outer surface of each core segment was parred away with a stainless spatula and discarded. The remainder was analyzed for metals and the radionuclide cesium-137 (for purposes of dating the core). All surfaces coming in contact with the sediment samples were stainless steel or glass, cleaned with LiquiNox^R detergent, 10 percent nitric acid, and deionized, distilled water.

Sediments samples were digested by adding 10 mL of concentrated redistilled nitric acid to one gram of sample and refluxing on a hotplate for 30 minutes. The sample was then allowed to cool, 2 mL of 30 percent hydrogen peroxide added, and the sample returned to the hotplate. Hydrogen peroxide was added in 1 mL increments until there was no change in the appearance of the sample, or after addition of 10 mL. The sample was heated without boiling until the volume had been reduced to 2 mL. The resulting digestate was resuspended in 10 mL of deionized, distilled water, filtered through a 0.8 µm acid-washed membrane filter and reconstituted to 100 mL with deionized, distilled water. The solution was analyzed by atomic absorption methods described in USEPA (1979).

Grain size distribution was determined by the method of sieves and pipettes (Holme and McIntyre 1971). Total organic carbon was analyzed by method 9060 in USEPA (1984). Cesium-137 activity was determined in dried, pressed sediment pellets subjected to gamma ray counting on a high resolution, lithium-drifted germanium diode equipped with an anti-coincidence shield.

The accuracy of the metals data was assessed by analysis of a standard reference material, National Bureau of Standards River Sediment (SRM-1645). The results, shown in Table 1, compared well with certified values. Cadmium and mercury analyses underestimated certified concentrations by approximately 25 percent.

Table 1. Analysis of NBS River Sediment (µg/g, dry)

Metal	Analysis for Present Study[a]	Certified Value
Zn	1640±10	1720±169
Cu	110±1.7	109±19
Pb	694±12	714±28
As	52.9±0.1	66[b]
Cd	7.6±0.2	10.2±1.5
Hg	0.82±0.06	1.1±0.5

[a] mean ± range of duplicates [b] not certified

RESULTS AND DISCUSSION

The results of sediment analysis are shown in Table 2. Lake Roosevelt sediments collected between the international border and Kettle Falls (i.e., Marcus Island station) consisted primarily of a blackish sand (81-99 percent). Below Kettle Falls, sediments were primarily silt and clay (80-97 percent), with the clay and sand content gradually increasing with distance downstream. Sediments in the upper lake had higher organic carbon concentrations than those in the lower lake.

Table 2. Analysis of Lake Roosevelt/Upper Columbia River sediment samples.

		Grain Size (%)					Metals Concentrations (µg/g, dry)					
Location	River Mile	Gravel (>2mm)	Sand (2mm-62um)	Silt (62um-4um)	Clay (<4um)	% Total Organic Carbon	Zn	Cu	Pb	As	Cd	Hg
-Upper Columbia River, British Columbia-												
Lower Arrow Lk.	790	1.50	65.61	26.91	3.82	1.7	51	17	19	3.0	0.6	0.02
Lower Arrow Lk. (replicate)		1.42	61.36	31.04	4.44	1.5	45	13	19	2.5	0.6	0.01
-Lake Roosevelt-												
Boundary	742.8	0.14	99.74	0.06	0.54	7.1	26,840	4,870	365	27.9	0.6	0.01
Deadman's Eddy	738.3	0.08	99.18	0.58	0.60	3.7	22,920	3,390	389	19.7	2.1	0.08
Northport	733.0	0.01	98.77	0.94	0.65	4.1	23,580	3,870	350	25.6	1.4	0.02
Northport (replicate)		0.00	98.51	1.06	0.61	3.7	25,380	3,960	406	31.2	1.4	0.03
Little Dalles	728.1	0.04	98.66	2.46	0.42	6.0	21,420	3,420	294	27.8	1.4	0.02
China Bend	723.8	0.09	81.39	17.19	2.62	4.4	3,840	600	431	15.1	4.8	0.58
Marcus Island	708.9	1.01	96.79	2.36	0.78	2.9	14,440	1,200	550	8.6	3.2	0.05
Marcus Island (replicate)		0.06	97.02	2.30	0.90	2.0	3,940	630	425	12.7	4.7	0.04
Frenchman Pt. Rks.	692.2	0.00	2.83	79.88	14.84	3.5	1,090	165	434	8.7	5.7	2.0
Gifford	676.4	0.00	0.63	68.95	27.73	2.8	1,060	111	467	11.6	5.7	2.7
Hunters	661.0	0.00	6.44	67.30	27.25	2.3	610	80	277	7.5	5.0	1.3
Castle Rock	644.8	0.04	5.87	60.02	29.30	2.7	954	67	349	10.4	5.5	1.0
Seven Bays	635.4	0.00	13.32	48.70	34.34	2.5	976	66	296	13.1	5.6	1.3
Seven Bays (replicate)		0.00	35.08	55.94	30.72	2.9	981	68	289	15.3	5.4	1.3
Swawilla Basin	604.9	0.83	24.05	45.62	34.36	1.5	757	65	206	7.6	5.2	1.0

Table 2. (continued)

Location	River Mile	Grain Size (%) Gravel (>2mm)	Sand (2mm-62um)	Silt (62um-4um)	Clay (<4um)	% Total Organic Carbon	Metals Concentrations (μg/g, dry) Zn	Cu	Pb	As	Cd	Hg
-U.S. Tributaries to Lake Roosevelt-												
Kettle R. Arm	706.4[a]	0.03	22.25	73.67	2.58	2.4	60	29	5	2.8	0.3	0.02
Colville R. Mouth	699.5[a]	0.14	38.84	54.20	4.94	3.2	112	27	21	3.0	1.4	0.05
Spokane R. Arm	638.9[a]	0.00	5.39	50.58	39.27	3.8	1,540	53	128	11.5	5.6	0.16
Sanpoil R. Arm	615.0[a]	0.00	9.72	77.34	8.99	2.9	101	33	12	4.0	0.8	0.05
Sanpoil R. Arm (replicate)		0.00	14.92	77.86	8.33	3.7	98	36	9	4.2	0.6	0.03

[a] river mile on Lake Roosevelt at tributary confluence

Table 3. Analysis of a Lake Roosevelt sediment core.

Depth Increment (cm)	Metals Concentrations (μg/g, dry) Zn	Cu	Pb	As	Cd	Hg	137Cs (pCi/g, dry)[a]
0-5	2,110	128	499	8.9	5.1	2.1	1.00 ± 6.6%
5-10	1,470	64	545	11.4	5.1	1.3	1.05 ± 4.7%
10-15	2,810	81	1,190	33.0	4.7	1.7	1.94 ± 2.1%
15-20	2,730	85	1,140	27.6	5.2	1.6	0.13 ± 12.4%
20-25	2,720	88	1,040	16.4	5.1	3.1	0
25-30	660	49	439	10.6	4.7	0.34	0
30-35	65	13	6	2.1	0.22	<0.07	0
35-40	53	13	4	1.0	0.12	<0.07	0
40-45	57	16	5	2.9	0.14	<0.07	0
45-50	53	17	3	0.6	0.10	<0.07	0

[a] picocuries per gram ± two-sigma, propagated, counting error

The lake sediments were elevated in concentrations of all metals analyzed. Peak concentrations of zinc, copper, and mercury (26,840, 4,870, and 2.7 µg/g, dry, respectively) were two orders of magnitude higher in the lake than in its tributaries or in Lower Arrow Lake. Lead (550 µg/g), arsenic (31.2 µg/g), and cadmium (5.7 µg/g) concentrations were elevated by up to one order of magnitude.

Metals concentrations in the sediments of Lake Roosevelt tributaries were within a factor of two of those in Lower Arrow Lake, except for the Spokane River Arm which was relatively high in all metals, especially zinc, lead, arsenic, and cadmium. However, when compared to Lake Roosevelt sediments above the Spokane confluence (i.e., Castle Rock and Hunters stations), the Spokane sediments were elevated only for zinc (1540 µg/g vs. 954 and 610 µg/g). The cadmium concentration measured in the Spokane Arm (5.6 µg/g) was in the upper end of the range found in the lake.

Strong longitudinal gradients in metals concentrations were evident - three patterns were observed. The sandy material collected in the upper lake contained the highest concentrations of zinc, copper, and arsenic. Maximum concentrations occurred at the border and decreased rapidly downstream to Kettle Falls, below which they remained relatively constant. Contrasting gradients were observed for cadmium and mercury which increased with distance downstream of the border and reached maximum levels in the finer sediments below Kettle Falls. Gradients in lead concentrations were less marked than other metals. The highest concentrations occurred in the middle reaches of the lake, however, the overall range in concentrations was not large (206 - 550 µg/g).

The high metals concentrations in Lake Roosevelt sediments are thought to be primarily due to discharges from the Cominco Limited lead-zinc smelter and refinery at Trail, BC, approximately ten miles above the international border (see Figure 1). It has been in existence since the turn of the century and is the largest such facility in the world. Extensive monitoring by the BC Ministry of Environment and Environment Canada has shown this is the major metals source to the Columbia River in British Columbia (Ministry of Environment 1979; Scheehan and Lamb 1987; Smith 1987).

The steep gradients in zinc, copper, and arsenic concentrations in upper Lake Roosevelt are probably due to slag which is discharged from Cominco in the form of a "coarse-grained sand"; an average of 460 metric tons/day was reported to have been discharged to the Columbia River in 1977 (Ministry of Environment 1979). Limited experimental data suggest slag may be relatively inert. Twenty-four hour leaching experiments with ground Cominco slag and Columbia River water were performed in 1976 (Ministry of Environment 1979). Zinc, lead, cadmium, and mercury were monitored. At a pH of 6.8 (lowest recorded for the river) negligible amounts of metals were leached. Greater leaching of zinc, lead, and cadmium occurred at pH 2.0 suggesting possible enhanced metals availability when slag is ingested by fish.

The distribution of cadmium and mercury in Lake Roosevelt sediments could be construed as indicating a source(s) below the border. Historically, there have been mining and milling operations along the lake's east shoreline between Northport and Kettle Falls (Orlob and Saxton 1950). These, however, were small-scale operations. It is more likely that the distribution of these metals is a function of the silt and clay content of the sediments.

The elevated zinc and cadmium concentrations observed in sediments of the Spokane River Arm probably reflect mining and related activities along the Coeur d'Alene River in Northern Idaho which resulted in contamination of the Spokane River. The Coeur d'Alene discharges were largely curtailed in the late 1970's (Mink 1971; Yake 1979; and Patmont et al. 1987).

The Lake Roosevelt sediment core data are in Table 3. The coring site, Frenchman Point Rocks, is in the region of maximum lead, cadmium, and mercury concentrations found in the surface sediments. Metals concentrations in the top 5 cm of this core were in good agreement with the nearby grab sample (Table 2).

Analysis of the fallout radionuclide cesium-137 ($t_{1/2}$=30 years) provides two dates: (1) 1963, the peak of nuclear weapons testing, and (2) 1954, the first major injection of bomb debris into the atmosphere (Schell and Nevissi 1980). In the Lake Roosevelt core, cesium-137 activity peaked at 10 to 15 cm and could not be detected below 20 cm. On this basis, the increments 10 to 15 cm and 15 to 20 cm were considered to contain material laid down in 1963 and 1954, respectively.

Metals contamination in this part of the lake appears to have begun before 1954. The contaminated layer may exceed the 30 cm thickness indicated because the type of coring device used can lead to core shortening (Hongve and Erlandsen 1979). Mercury concentrations in recent lake sediments are at least two orders of magnitude above historical background levels (i.e., below 30 cm), while zinc, copper, lead, arsenic, and cadmium are one to two orders of magnitude above historical background. Except for lead, the level of contamination has apparently not changed appreciably since the 1950's. The steep gradient in increasing metals concentrations between the 30 to 35 cm and 20 to 25 cm increments may represent early discharges by Cominco -- probably corresponding to the 1920's when the lead-zinc smelter was expanded to its current size. Recent improvements in the quality of Cominco discharges, described below, may not be detectable due to the relatively thick increments analyzed.

In the 1970's Cominco embarked on a program to reduce metals discharges to the Columbia River. Smith (1987) reports data, collected by industry, showing zinc, lead, cadmium, and mercury loads in liquid effluents for the period 1977 through 1985 have been reduced by 69-85 percent, depending on the metal in question. Cominco will continue to discharge slag. Although the present permit allows discharge of up to 900 metric tons/day, current discharges are in the range of 250-400 metric tons/day (R. Crozier, personal communication). Lake Roosevelt is the ultimate sink for this material.

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REFERENCES

Holme NA, McIntyre AD (1971) Methods for study of the marine benthos. International Biological Program Handbook No. 16

Hongve D, Erlandsen AH (1979) Shortening of surface sediment cores during sampling. Hydrobiolgia 63:283-287

Hopkins BS, Clark DK, Schlender M, Stinson M (1985) Basic Water Monitoring Program: Fish tissue and sediment sampling for 1984. Wash St Dept of Ecology. Olympia, 43 pp

Johnson A, Norton D, Yake W (1988) An assessment of metals contamination in Lake Roosevelt. Wash St Dept of Ecology, Olympia, 77 pp

Lowe TP, May TW, Brumbaugh WG, Kane DA (1985) National Contaminant Biomonitoring Program: Concentrations of seven elements in freshwater fish, 1978-1981. Arch Environ Contam Toxicol 14:363-388

Mink LL, Williams RE, Wallace AT (1971) Effect of industrial and domestic effluents on water quality of the Coeur d'Alene River basin. Idaho Bureau of Mines and Geology, 149 pp

Ministry of Environment (1979) Kootenay air and water quality study, phase II. Victoria, BC, 238 pp

Orlob GT, Saxton WW (1950) Mining and mill waste pollution in the upper Columbia River basin and Lake Roosevelt. Wash Pollut Control Comm, Olympia, 12 pp

Patmont CR, Pelletier, Singleton LR, Soltero RA, Trial WT, Welch EB (1987) The Spokane River basin: Allowable phosphorus loading. Wash St Dept of Ecology, Olympia, 178 pp

Schell WR, Nevissi A (1980) Detrital sedimentation in lakes and reservoirs. Univ of Wash, Seattle, 27 pp

Sheehan SW, Lamb M (1987) Water chemistry of the Columbia and Pend d'Oreille Rivers near the international boundary. Environment Canada, Vancouver, BC. 312 pp

Smith AL (1987) Levels of metals and metallothionein in fish of the Columbia River near the international boundary. for BC Ministry of Environment, and Environment Canada, 133 pp

USEPA (1979) Chemical analysis of waters and wastes. USEPA 600/4-79, March 1983 revision

USEPA (1984) Test methods for evaluating solid waste, SW846. Washington DC

Yake WE (1979) Water quality trend analysis - the Spokane River basin. Wash St Dept of Ecology, Olympia, DOE-PR-6, 39 pp + app

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