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Sediment Quality and Ecorisk Assessment Factors for a Major River System



Prepared for the U.S. Department of Energy
Office of Environmental Restoration and
Waste Management



Westinghouse
Hanford Company Richland, Washington

Hanford Operations and Engineering Contractor for the
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Sediment Quality and Ecorisk Assessment Factors for a Major River System

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ABSTRACT

Sediment-related water quality and risk assessment parameters were developed from metal loading and concentration data from Lake Roosevelt (river mile 703) to the mouth. Correlation of lead, zinc, mercury, and cadmium concentrations in downstream sediments with refinery operations in British Columbia suggest solutes with K_d 's $> 10^5$ reach about 1 to 5 $\mu\text{g/g}$ per metric ton/yr of input. A low suspended load (upriver average < 10 mg/L) and high particle-surface reactivity account for the high clay-fraction contaminant concentrations. In addition, a sediment exposure path was demonstrated with post-shutdown biodynamics of a heavy metal radiotracer (^{65}Zn) following termination of the last Hanford single-pass plutonium production reactor in 1971. Residence times for decay-corrected ^{65}Zn in suspended sediment, biota, and bed sediment were 600 to 900 days versus approximately 20 days for water. The slow decline in sediment was attributed to resuspension, bioturbation, and anthropogenic disturbances. The above findings suggest that conservative sediment quality criteria should be used to restrict additional contaminant loading in the upper drainage basin. The issuance of an advisory for Lake Roosevelt, due in part to mercury accumulation in large sport fish, emphasizes the need for more restrictive controls. An approach for assessing relative human exposure and ecological risk due to particle-bound contaminants in the Columbia River is also proposed.

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INTRODUCTION

Sediment quality, as an adjunct to water quality, has become an important parameter in ecological risk assessments. Streambed sediments and floodplain soils constitute a sink for many pollutants. Once contaminated, the sediments are a potential source of water pollution that persists after the original source is terminated and remediation is completed. Disturbance of sediments, especially during large-scale earth-moving remediation or unusual erosional events, is a major potential vector for contaminant dispersal. Pollutants for which a sediment or particulate phase pathway is important include most heavy metals, many radionuclides, and a wide array of organic materials (Olsen et al. 1982).

Ecological risk assessments for major remediation projects are required by the U.S. Environmental Protection Agency (EPA). These involve definition or characterization of the hazard followed by exposure and effects assessments. The exposure assessment relies on transport and fate models, while the effects assessment relies on toxicity testing (Suter and Loar 1991). Such assessments for projects located in major drainage basins invariably include a fluvial and estuarine component or pathway. Exposure levels for contaminants with little or no affinity for particulate phases can be estimated from simple mixing and dilution given the mass input rate to the receiving water. If the contaminants have an affinity for particulate phases, particle transport and sedimentary processes must be considered.

While many assessment models incorporate sediment-contaminant transport components, the models require input parameters that are often site dependent. Such information is rarely available for most large aquatic ecosystems. Anthropogenic sources, however, can sometimes serve as large-scale sediment "tracers" to provide some of the site-specific input parameters for the exposure assessment models. This paper describes contaminant-sediment transport and behavior in a large fluvial-estuarine ecosystem that was uniquely "labeled" with heavy metals and radionuclides. These tracers permitted evaluation of the net effect of scour and deposition, contaminant-sediment dilution and mixing, rates of sediment purging, water-to-sediment mass transfer, particle size contaminant concentration effects, and evaluation of a major estuarine sediment exposure pathway.

Previous Studies. Transport and behavior of sediment and radionuclides in the lower Columbia River and estuary were the subject of several previously reported investigations (Pruter and Alverson 1972, Haushild et al. 1966, Haushild et al. 1975, Hubbell and Glenn 1973, Perkins et al. 1966). These studies were conducted during long-term input of radiocontaminants to the lower Columbia River by the Hanford Site near Richland, Washington, from September 1944 until January 1971, when the last "once-through" plutonium production reactor was terminated. Valuable insights concerning sediment and contaminant transport processes were gained from these studies. This work, an extension of the "steady-state" studies, focuses primarily on the post-shutdown period from 1971 to 1976. This study also focuses on other related tracers, such as the heavy metals in the upper Columbia River drainage basin from mining and associated refinery activities. These historical contaminant releases provided opportunities to evaluate contaminant-sediment transport and fate directly in the second largest river system in the conterminous United States.

Purpose. The specific purposes of this paper are to (1) report previously unpublished radionuclide and heavy metal data, (2) interpret the environmental significance of these and related results, and (3) generalize the findings for sediment quality and ecological risk assessments in the lower Columbia River and coastal zone.

Data Sources. Data reported in this paper were acquired by the authors from ca. 1971 to 1976. Sources include thesis and related unpublished research as well as sediment quality surveys conducted by the authors for private and governmental agencies under contract with the School of Oceanography, Oregon State University, and supplemented by reference to other studies.

CONTAMINANT-TRACER SOURCE CHARACTERISTICS

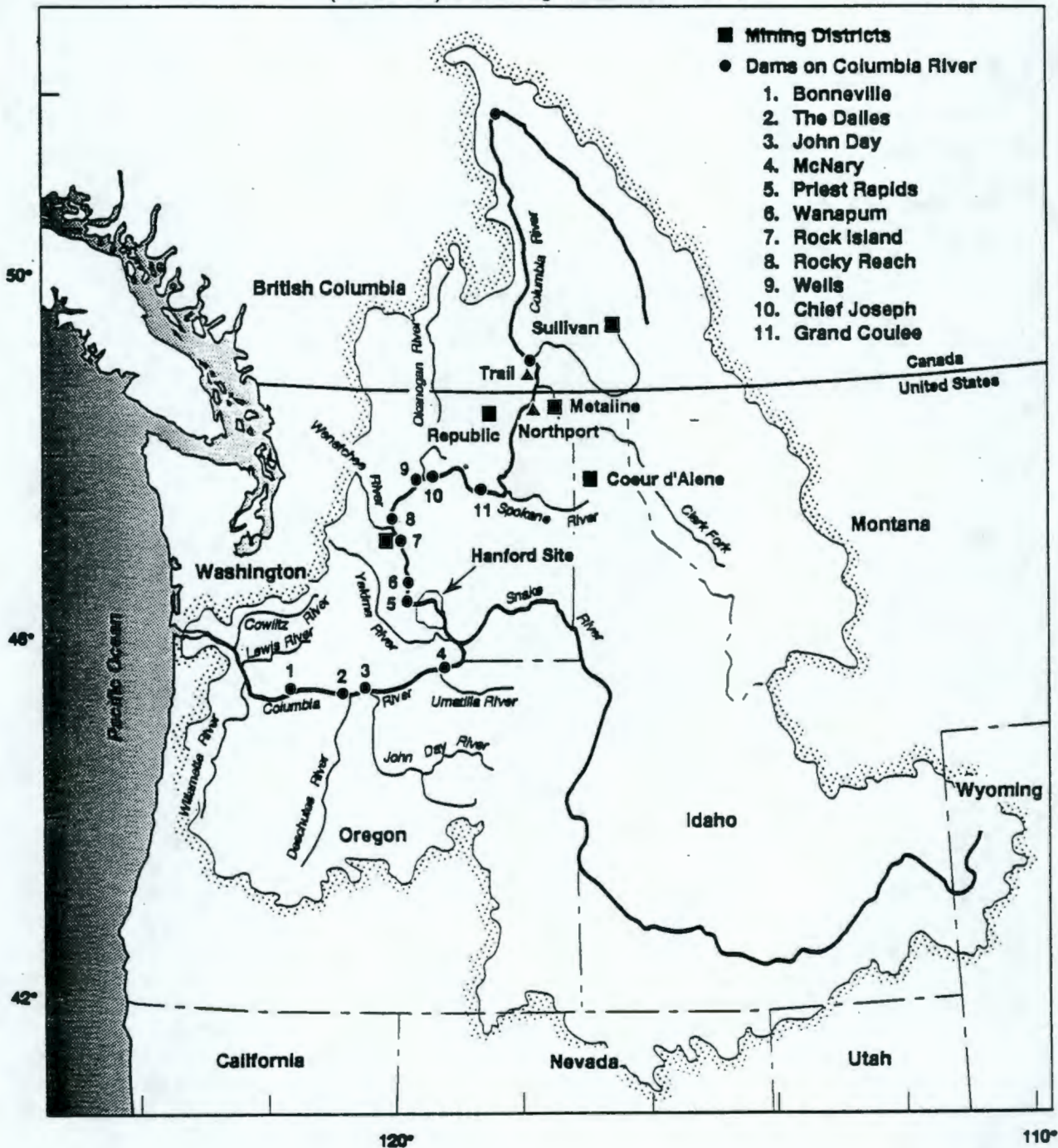
The sediment "tracers" to be evaluated consist of two types: heavy metals introduced by mining and refinery activities that began in the late 1800's in the upper Columbia River drainage basin, and neutron activation products in reactor cooling water released to the Columbia River by the Hanford Site between 1944 and 1971. The contaminant-tracer source areas and major geographic features are shown in Figure 1. Mining-related metals and radiotracer sources are discussed in the following paragraphs.

Mining Activities

Source Areas. The mining districts in northeastern Washington, the panhandle region of Idaho, and southern British Columbia (see Figure 1) have been active since the late 1800's. The mineralized areas are located in the Okanogan Highlands Province (see Figure 1) and the Kootenay Arc in southern British Columbia. The Okanogan Province has three principal mining districts, and the Kootenay Arc has two principal districts. Metal production from these districts is listed in Table 1. Both the Coeur d'Alene and the Metaline Districts are located near streams that drain to the Coeur d'Alene Lake system. The Kootenay Arc mines are located near streams and rivers that drain to the upper Columbia River. Some of the arc-related mineral deposits may have been subjected to glacial erosion during the Pleistocene and might elevate the metal concentrations downstream. However, the absence of anomalous zinc and lead concentrations in Columbia River sediments of approximately Pleistocene age from the Hanford Site suggests little if any such influence on sediment concentrations for these metals. Little influence is suggested because metal contents of sediment samples with the textural equivalent of shale are consistent with world average shale values.

Waste Input. Mining and associated processing activities released metal-bearing process wastes to streams within the drainage basins where the mines are located. For example, untreated mine tailings, mine drainage, and refinery wastewater were released directly to the Coeur d'Alene River from ca. 1885 to 1968 (Rember et al. 1991). Zinc and cadmium concentrations in this river were above toxic levels for fish. Benthic biota were virtually absent from the streambed until after 1968 when some degree of effluent treatment was instituted (Mink et al. 1971). In addition to metals transported in solution, finely divided ore particulates were also transported into downstream tributaries and deposited in the connected lakes (Maxfield et al. 1974, Rember et al. 1991). Elevated concentrations of zinc and lead (up

Figure 1. Columbia river Drainage Basin Showing the Main River Courses (in Bold) and Major Tributaries.



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Dams on the main stem of the Columbia River up to the Canadian border are identified with numbers 1 through 11. Lake Roosevelt, formed by Grand Coulee Dam (11), and Lake Wallula, formed by McNary Dam (4), are the largest reservoirs and the principal sediment traps in the river-reservoir complex. Lake Roosevelt is 240 km long, has an average depth of 40 m, a maximum depth of 120 m near the dam face, and a full pool volume of $1.2 \times 10^{10} \text{ m}^3$; average residence time for water in storage is about 40 days. The other reservoirs are low head, "run of the river" hydroelectric projects that operate with minimal water in storage. The only free-flowing stretches remaining below Grand Coulee Dam are within the Hanford Reach below Priest Rapids Dam (5) and the reach below Bonneville Dam (1) to the estuary. Annual average freshwater and sediment discharge to the Pacific Ocean is $23 \times 10^{13} \text{ L}$ and $9 \times 10^{12} \text{ g}$, respectively, or 40 mg/L of suspended sediment (Cutshall et al. 1986). The relatively low suspended sediment concentration, compared to other major rivers, is attributed to the dominance of crystalline or igneous and metamorphic rock in the drainage basin.

Table 1. Metal Production from Major Mining Districts in the Upper Columbia River Drainage Basin.

Mine or District	Starting Year	Total tons x 1000			
		Zinc	Lead	Copper	Reference
Coeur d'Alene District	1884	2450	6900	112	(1)
Metalline District	1906	400	178	-	(2)
Van Stone Mine	1926	120	11	-	(3)
Reeves-MacDonald, B.C.	-	198	57	-	(4)
Sullivan, B.C.	1920	9600*	9600*	-	(4)

* based on estimated original size of orebody; also contain manganese, arsenic, copper, tin, tungsten, antimony, cadmium, indium, thallium, and mercury (Hoy et al. 1985).

(1) Hobbs et al. (1968), (2) McConnell and Anderson (1968), (3) Cox (1968), and (4) Hoy et al. (1985)

to 3,000 to 5,000 ppm) and copper and cadmium (up to 100 to 200 ppm) in surface sediments of Coeur d'Alene Lake were first reported by Maxfield et al. (1974). More recent studies of heavy metal depth distribution in cores from the Coeur d'Alene River and lateral lakes reflect the variation of input over time and document the historical record of mining and refining activities in the originating drainage basin.

The Sullivan mine in British Columbia generated approximately 5×10^9 L/yr of wastewater that ultimately drained into the St. Mary's River and then to the upper Columbia River and Lake Roosevelt behind Grand Coulee Dam. The major heavy metal contaminants in this wastewater (from the ore flotation process) were lead and zinc, with average concentrations of $1,400 \mu\text{g/L}$ and $16,000 \mu\text{g/L}$, respectively (Robbins 1978). If these concentrations are representative of input during and before the early 1970's (the period of interest for this study), an estimated 80 metric tons (MT)/yr of zinc and 7 MT/yr of lead were added to the upper Columbia River from this source alone. Based on the cadmium/zinc ratio for zinc ores, the corresponding estimate for cadmium is 1 MT/yr. Robbins further pointed out the potential impact of contaminated sediments on downstream benthos in the St. Mary's River and cited evidence of fish toxicity from live cage tests above and below the outfall from the Sullivan mine (e.g., a survival rate of 47% relative to upstream controls). A reduction in stream benthos by as much as 95% was also reported. The importance of sediment in controlling metal accumulation in fish tissue in freshwater environments has been noted in Delisle et al. (1975).

In addition to metal sources caused by mining activities, smelter and refinery wastewater is a major source of heavy metal input to the upper Columbia River. The most significant source is the Cominco, Ltd. refinery located near Trail, British Columbia (see Figure 1, Johnson et al. 1989). The Cominco, Ltd. refinery has operated since the 1900's and is the world's largest lead-zinc smelter and refinery. The facility released dissolved iron, manganese, zinc, copper, lead, arsenic, cadmium, and mercury via liquid

effluents and as solids in the form of slag. Reported mass loading rates for the dissolved component in 1977, the earliest available recorded data, are listed in Table 2.

Input rates for subsequent years were comparable to the above until about 1978. By 1986, discharge rates had declined to about 15% of the Table 2 rates because of improved effluent controls. Greater reductions, especially in mercury, may have occurred after a new lead smelter was built in 1987. Metal concentrations versus depth in a 1986 sediment core from the upper end of Lake Roosevelt (ca. river mi 703) suggest that the input rates for the early to mid 1970's were fairly uniform and probably similar to the input rates for 1977 shown above (Johnson et al. 1989). The ionic nature of the metals transported into Lake Roosevelt from Canada as dissolved and suspended load is suggested by their relative affinity for particulates. For example, the percent retained on 0.4 μm filters was <10% for arsenic, 53% for zinc, 74% for lead, and 80% for copper at Northport (based on mean values for the period 1978 through 1987 [Johnson et al. 1989]). The observed order of affinity for particulates is consistent with the expected oxidation states of these metals and the order of their surface-chemical reactivity (Olsen et al. 1982).

Based on sediment core data and an age/event marker (fallout ^{137}Cs profile), sediment concentrations in the mid to upper reservoir during the mid-1970's are estimated to have been on the order of 3,000 for zinc, 1,000 for lead, 30 for arsenic, 5 for cadmium, and 1 $\mu\text{g/g}$ for mercury (Johnson et al. 1989). Zinc concentrations in sediments near the inlet to Lake Roosevelt are nearly ten-fold higher. This is attributed to the larger, more dense slag particles transported as bed load that settle out in the slack water of the lake. The slag from the Cominco, Ltd. refinery contains non-leachable metals consisting primarily of iron, manganese, and zinc.

Two additional major tributaries contributed mining-related metals to the upper Columbia River during the study period. These tributaries are the Pend Oreille, which enters the Columbia River in Canada, and the Spokane River, which discharges into the lower end of Lake Roosevelt (see Figure 1). The Clark Fork enters Lake Pend Oreille, which becomes the Pend Oreille River. The Clark Fork may contribute some heavy metals from the copper mining and smelter operations in Montana.

Estimates of input from the above sources were made using discharge-weighted average concentrations for water years 1976 and 1977 (October 1, 1975, through September 30, 1977) (USGS 1976, 1977). However, the only consistently detected results for this period were for zinc. The average transport rate for water years 1976 and 1977 was 830 MT/yr for the Pend Oreille and 850 Mt/yr for the Spokane River.

The total estimated input rate of zinc from the major sources described above is $80 + 3,100 + 830 + 850 = 4,860$ MT/yr. The amount entering the upper end of Lake Roosevelt was also calculated using discharge-weighted concentrations for the Northport sampling station (river mile 733). The average for water years 1976 and 1977 was 5,200 MT/yr. This compares with a total of 4,010 MT/yr based on the effluent records for the Sullivan mine plus effluent data for the Cominco, Ltd. refinery and the discharge estimate for the Pend Oreille. The somewhat higher value for the Northport estimate may be sampling related. For example, Windom et al. (1991) estimated that water samples of east coast rivers collected with the same device used for the

Table 2. Heavy Metal Input Rates to the Columbia River
Near Trail, British Columbia.

Metal	¹ Form	kg/day	MT/yr
Zinc	Zn ²⁺	8400	3100
Lead	Pb ²⁺	250	91
Cadmium	Cd ²⁺	95	35
Arsenic	AsO ₄ H ²⁻	10	3.7
Mercury	Hg ²⁺	7	2.5

(Johnson et al. 1989)

¹Expected chemical states in Columbia River water. Divalent heavy metals may exist in a variety of ion-pair associations (e.g., PbOH⁺, PbCO₃⁰; Jenne et al. 1980).

Columbia River sampling produced a blank of about 20 µg/L for zinc. A correction of this magnitude would lower the Northport estimate to 3,500 MT/yr. The natural background contribution is assumed to be negligible based on more recent sampling results upstream of the lead-zinc refinery showing total zinc levels of about 1 µg/L.

The estimates of metal input and/or transport suggest that (1) refinery effluent records are consistent with transport estimates at Northport and (2) the Cominco, Ltd. refinery alone contributed the majority of heavy metal input to the upper Columbia River during the period of interest for this study.

Metal Uptake by Sediment. The conceptual model consists of metal sorption from solution (as divalent heavy metal cations) by suspended sediments after the main metal-carrying stream or wastewater mixes with the Columbia River. Major controls on the degree of heavy metal-particle-association in the Columbia River are as follows.

pH. Heavy metal sorption by particulate phases is in part controlled by pH. For example, changes in pH from acidic to basic can result in significant increases in the uptake of zinc, cadmium, and especially lead (O'Connor and Renn 1964, Erel and Morgan 1992). The Couer d'Alene River pH was in the range of 5 to 6, while the Spokane and Columbia Rivers were slightly alkaline (pH of about 8). Thus, heavy metal sorption was probably enhanced upon mixing the rivers.

Competing Ion Effect and Biological Uptake. Uptake of divalent metals such as Zn²⁺ by diatoms and organic aggregates has been shown to occur. However, this phenomenon appears to be more related to surface chemistry than to active cell processes because living and killed seston exhibit similar radioactive zinc uptake (Cushing and Watson 1968). In addition to pH, other secondary chemical controls include the competing ion effect where the more abundant divalent cations of magnesium and calcium may reduce the uptake of Pb²⁺, Zn²⁺, and Cd²⁺. This effect, however, is apparently minimal for divalent lead in aqueous solutions (Erel and Morgan 1992). Average concentrations (mg/L) of the major cations and anions in the Columbia River at the Dalles,

Oregon, are as follows: Mg^{2+} (6.2), Ca^{2+} (23), Na^{1+} (16), HCO_3^{2-} (108), SO_4^{2-} (19), Cl^{1-} (4.9), and F^{1-} (0.5). In general, the chemical composition of the Columbia River has been characterized as a slightly alkaline, dilute calcium bicarbonate solution (Johnson 1979). Because of apparent differences in sensitivity to pH changes, competing ion effects, and divalent heavy-metal chemistry, the sorption of lead by Columbia River seston and suspended sediments is expected to be greater than cadmium and zinc. These chemical differences are reflected in the percent particulate averages at the Northport station as previously indicated.

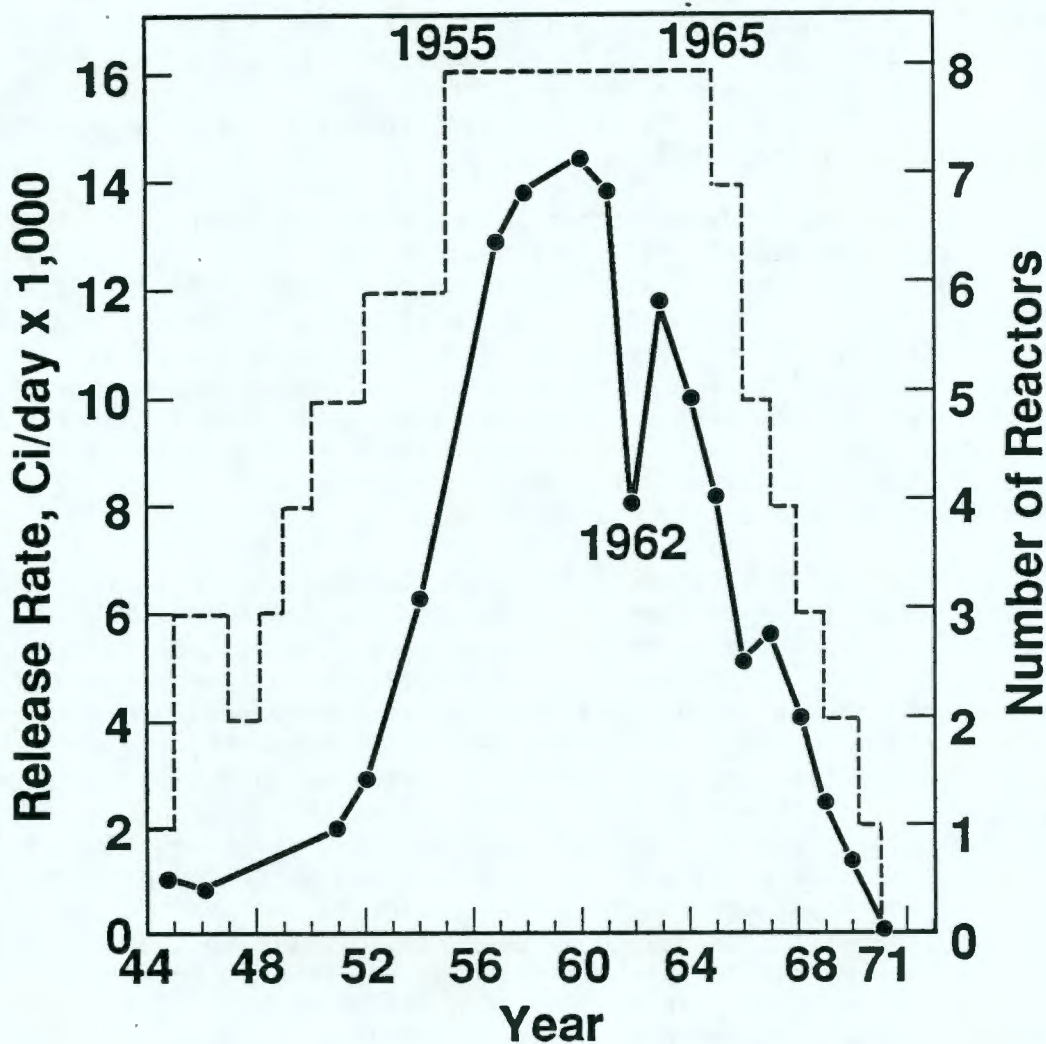
Suspended Sediment Concentration. The fate of the particle-bound metal fraction in transport depends on stream velocity, particle size, and particle density. It is likely that only colloidal or clay-size particles would be transported through the impoundments because larger particles or high-density ore fines are trapped, especially in the larger reservoirs such as Lake Roosevelt and Lake Wallula behind McNary Dam. Smaller particles may also form larger aggregates (diatom-particle associations [Meade 1972]) that can settle in low-velocity zones and thus become incorporated in bed sediments. Sorption by both suspended and bed sediment probably occurred over the entire downstream distance in the Columbia River.

The average suspended load of the upper Columbia River is less than 10 mg/L, increasing to an average of 40 mg/L at the mouth (Cutshall et al. 1986). Based on laboratory sorption studies of zinc by stream sediments (O'Connor and Renn 1964), the very low suspended load of the Columbia River probably accounts for the relatively high soluble heavy metal fraction. Regardless of the major controls on sediment-metal sorption, the mining related metal concentrations in bed sediment below Grand Coulee Dam should decrease in a systematic manner with distance downstream because of sediment "dilution" by tributaries as observed in other drainage basins (Hawkes 1976). This hypothesis and the magnitude of the effect can be tested using both suspended sediment loads and the heavy metal labeled sediment to quantify the mixing or dilution. It should also be possible to estimate the resulting metal-sediment concentrations per unit loading (MT/yr) of the respective heavy metal solutes from upstream mining and refining operations. The latter parameter is a measure of the net solute distribution between the water and sedimentary compartment in this fluvial-estuarine ecosystem. This information may be used as exposure "bench marks" or reference points for assessment of contaminant pathways and effects of chronic loading from upstream sources.

Past Hanford Site Operations. The single-pass plutonium production reactors at the Hanford Site (see Figure 1) began operation in 1944 and ended when the last of eight original reactors was shut down in 1971. Between 1965 and 1971, approximately one reactor per year terminated operation, as shown in Figure 2. These reactors used chemically treated coolant water from the Columbia River. The treated water was then passed through the reactors, stored in large concrete retention basins for a short time, and returned to the river. Radioisotopes of many different metals were produced by the intense neutron flux (Foster 1972). The most significant radioisotopes for sediment tracer purposes are listed in Table 3.

Cationic radiotracers were readily taken up by suspended particulates while the anionic radioisotopes were transported almost exclusively as solutes (Cutshall 1967, Cutshall et al. 1967, Johnson et al. 1967, Johnson 1979).

Figure 2. Radioactive Waste Discharge History for the Single-Pass Plutonium Production Reactors, Hanford, Washington, 1944-1971.



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The variations in radionuclide release rates between 1955 and 1965, when all eight reactors were in continuous operation, reflect changes in effluent control procedures. For example, the sharp decrease during 1962 was caused by the use of increased amounts of alum (aluminum sulfate) to remove particulates from influent. The enhanced water pretreatment significantly reduced neutron activation products but was abandoned because of mechanical/filtration problems. Other changes resulted in a gradual reduction in radionuclide discharge rate during the latter half of the period when all eight single-pass reactors were in operation (i.e., 1961-1965).

Table 3. Selected Radionuclides Released to the Columbia River from Plutonium Production Reactor Operations, Hanford, Washington.

Radioisotope	Half-life	Chemical State
^{152}Eu	13.6 yr	Eu^{3+}
^{60}Co	5.27 yr	Co^{2+}
^{65}Zn	245 days	Zn^{2+}
^{51}Cr	28 days	CrO_4^{2-}

Sufficient tracer was added to the river system to make these radioactive "tags" easily detectable in sediments from below the reactor area to the estuary and on the continental shelf (Cutshall et al. 1971). In 1965-1966, annual average input rates (Ci/yr) to the river were ^{65}Zn , 12,500; ^{60}Co , 310; ^{51}Cr , 340,000 (Hall 1968); and ^{152}Eu , 1,150 (estimated).

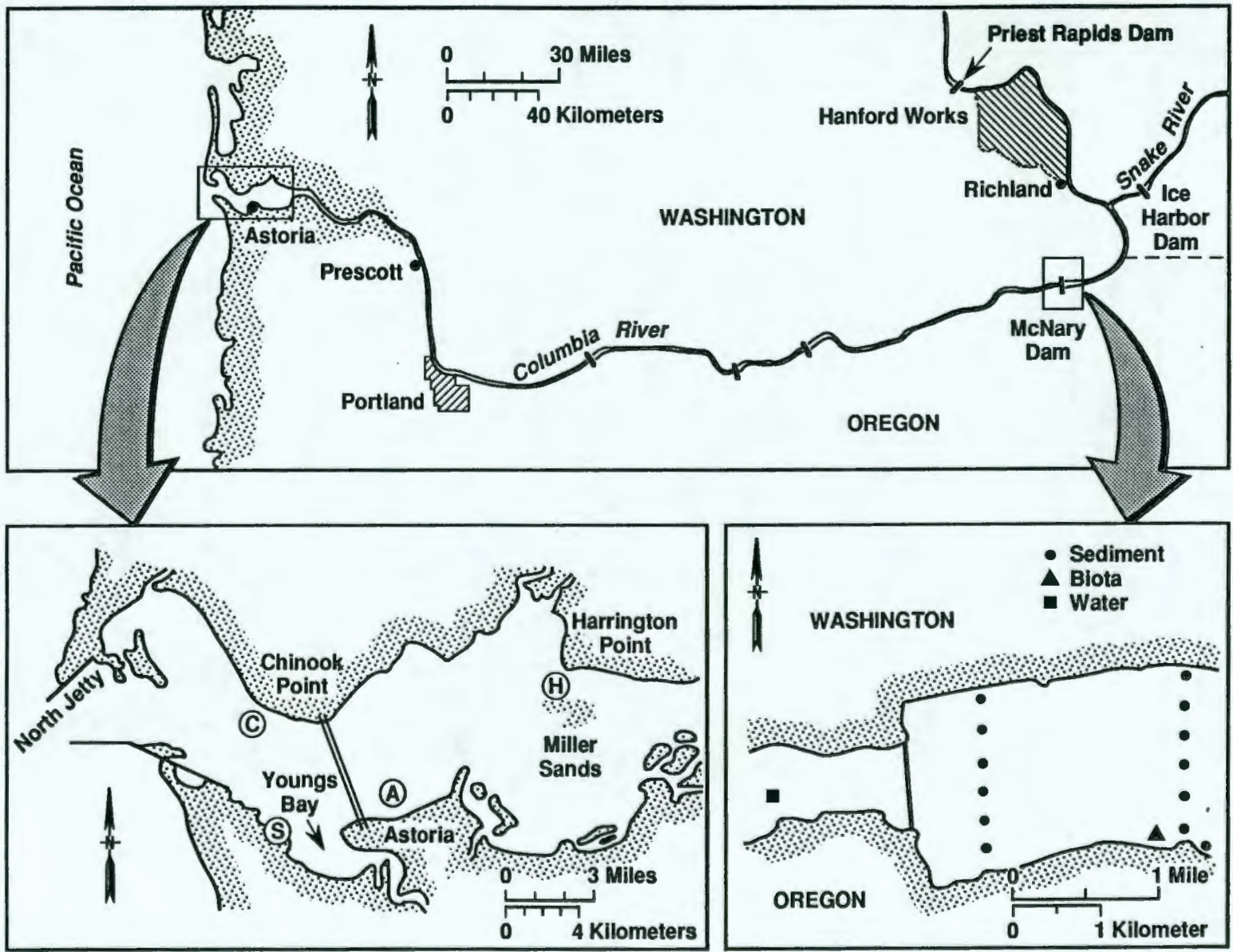
As indicated above, sediment labeling occurred on a very large scale during the operating years of the plutonium production reactors. When the last reactor was shut down, an additional opportunity was provided to evaluate contaminant or tracer dynamics on an ecosystem scale. It is the post-shutdown period between 1971 and 1975 that is of particular interest for evaluating the rate of decline or turnover of heavy metals and radionuclides in biologic and sedimentary components of the riverine-estuarine ecosystem. During this same period, sediment quality surveys, which included heavy metals and radionuclides, were conducted in the lower river and estuary (Johnson 1979, Johnson and Cutshall 1975, Cutshall and Johnson 1976) and subsequent metal-particle-size relationships (Wagner 1976). The unique tracer combination provided by the availability of both stable zinc from upstream mining/refinery operations and the radioisotope, ^{65}Zn , from the Hanford Site, is valuable for this review and is thus emphasized in the following sections. The longer-lived radionuclides (^{60}Co and ^{152}Eu) are also useful as dating tools for evaluating depositional history (Johnson 1979).

SAMPLING AND ANALYSIS

The study areas and key sample locations for the studies already discussed are shown in Figure 3. Analytical and sampling conditions for the radiotracer studies (ca. 1971 to 1975) and the sediment quality surveys are as follows.

Radiotracer Study. Lake Wallula, formed by McNary Dam (Figure 3), was chosen because it was the first downstream impoundment below the reactor areas and it had been previously shown to contain a major portion of the radionuclide inventory (estimated 40% to 50% [Johnson 1979]). Selected stations were cored with a 6-in. corer to obtain sufficient samples to assess depth distribution of radionuclides and to estimate sedimentation rates. Grab samples of surface sediment were collected along two transects (see Figure 3) for estimating turnover in surficial sediment. The sediment grab samples were leached with a 0.05M solution of copper sulfate to facilitate measurement of both zinc and

Figure 3. Location Map of Study Areas and Sample Sites.



H, A, C = Biological Collection Stations
 S = Sediment Collection Site (See Figure 14)

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^{65}Zn (this treatment was shown to remove about 50% of the ^{65}Zn , Johnson et al. 1967). Biota were obtained as available (i.e., fish, freshwater mussels, pond weed). Grab samples of water were collected near the center of the river just downstream of the dam every two weeks initially and then monthly. Water samples were analyzed for soluble ($0.4\ \mu\text{m}$ Nuclepore [trademark of CoStar Corporation] filtered) and particulate stable and radiozinc.

The estuary (see inset, Figure 3) is the other major fine-sediment trap in the river system below the Hanford Site. Within the estuary, the post-shutdown studies between 1971 and 1973 involved monthly collections of fish, invertebrates, water, and sediment at the locations indicated. In addition, a composite water sample station was maintained approximately 50 miles upstream from the estuary at Prescott, Oregon (see Figure 3). The National Marine Fisheries Service maintained a research barge moored on the edge of the main channel of the river at this location. A daily sample (approx. 6 L) was collected from the barge, transferred to a 200-L barrel, and the resulting composite sample processed monthly. This provided a time series estimate of radiolabeled suspended sediment (primarily ^{65}Zn) for determining the rate of decline of the source and post-shutdown input to the estuary. The sediment collection station (S) in Figure 3 involved compositing of the top 1-cm layer of sediment from 10 locations in a semi-protected area adjacent to the main channel of the Columbia River. This sample permitted evaluation of turnover rate in surficial sediments at a fine-sediment depositional site in the estuary. Sediment and biota were collected monthly and analyzed for ^{65}Zn and total zinc. Grab samples of water from the various estuary sites were analyzed for soluble and particulate radionuclides (Johnson 1979).

Sediment quality surveys were conducted in the estuary at Young's Bay and Miller Sands Island (see Figure 3). These studies involved collecting sediment grab samples and 6-in. cores in transects across bathymetric transitions at the two sites. Sediment samples were analyzed for selected heavy metals using acid extraction procedures and atomic absorption spectrometry. Grain size analysis was accomplished by (1) sieving through a $63\text{-}\mu\text{m}$ screen to determine percent silt/clay and (2) using the sieve-pipet method for determining median grain diameter (Guy 1969). Companion biological surveys were also conducted, the details of which are reported in Higley and Holton (1975) and Higley et al. (1976).

A special study to relate metal concentration and downstream sediment dilution by tributaries involved collecting sediment from major tributary sites; above the reactor area; and selected locations in the river, estuary, and offshore near the river mouth (see Figure 3). Sediments were subjected to particle-size separation and metal analysis by flame atomic absorption following a nitric-hydrofluoric acid or nitric-hydrochloric acid total digestion procedure. The size separation procedure involved wet sieving through a $63\text{-}\mu\text{m}$ stainless steel screen followed by mechanical dispersion and mixing of the silt-plus-clay fraction in a 1-L graduate cylinder. At a fixed depth below the surface at various predetermined times, 20-mL aliquots were drawn (Guy 1969). The 20-mL aliquots were drawn through preweighed Nuclepore membrane filters, air dried, desiccated, and sediment-weight determined gravimetrically. The sediment fractions were analyzed for metals as described previously. Concurrent analyses of certified reference geologic media (e.g., Abbey et al. 1971) indicated greater than 85% recovery for the metals of interest in this study.

Radioanalysis was accomplished by gamma ray spectrometry using a NaI(Tl) well detector and spectral stripping computer program and by high-resolution gamma energy analysis on a Ge(Li) detector. Efficacy of spectral resolution was checked by analyzing prepared mixtures of certified reference sources of various radionuclides known to be present from discharge records and previous studies. Details of the above sampling and analysis are described in Johnson (1979), Johnson and Cutshall (1975), and Cutshall and Johnson (1976).

RESULTS

Sediment dilution and particle-size effects for the mining-related metals and associated interpretations are presented first followed by the radioisotope results.

Heavy Metals. Metal results for the main stem of the Columbia River and major tributaries and associated sediment dilution considerations are followed by results for the two sediment quality surveys (i.e., Young's Bay, Oregon, and Miller Sands Island, Oregon). The influence of particle size on metal concentrations is fundamental to all of the sediment metal results and is discussed first.

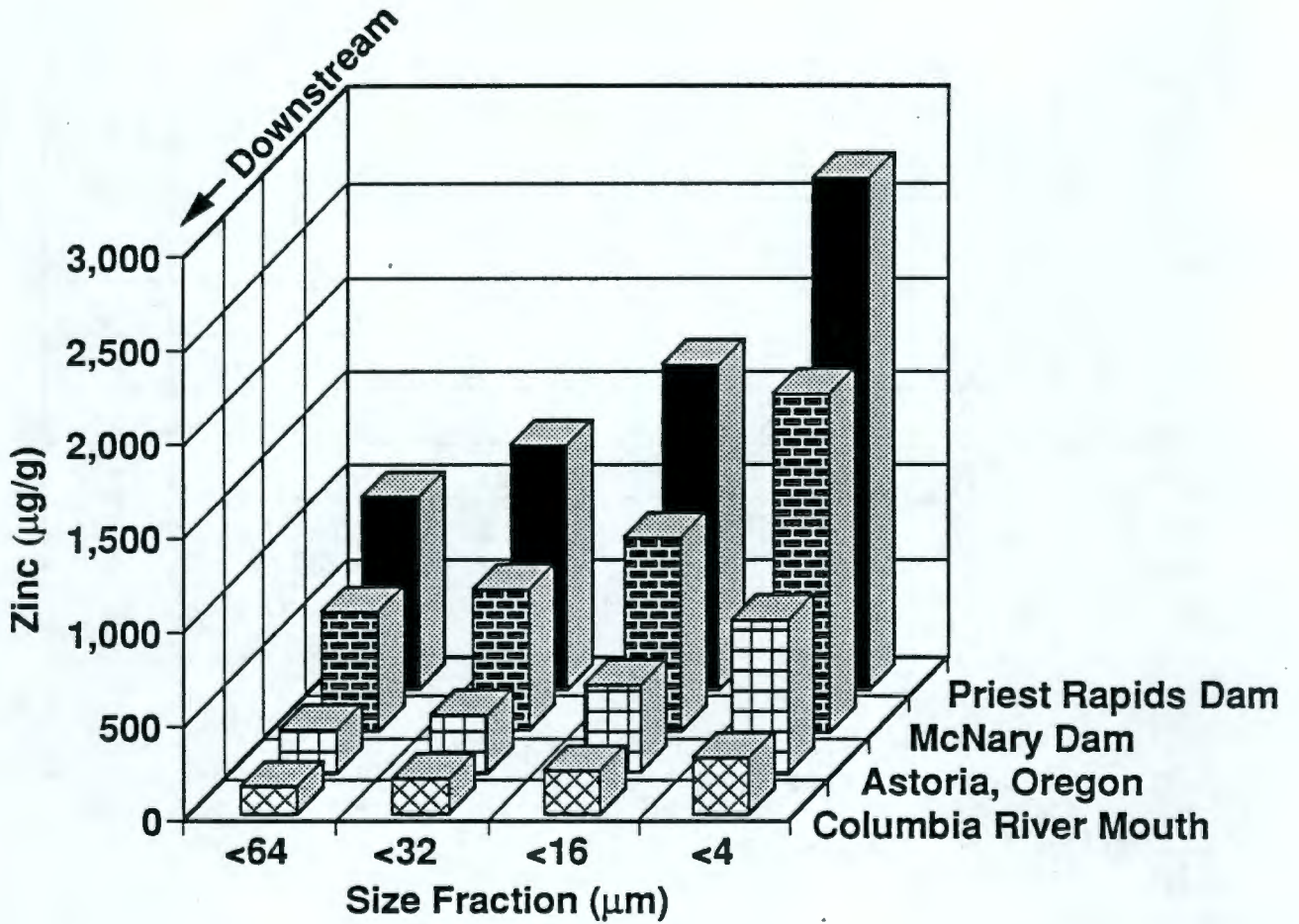
Particle-size considerations. The effect of particle size on sediment metal concentration was evaluated by analyzing size-class separates from surficial sediment samples collected at selected tributary and main stem Columbia River locations. Surface area dependent sediment-metal concentrations should show a progressive increase in concentration with decreasing size class (i.e., in those cases where the metal abundance is significantly greater than natural background). Metal concentrations associated with bulk properties of the sediment (minerals or rock fragments) should not exhibit a strong particle-size dependence.

Because zinc was consistently detectable in all size fractions at all locations, it was used to evaluate the effect of particle size on heavy metal concentrations. Results of the size class separates at four locations from Priest Rapids Dam to the mouth of the Columbia River are shown in Figure 4. For Priest Rapids Dam sediments, a strong particle-size dependence of zinc concentrations was found, which is characteristic of particle surface contamination. Further downstream, where concentrations are lower and dominated by the bulk sediment composition, the particle-size dependence is attenuated. The implications of the significantly higher metal concentrations in the smaller size classes are discussed later.

Tributary and Main Stem Metal Concentrations. Metal contents for the <62- μm size class of sediment samples from the major tributaries and main stem Columbia River are shown in Table 4. Complete analytical results for all metals of interest at all locations were obtained for only the <62- μm fraction that previous studies have shown accounts for a large fraction of the sediment-bound radionuclide transport in the lower river (Haushild et al. 1966, Glenn 1973). During peak sediment discharge, the <62- μm component accounts for approximately 50 wt% of the depth-integrated sediment load.

Metal concentrations (see Table 4) show a decreasing downstream trend for the mining-related metals (lead, zinc, and cadmium), while the other metals are more randomly distributed. Comparison with world average shale

Figure 4. Zinc Concentrations in Various Particle Size Fractions of Bed Sediment from Selected Upper and Lower Columbia River Locations.



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Table 4. Trace Metal Concentrations in Bed Sediment (<62 μm) from the Main Stem Columbia River and Major Tributaries, 1973-1974.

Location	Zn $\mu\text{g/g}$	Cd $\mu\text{g/g}$	Pb $\mu\text{g/g}$	Cu $\mu\text{g/g}$	Mn $\mu\text{g/g}$	Ni $\mu\text{g/g}$	Co $\mu\text{g/g}$
Spokane River	735	5.7	250	35	830	18	16
Priest Rapids Dam	845	9.9	120	40	750	39	18
McNary Dam	572	8.0	81	40	650	27	20
Prescott, OR	197	2.4	46	59	890	41	26
Columbia River mouth	103	1.5	33	30	360	65	17
Ice Harbor Dam	99	1.2	39	38	670	28	23
YBC Composite	157	1.6	29	30	630	25	20
Young's River	90	1.2	31	25	880	19	23
Willamette River	107	2.0	25	44	610	39	26
World Average Shale	95	0.3	20	45	850	68	19

Relative Standard Deviation (%); Mn, Cu, Pb = 1-4%; Cd, Ni, Co = 10%-15%.
YBC: Young's Bay Composite (70 sample sites).

also shows the mining metals anomaly. The general trend of decreasing zinc, cadmium, and lead continues through the estuary. Offshore sediment-metal concentrations are nearly indistinguishable from natural background. Zinc contributions from the upstream mining source in the mixtures are evident over a larger range of apparent sediment dilution or mixing fractions.

Dilution Factor Calculation. Dilution of the elevated metal concentrations by mixing with tributary sediment loads follows a two-component mixing equation:

$$C_m = aC_1 + (1-a)C_2 \quad 1)$$

where

- C_m = Sediment-bound contaminant mixture at downstream locations in the Columbia River
- C_1 = Concentration of component 1 (Columbia River sediment-metal concentration above Priest Rapids Dam)
- C_2 = Tributary sediment-metal concentration
- a = Fraction of C_1 in C_m .

The zinc data are the most useful for Equation 1. For example, Table 4 indicates a fairly uniform concentration for zinc from the various tributaries analyzed. A value of 99 ppm is assigned for C_2 based on an average of the two major downstream sediment-contributing tributaries (the Snake River and Willamette River) and the Young's River. Using 99 ppm for C_2 , the mixing fraction and a corresponding dilution factor, DF, are calculated as follows:

$$DF = 1/a = 1/[(C_m - C_2)/(C_1 - C_2)] \quad 2)$$

Similar assumptions and calculations for the other metals yield calculated dilution factors listed in Table 5. This tabulation demonstrates that lead, zinc, and cadmium exhibit similar downstream dilution trends consistent with the known inputs from mining activities. Additional discussion and application of sediment dilution is provided together with related radiotracer results in the summary and discussion.

Areal and Vertical Distribution Patterns in Young's Bay Sediments. The mouth of Young's Bay opens directly to the main channel of the Columbia River near Astoria, Oregon (Figure 5). Sediment accumulation in the bay is predominantly from the Columbia River and estuary.

Results for the sediment texture and mining-related metals for the sediment quality survey of Young's Bay are summarized in Figure 5. Metal concentrations in surficial sediments from 70 grab sample locations were generally co-variant with sediment texture (Johnson and Cutshall 1975). Metal concentrations are all based on an acid leach method.

The most obvious general pattern by the metal depth profiles shown in Figure 5(a) is the strong correlation between metal concentration and sediment texture. Areas with the highest fine-sediment contents exhibit the highest metal concentrations. This occurs in protected areas such as in the Skipanon Waterway near Warrenton (core site A), behind the solid-fill causeway on the western side of the entrance to Young's Bay (near core site E), and immediately north of the east end of the Young's Bay Bridge. Strong tidal currents prevent accumulation of fine sediment in the main channels. Likewise, exposed tidal flats, which account for a large portion of the area shown in Young's Bay, are subject to sediment mixing and redistribution from natural and/or human activities.

Of the metals shown in Figure 5(b), copper is most consistently correlated with depth-textural variations. The distribution patterns for copper are judged to be caused primarily by naturally occurring copper concentrations because the moderately elevated copper from upstream mining is diluted to natural background levels in Young's Bay. Total fluoride, which reflects the bulk composition/mineralogy (Johnson and Cutshall 1975), has a pattern similar to copper, lending support to the view that the copper is present at naturally occurring levels. In contrast, cadmium, zinc, and lead show depth distribution patterns in the lower core sections that are clearly *not* related to texture alone. For example, elevated metal concentrations appear in the upper sections of cores with fine-textured sediments, but significant departures from this pattern occur with depth. The most dramatic departure is shown in core I. This core exhibits an inverse relationship between texture and metal concentration for lead, zinc, and cadmium. The depth below which no Hanford Site radioactivity was detected is indicated with a solid circle on the texture profiles shown at the top of Figure 5(a). Sediment below this depth is taken to be pre-Hanford Site in age (deposited before 1944). The very low metal content of the deeper, fine-textured sedimentary layer suggests this horizon may be pre-mining era in age. This pattern is also evident in cores C, G, and J.

Cores exhibiting a consistent positive relation between texture and metal content are in the protected areas where fine sediment can accumulate.

Table 5. Sediment Dilution Factors Based on Two-Component Metal Mixing Model (Metal Concentrations from Table 1 for <62 μm Size Fraction; Calculated Using Equation 2 Where C_2 is Average of Ice Harbor, Willamette River and Young's River Values for Tributary Input Concentrations).

	Dilution Factor			
	Zinc	Cadmium	Lead	Mean
McNary Dam	1.6	1.3	1.8	1.6
Prescott, OR	7.7	9.1	6.3	7.7
Young's Bay	13	21	--	17
Mouth/Astoria Canyon	>20	--	--	--

These are also the cores in which Hanford Site radionuclides were still detectable over the full length of core recovered, e.g., core site E behind the solid fill causeway. The fairly uniform concentrations of zinc in the upper 20-cm section of this core (apparent sedimentation rate of 4 cm/yr) suggests the input rate of this metal to the river was fairly constant during the period of interest for this study. This is consistent with the uniform zinc concentration versus depth for the core section corresponding to the early-to-mid 1970's time period in the Lake Roosevelt core discussed previously.

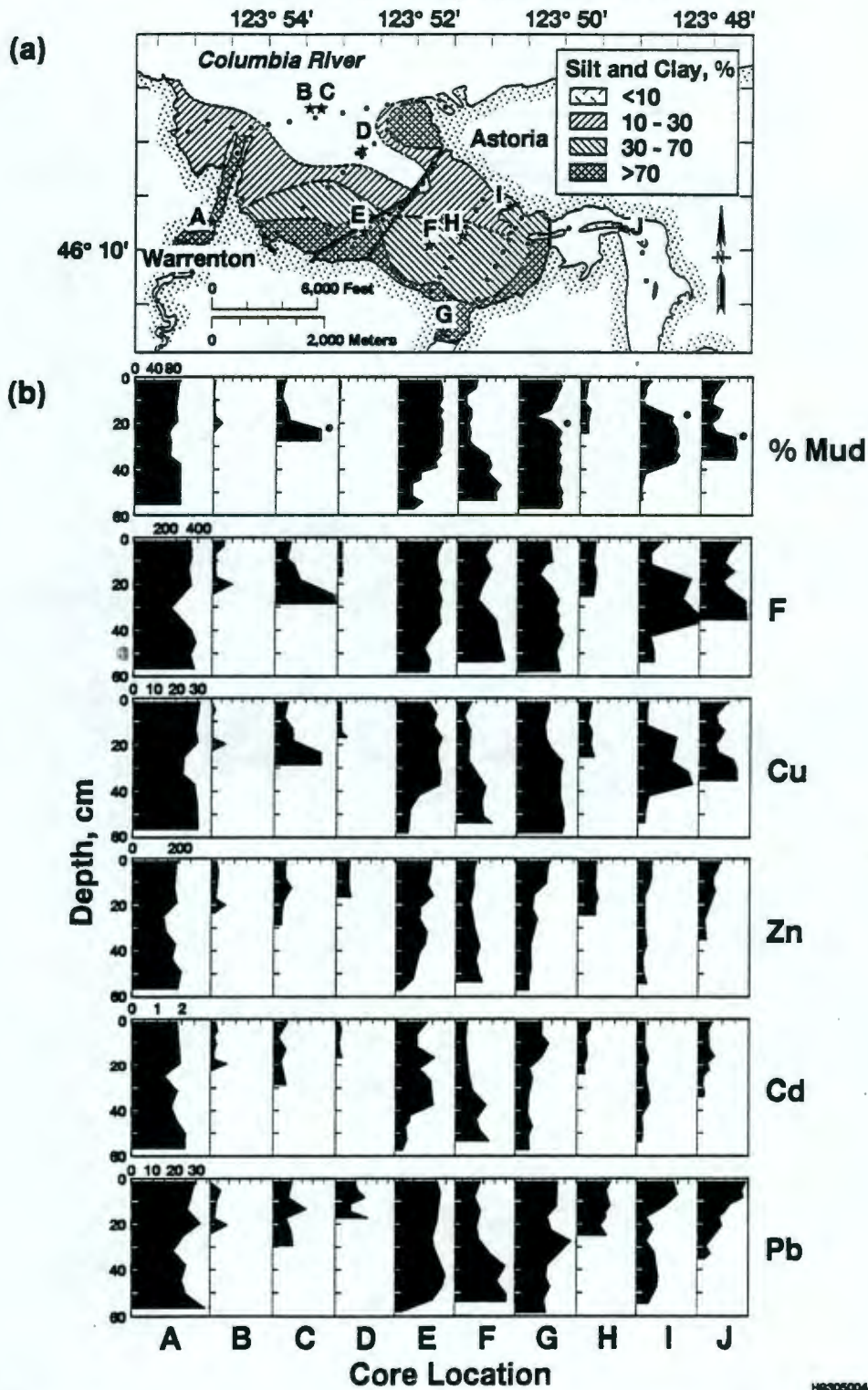
The wide range of areal and vertical metal concentrations illustrates the importance of accounting for the impact of upstream sources on downstream locations and the effect of grain size and depositional history on metal concentrations. In addition, some of the depth profiles illustrate the effects of natural mixing processes in a high-energy environment. The data also suggest that deeper or shielded sediments may be useful as site-specific background reference concentrations.

Metal Content versus Median Grain Diameter (Miller Sands). Miller Sands sample sites adjacent to the main channel of the Columbia River are shown in Figure 6. Baseline physical and chemical conditions were assessed at this site for a dredge spoil disposal, stabilization, and habitat development study (Cutshall and Johnson 1975). Samples of the inner cove, marsh area, and island (previous dredge spoil and naturally deposited sediments) were collected and treated with a reducing acid leach procedure intended to remove surface or adsorbed metals.

If the dominant metal-sediment association is surface area-related sorption and the sediment particles can be approximated as uniform spheres, metal concentration should be proportional to median particle diameter and specific surface, cm^2/g (Sayre et al. 1963). The expected response in this case is:

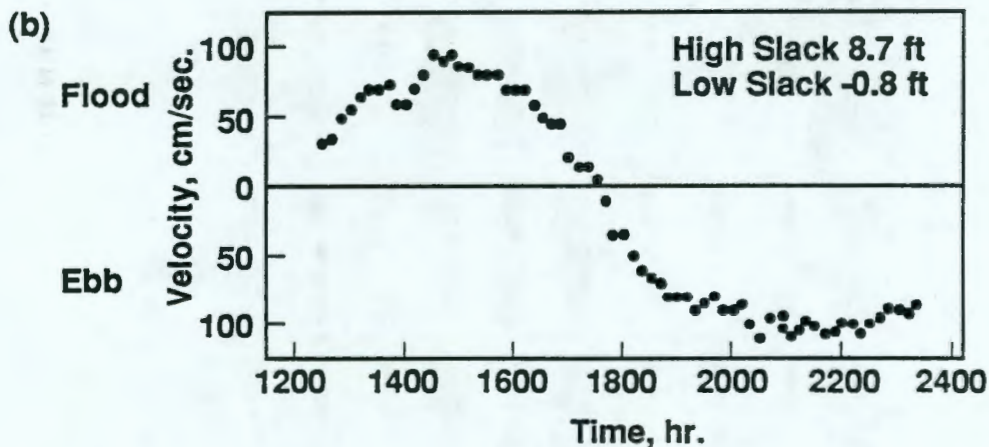
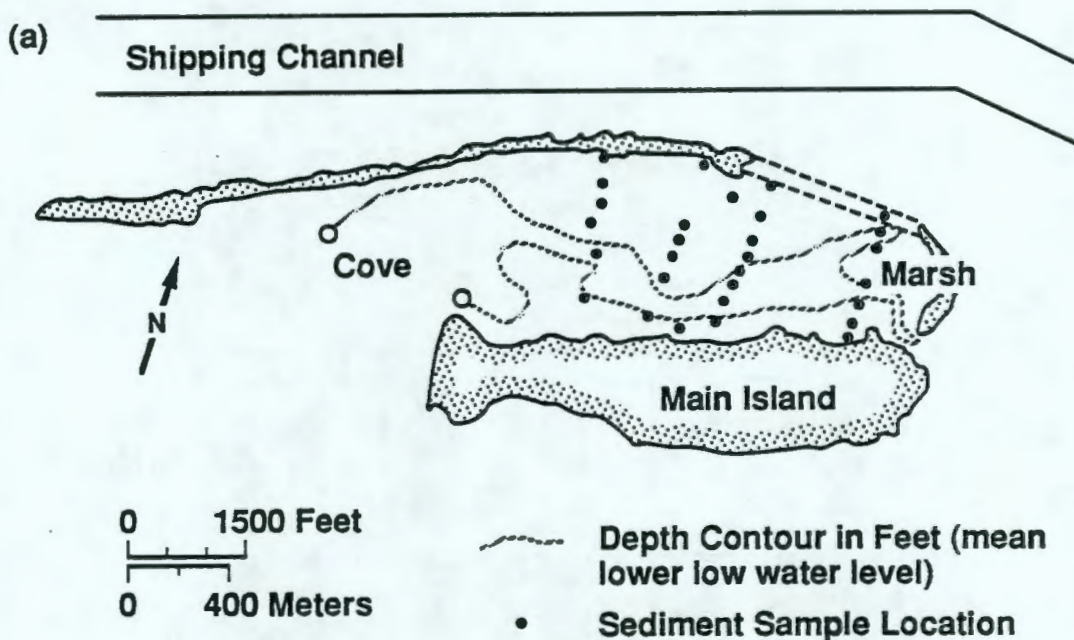
$$C_1 = C_2[d_2/d_1] \quad 3)$$

Figure 5. Sediment Texture and Heavy Metal Content, Young's Bay, Oregon, June 1974.



Inset (a) shows surface grab (•) and core locations (★) and surface sediment texture. Depth distribution patterns (b) for sediment texture (wt% passing 62 μm screen) and acid extractable heavy metal content ($\mu\text{g/g}$). Core locations are keyed to the site map (a) with letters A through J. The depth below which no Hanford Site radioactivity was detected is designated with a solid circle on the texture profiles; the absence of this flag indicates Hanford Site radionuclides were detected over the full length of the core.

Figure 6. Sample Site Locations at Miller Sands, June 1974.



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The island is located immediately adjacent to the main shipping channel and approximately 30 km upstream from the mouth near Harrington Pt. (see Figure 13). Inset shows tidal current velocity adjacent to island over a 24-hour period during low river flow (August 1974). The velocities shown are more than sufficient to prevent fine sediment accumulation, illustrating one reason for the limited occurrence of fine sediment in the estuary proper, as reported in Hubble and Glenn (1973).

where

- C_1 = Higher metal concentration for the sample with a median grain diameter of d_1
 C_2 = Lower concentration with the larger grain diameter of d_2 .

This relationship is illustrated in Figure 7, including an example calculation. Nonspherical sediment particle shapes and particle aggregation may cause deviation from this idealization. Mining-related metal concentrations for sediment samples from the sites shown in Figure 6 are plotted as a function of median grain diameter (Figure 8). Although there are deviations from linearity at the lower end of the concentration ranges for lead and zinc, the four metals approximate the idealized distribution of Figure 7. A similar linear relationship was also reported for ^{65}Zn concentrations in the fine- to medium-sand size classes of bed sediments of the upper portion of the Columbia River estuary (Hubbell and Glenn 1973, Forster 1972). The average shale concentration is shown as a horizontal dashed line for reference. The deviations from linearity, most apparent for lead and zinc, appear to be related to the samples from the main or older island and marsh. These sediments may be pre-mining in age or may have been subjected to natural leaching and/or depletion by plant growth.

Sediment Quality. Some guidelines for evaluating sediment quality survey data suggested from the above considerations are as follows.

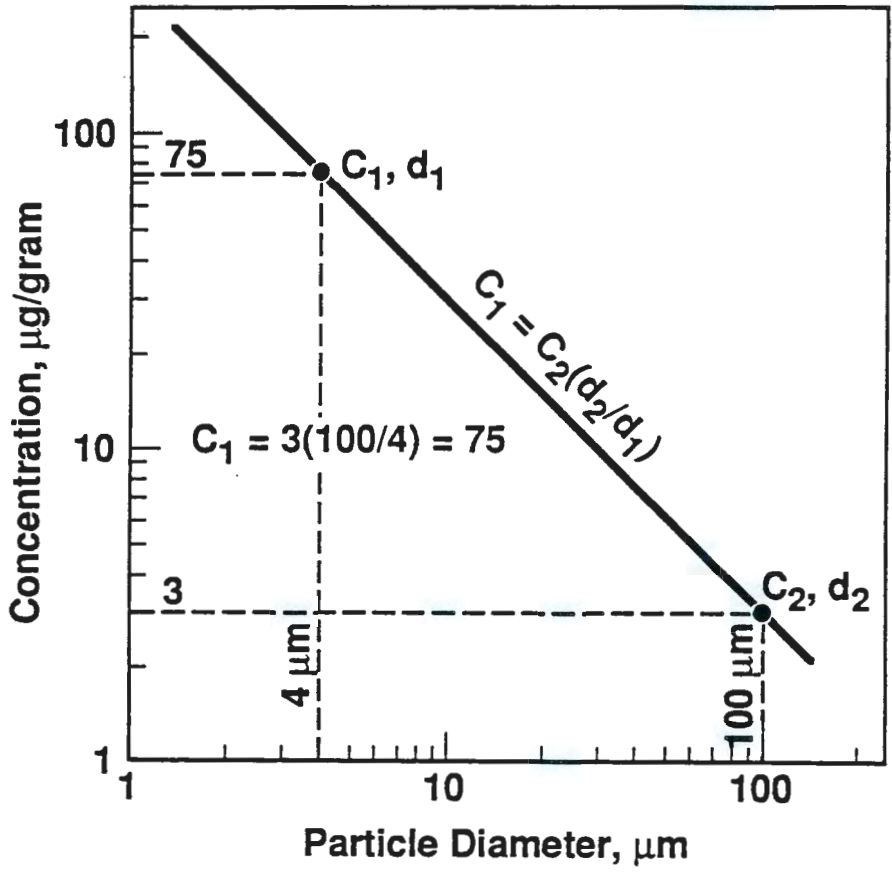
Metals. Only like size-class ranges of reference standards and survey samples should be compared (e.g., world average shale versus the fine-silt-to-clay size range as shown in Figure 8). Alternatively, samples can be sized so that only fine-silt-to-clay size fractions of a local reference standard and survey samples are compared. In lieu of site-specific background, the world average shale (Bowen 1966) appears to be a suitable surrogate for a reference standard. Finally, historical contaminant inputs need to be addressed in assessing local sites. In these cases, a higher "background" threshold level may be needed to allow for elevated "ambient" contaminant concentrations from upstream sources.

Synthetic Contaminants. Artificial contaminants require a different approach because there is no "natural" background reference as discussed above for heavy metals. In these cases, ingestion-based toxicity data may be used to set sediment quality/concentration criteria. One approach is to use median grain diameter and analytical results from the sediment survey, adjust the observed concentration to a fine-silt-to-clay size equivalent using the surface area relationship (Equation 3), and compare this concentration to the ingestion-based toxicity standard for appropriate indicator test species. Site-specific grain-size versus sediment concentration data could be determined in laboratory experiments to improve the sediment or particle-size-dependent estimate. In both the above cases, an appropriate sediment size class must be chosen for reference purposes.

Radiotracer Results.

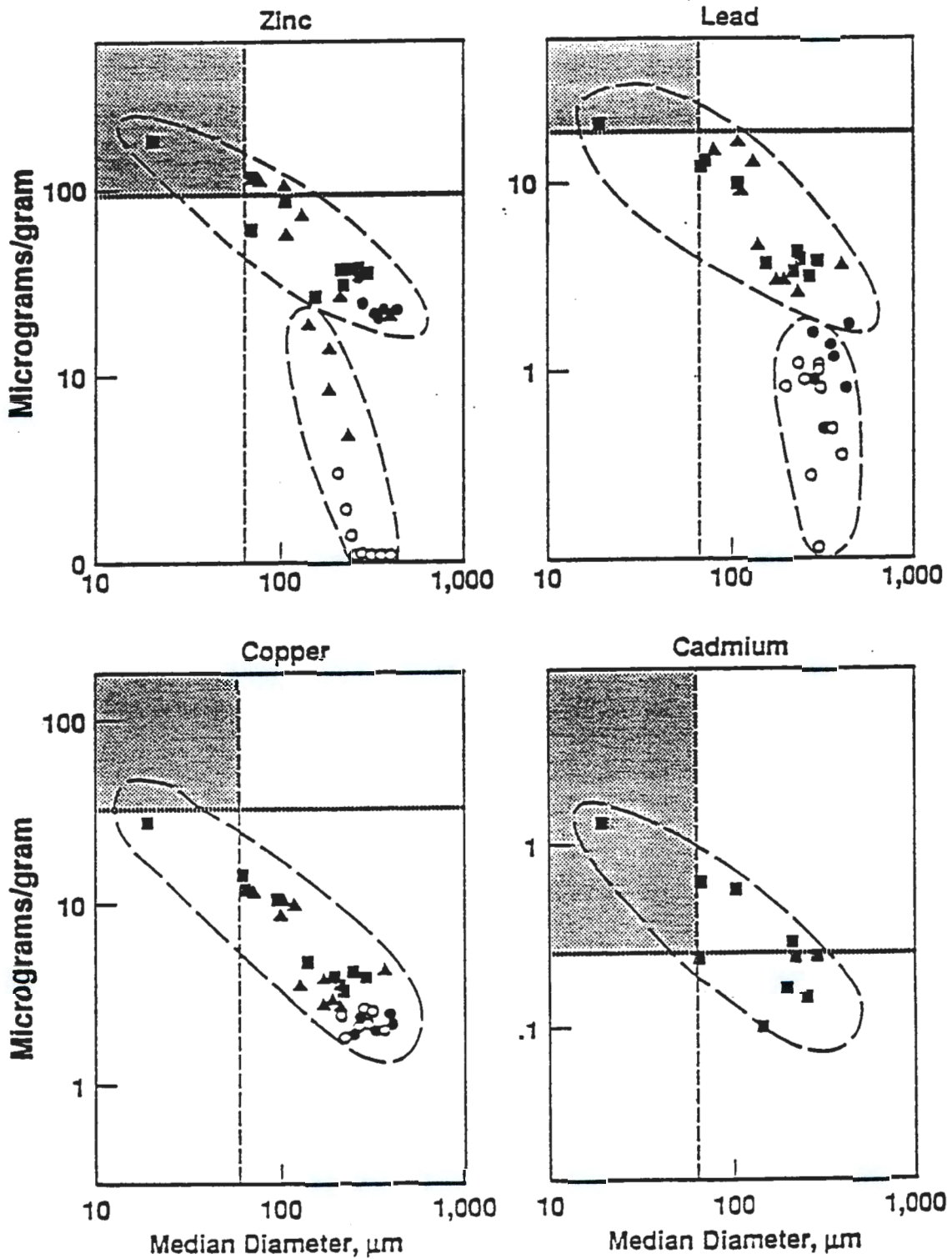
Suspended Sediment and Radionuclide Transport. Time series measurements in the water column below McNary Dam following shutdown of the last once-through production reactor are shown in Figure 9. Primary attention was given to ^{65}Zn for this plot because of the relative amount of this radionuclide and ease of measurement. Additionally, other radionuclides (^{54}Mn and ^{60}Co) were

Figure 7. Theoretical Relationship Between Contaminant Concentration and Particle Diameter Based on the Assumption that Sediment Particles Are Uniform Spheres of Diameter, d .



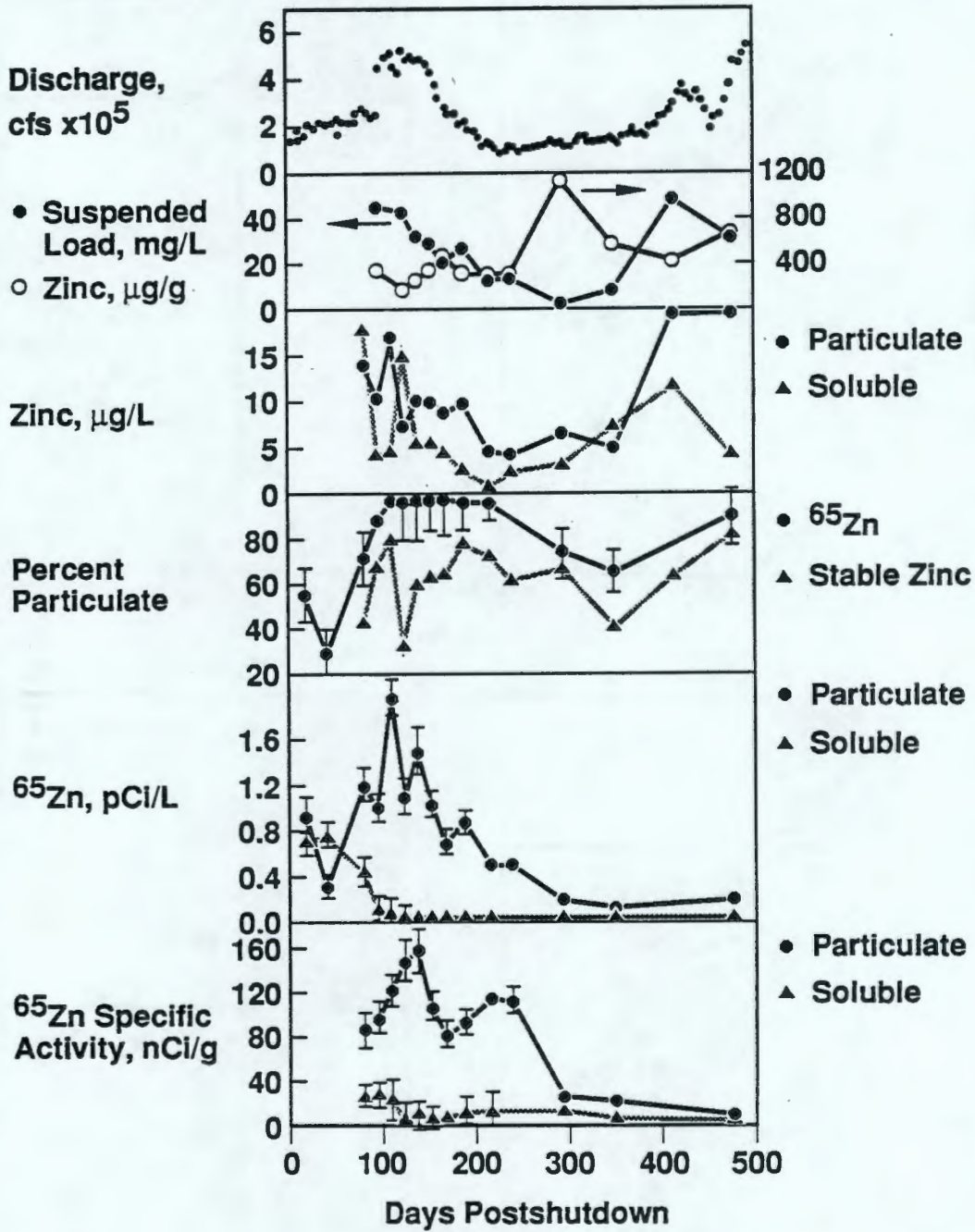
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Figure 8. Heavy Metal Content Versus Median Grain Size.



World average shale concentrations are shown as horizontal line. Vertical dashed line (62 μm) corresponds to the approximate textural equivalent of the rock term "shale." The shaded area to the left of the vertical line and above the horizontal line designates above-background concentrations.

Figure 9. Soluble and Particulate ^{65}Zn and Stable Zinc Concentrations in River Water Below McNary Dam Following Reactor Shutdown on January 17, 1971.



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The abrupt increase in discharge in 1971 (ca. day 100) was caused by a reservoir storage and release or drawdown test.

shown to be influenced by additional inputs from N Reactor, a reactor with a secondary coolant loop operated between 1962 and 1986. However, ^{65}Zn and ^{152}Eu from this source were insignificant based on effluent records and other considerations (Johnson 1979).

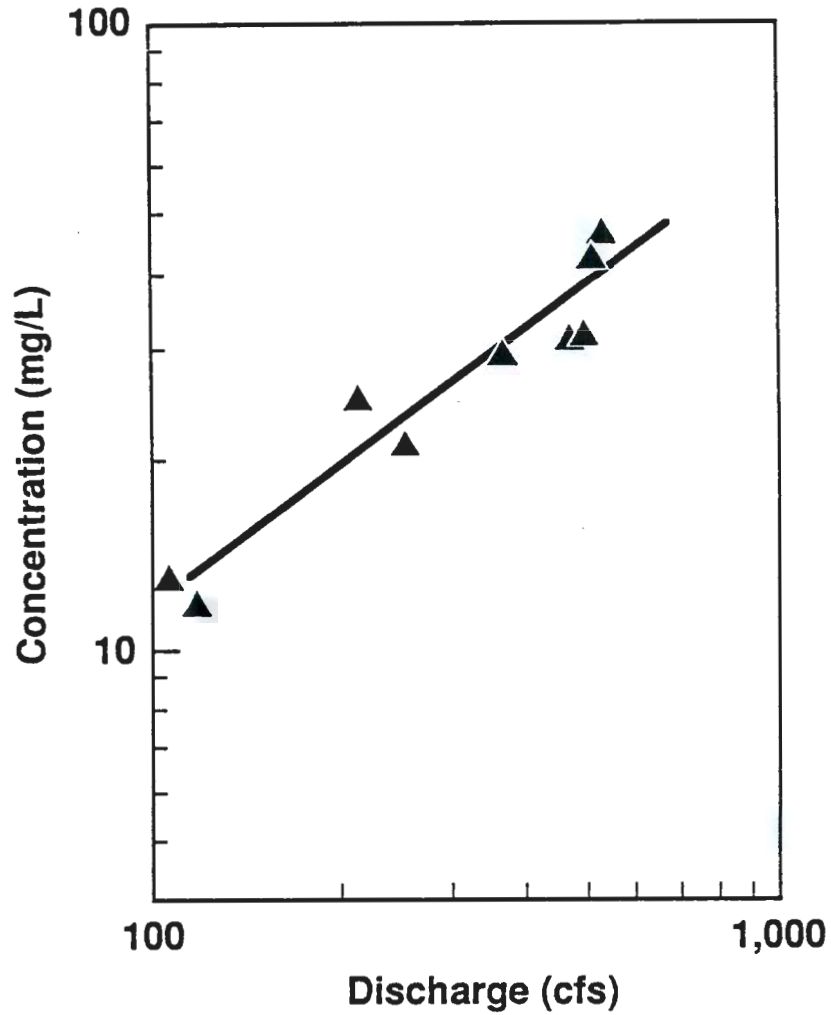
Figure 9 also illustrates the importance of stream velocity or discharge in resuspension of sediment-bound contaminants. The corresponding suspended sediment load plotted as \ln (concentration) versus \ln (discharge) below McNary Dam is shown in Figure 10. The linear response (Graf 1971) implies scour or resuspension of deposited sediment from above McNary Dam. In addition, the stable zinc data shows increasing sediment-zinc concentrations as the discharge rate drops (i.e., as stream velocity decreases, only the finer sediment fractions are in transport). The observed concentrations are very similar to the size fraction results for the composite Lake Wallula sediment. For example, zinc concentrations in the suspended sediment are similar to the $<62\text{-}\mu\text{m}$ fraction during high discharge conditions and approach the concentration for the $<4\text{-}\mu\text{m}$ fraction during low flow. These particle-size-related metal concentrations are consistent with previous studies of suspended sediment particle size versus discharge below McNary Dam (Haushild et al. 1966). For example, during the low flow period, nearly 90% of the depth-integrated, suspended sediment in transport was in the $<4\text{ }\mu\text{m}$ size fraction; during high discharge, approximately 50% of the depth integrated total was $<62\text{ }\mu\text{m}$.

Note that the total radiozinc increased in direct response to Columbia River discharge. This is attributed to resuspension of sediment-bound radiozinc and not to new input. For example, during the years of reactor operations, the radionuclide concentrations in river water decreased during the annual spring high-discharge period. This pattern was complicated by resuspension; however, even with the resuspension, a decrease in river water radionuclide concentration was observed during high river flow (Perkins et al. 1966). Thus, the pattern of increasing ^{65}Zn with discharge shown in Figure 9, as compared to a decrease during reactor operating years, is evidence that little if any additional radiozinc was added to the river from reactor operations during the post-shutdown period. Figure 9 also shows that nonparticulate radiozinc declined rapidly following termination of new input to the river. This further emphasizes the particulate nature of the radiozinc during the post-shutdown period. In contrast, during the years of reactor operations, the distribution of ^{65}Zn between water and particulates was about equal at McNary Dam (Perkins et al. 1966).

The specific activity (ratio of radiozinc to stable zinc expressed as $\text{nCi } ^{65}\text{Zn/g Zn}$) for particulate and soluble fractions (passing a $0.4\text{-}\mu\text{m}$ Nuclepore filter) suggests an irregular discharge-resuspension effect. This more complicated pattern compared to ^{65}Zn only is probably because of the differential mobilization of sediment-bound zinc from above the Hanford Site that mixed with the radiolabeled suspended sediment. The result was a specific activity-discharge pattern that reflects not only erosion of sediments containing ^{65}Zn and stable zinc from below the Hanford Site, but also erosion of sediments containing stable zinc from above the Hanford Site.

The specific activity (see Figure 9) also suggests post-shutdown, particle-bound radiozinc did not re-equilibrate with stable zinc in the aqueous phase (i.e., the specific activity should be the same for the soluble and particulate phases if the isotope of zinc is in equilibrium with stable

Figure 10. Suspended Sediment Concentration Versus River Discharge Below McNary Dam, 1971-1972.



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Mean daily flow rates at McNary Dam were used for the sample collection dates.

zinc). This suggests the adsorbed radiolabeled zinc "pool" is irreversibly bound or exchanges very slowly with zinc in solution. This is in contrast to laboratory studies that have shown complete isotopic equilibrium of ^{65}Zn and stable zinc associated with clay minerals and the aqueous phase (Johnson 1979). Sorption by natural sediments apparently involves more complex interactions than simple clay mineral-ion-exchange bonds. Possible explanations include coordinate covalent bonding between hydrous oxide coatings on sediment particles and divalent zinc (Johnson 1979). Once the "radiotracer" is incorporated into this bound fraction, it may not readily exchange with the stable counterpart in solution. Similar kinetic hindrances have been noted in laboratory studies of metal-ligand-tracer exchange reactions (Turse and Rieman 1961). This interpretation is consistent with earlier observations (Johnson et al. 1967) showing that very little ^{65}Zn could be displaced from Columbia River sediment with neutral, one normal NH_4Ac or seawater, whereas dilute (0.05 molar) copper sulfate easily displaced a major fraction of the sediment-bound ^{65}Zn . Thus retention of zinc and probably other heavy metal solutes by Columbia River sediment involves more complex bonds than simple ion exchange.

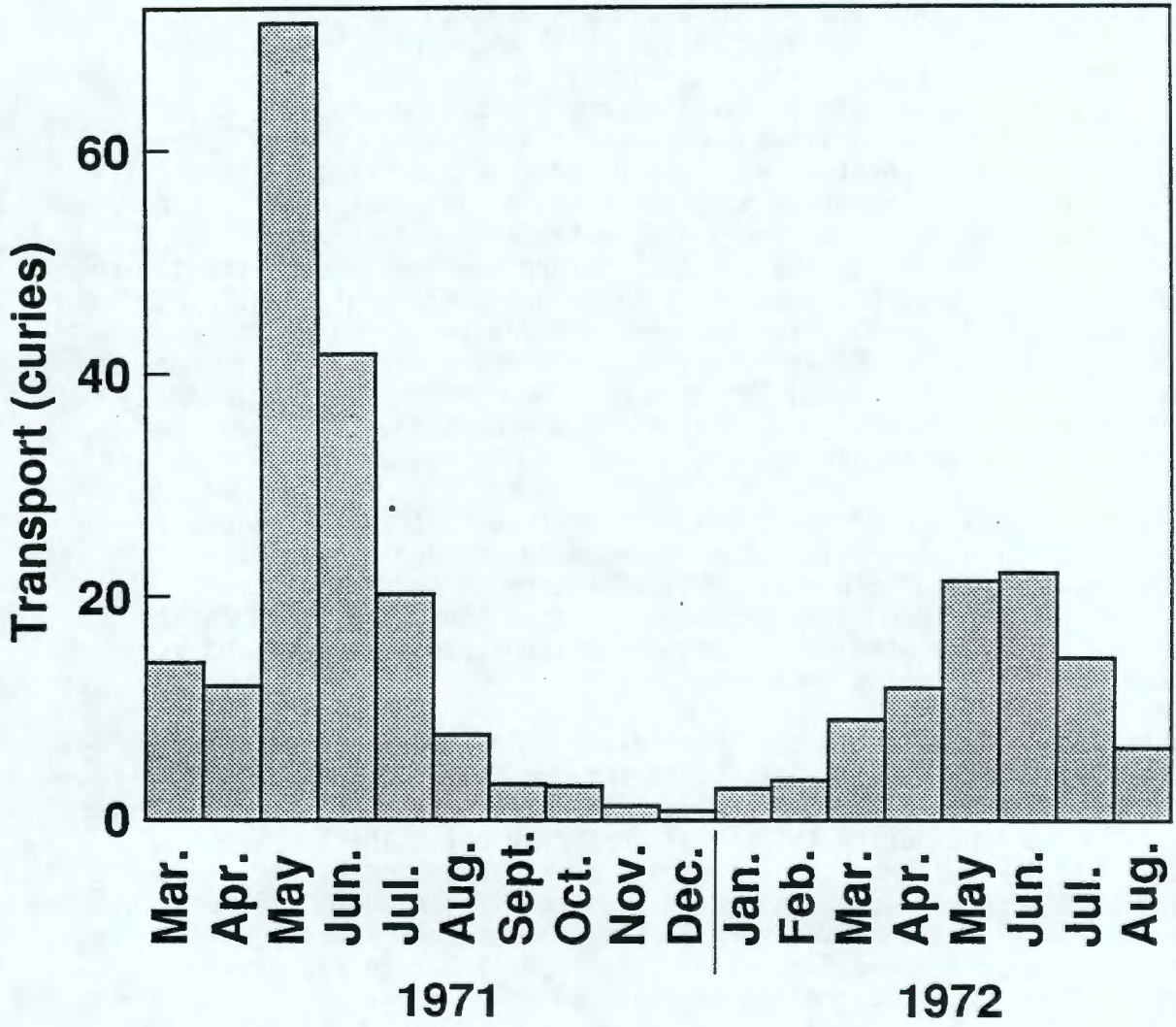
The above feature of the post-shutdown transport and behavior of radiozinc may allow some distinction to be made between biological uptake via sediment ingestion and uptake via solute absorption and or solute-phytoplankton-based food chain exposure routes. These are fundamental questions directly related to evaluating sediment exposure pathways as discussed in the following section.

Transport and Biodynamics in the Lower River. Transport of radiozinc at Prescott, Oregon, (Figure 11) exhibited the same river discharge-resuspension relationship observed below McNary Dam. Radiozinc transport increased with increasing discharge during the annual spring runoff. The "spike" in transport for May 1971 was in part caused by resuspension of bed sediments in response to a reservoir storage and release test intended to aid out-migrating salmon smolts. Release of water following a short-term storage period during the spring freshet doubled flow rates below McNary Dam in less than a day. The flow perturbation occurred throughout the lower river.

The area under the decay-corrected, monthly mean totals of ^{65}Zn for 1972 shown in Figure 11 is approximately 50% of the area for the 1971 monthly means. Multiple regression of decay-corrected ^{65}Zn concentration versus time and discharge resulted in half-times of decline of 360 ± 99 days and 445 ± 56 days at McNary Dam and Prescott, respectively (Johnson 1979).

Estuarine biota collected from 0 to 700 days post-shutdown at sites H, C, and A (see Figure 3) showed a median half-time of decline of approximately the same rate as indicated for particle-bound ^{65}Zn transported into the estuary in water, i.e., 350 to 400 days as suggested in Figure 12. The only departure from this generalization was for the pond weed *Potamogeton*, near McNary Dam, which exhibited a decay-corrected half-time of decline of ca. 75 days (Johnson 1979). However, this aquatic plant takes up major and micronutrients directly through its leaves. Because there was very little dissolved ^{65}Zn available after the first 2 to 3 months post-shutdown (i.e., only particle-bound ^{65}Zn was available [see Figure 9]), incorporation of additional ^{65}Zn into *Potamogeton* tissue was greatly diminished.

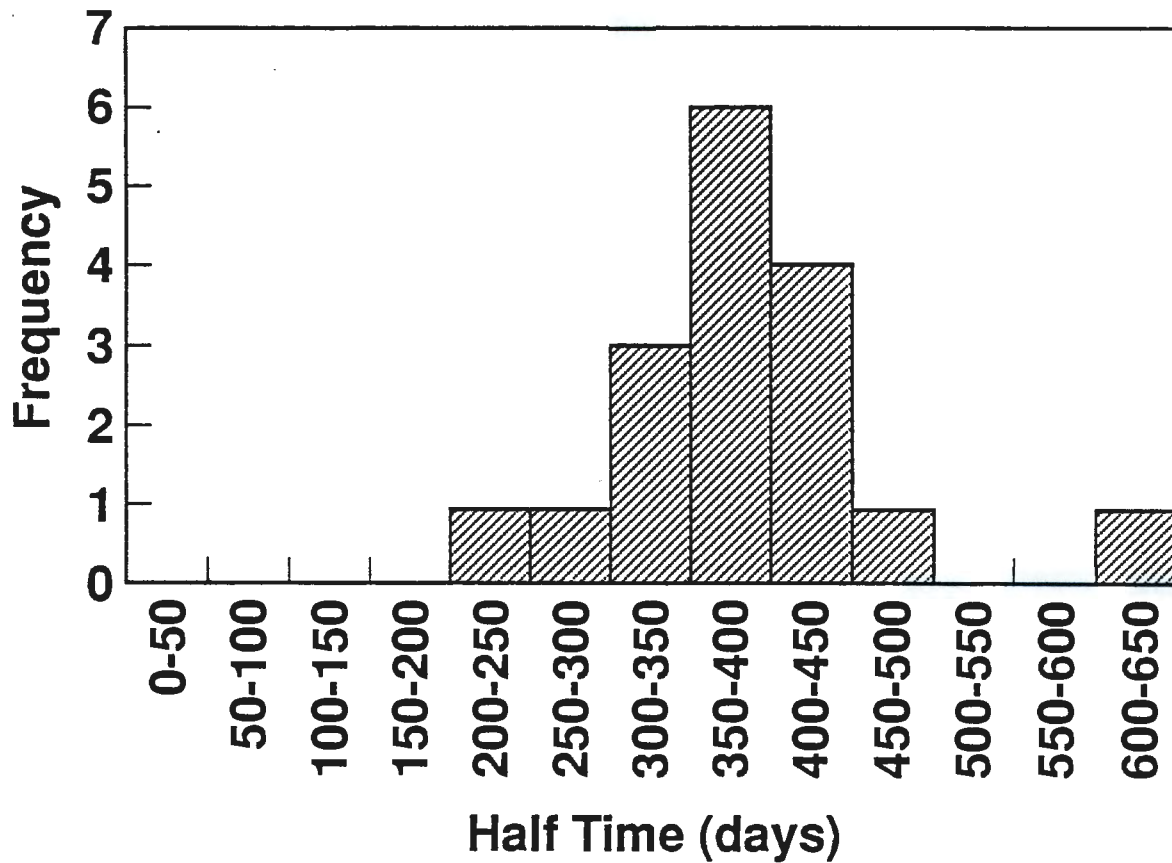
Figure 11. Radionuclide Transport at Prescott, Oregon, 1971-1972.



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Mean monthly flow rates were combined with the corresponding monthly composites of daily samples to estimate total curies of ⁶⁵Zn per month (decay corrected to time of shutdown, January 1971).

Figure 12.. Decay-Corrected Half-Times of Decline for ^{65}Zn Specific Activities in Selected Biota from the Columbia River Estuary, 1971-1972.



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The decay correction was made by subtracting the physical decay constant (0.693/245 days) for ^{65}Zn from the slope of the regression of \ln (specific activity) on time and then solving for the resultant half-time (Johnson 1979). Average standard deviation of the regression coefficient was $\pm 18\%$. Species included in the frequency plot are listed in Table 6 (see Figure 13 for trawl locations).

Figure 12 is based on a diverse group of estuarine biota that includes (1) various species of juvenile fishes of two size classes (0 to 10 cm and 10 to 20 cm), (2) small crustaceans, and (3) both freshwater and marine mussels (Table 6). A wide range of biological half-lives (e.g., <30 to 300 days [Johnson 1979]) was expected for biota with such divergent food sources and metabolic activities. Yet the median half-time of decline (decay corrected) is about the same magnitude as the decay-corrected rate of decline for the radiozinc in transport at Prescott (see Figure 11). The apparent concordance between the rates of decline in water and biota suggested a common exposure path involving either direct sediment ingestion and/or consumption of sediment-ingesting prey species. The hypothesized sediment-consumer pathway should follow simple source-receptor biodynamics. For this purpose, an uptake and loss equation is used with an exponentially declining source term, i.e., the rate of decline of particle-bound ⁶⁵Zn transported into the estuary. If the hypothesis is correct, the apparent rate of decline of the receptor should be invariant over a range of reasonable biological half-lives.

The first-order differential equation and its solution used to model the above conditions are summarized from Johnson (1979) as follows:

$$dS/dt = \frac{\text{uptake}}{BCe^{-kt}} - \frac{\text{loss}}{uS} \quad (4)$$

$$S = S_0 \left[\left[\frac{u}{u-k} \right] e^{-kt} + \left[1 - \frac{u}{u-k} \right] e^{-ut} \right] \quad (5)$$

where

- S = ⁶⁵Zn specific activity (nCi/g Zn) in the receptor organism as a function of time, t
- S₀ = Steady state or initial specific activity just before shutdown
- u = [(0.693/T_b) + (0.693/245 days)]
- T_b = Biological half-life of zinc in the receptor organism and 245 days is the physical half-life of ⁶⁵Zn
- k = The effective source term decay constant = [(0.693/T_w) + (0.693/245 days)]
- T_w = Washout half-time in days
- B = 0.693/T_b
- C = A boundary condition that specific activity of the organism equals its steady-state specific activity just before reactor shutdown or termination of the source = S₀*(u/B)

Semi-log plots of Equation 5, i.e., log (S) versus time from 0 to 1,000 days for various values of T_b (30, 300, and 3,000 days) and T_w (Johnson 1979) are curvilinear. The moderate curvature occurs in the early post-shutdown period (~100 days). As T_w values approach the observed washout half-time of about 1 yr, the apparent (decay-corrected) slopes of the straight-line portions of the plots for values of T_b between 30 and 3,000 days converge on values of about 1 yr; i.e., they are approximately independent of biological half-life for values of T_w ≥ 1 yr. Mathematically, this means that the second exponential term in Equation 5, e^{-ut}, which accounts for biological turnover or depuration, is significantly smaller than e^{-kt}.

Based on the biokinetic argument summarized above, the sediment ingestion hypothesis is not rejected. Alternatively, sediment ingestion may

Table 6. List of biota and feeding habits, Columbia River estuary (summarized from Haertel and Osterberg, 1967).

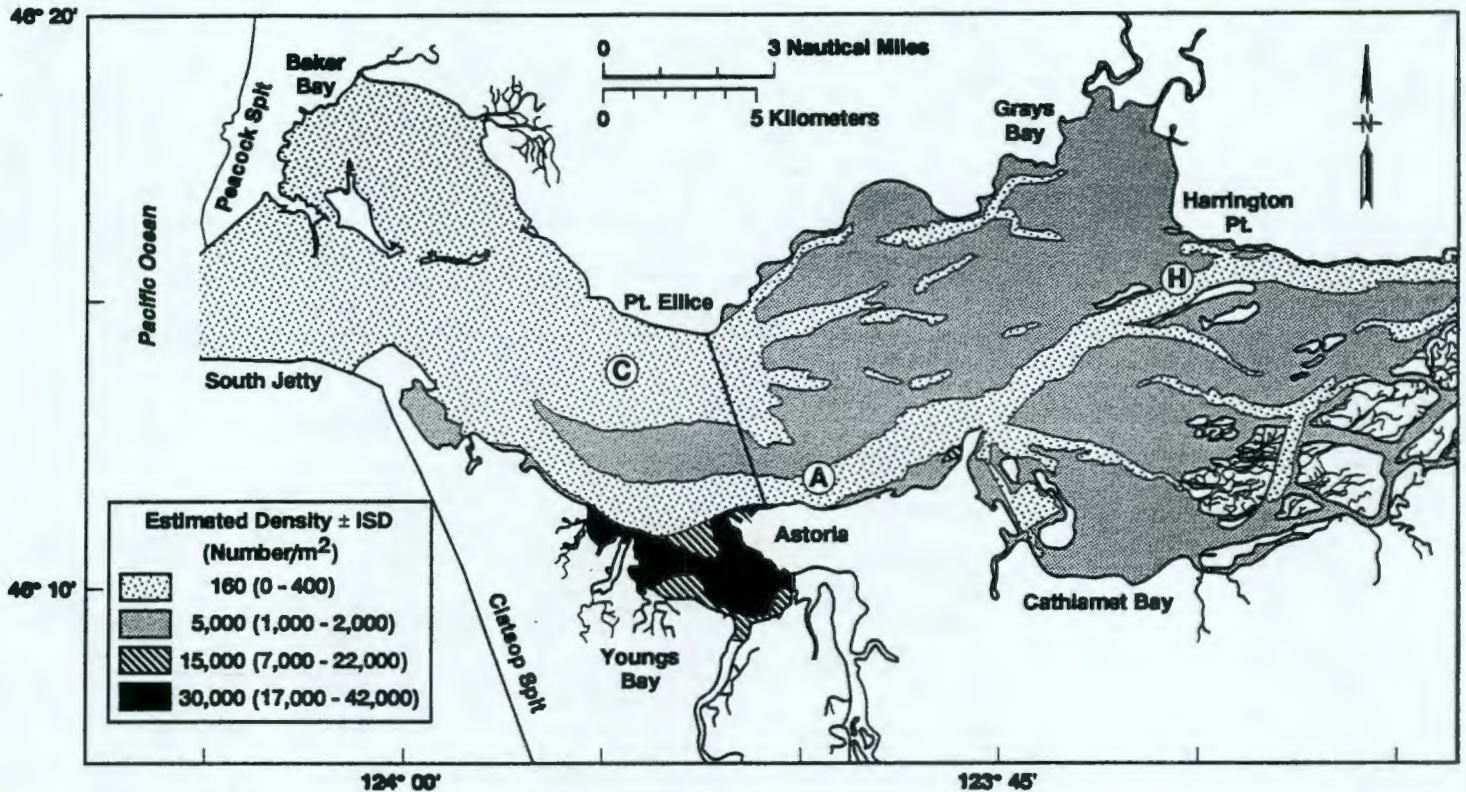
Common Name	Scientific Name	Food Type
Shiner perch	<i>Cymatogaster aggregata</i>	P, B
Pacific staghorn sculpin	<i>Leptocottus armatus</i>	<u>E</u> , B
Prickly sculpin	<i>Cottus asper</i>	<u>B</u> , F
Starry flounder	<i>Platichthys stellatus</i>	<u>B</u> , P
Pacific tomcod	<i>Microgadus proximus</i>	P, B, F
Sand sole	<i>Psettichthys melanostictus</i>	<u>B</u> , P
Lemon sole (or English sole)	<i>Parophrys vetulus</i>	<u>B</u> , P
Sand shrimp	<i>Crangon franciscorum</i>	P
Freshwater clam	<i>Anodonta oregonensis</i>	FF
Marine mussel	<i>Mytilus californianus</i>	FF
Marine mussel	<i>Mytilus edulis</i>	FF

B = benthos F = fish P = plankton FF = filter feeder
Where more than one food type is indicated, the more dominant form is underlined.

be a significant contaminant pathway for exposure of the biota listed in Table 6. Additional field evidence supporting a benthic sediment ingestion pathway is found in previously reported ecological studies summarized as follows.

Estuarine Benthos Studies. As indicated in Table 6, benthic amphipods and infauna are a major food source for the juvenile fish species evaluated in this study. Amphipods, nematodes, oligochaetes, and polychaetes, which digest sediment, were common in juvenile fish stomachs (Higley and Holton 1975, Higley et al. 1976; Haertel and Osterberg 1967). Benthic amphipods (predominantly *Corophium salmonis*) are the most abundant benthos and are most commonly found in fine sand (0.063 to 0.246 mm) while oligochaetes are most abundant in "mud" (<0.063 mm). However, even in mud bottoms, densities of *Corophium* of 15,000/m² were found in Young's Bay locations with median grain sizes in the range of 2 to 8 μ m (Higley et al. 1976, Slotta 1975). In fact, the second highest *Corophium* densities in the Columbia River estuary were found in the mud bottom tidal flats of Young's Bay (Figure 13) in predominantly clay-size substrates (Higley et al. 1976). *Corophium* builds small tubes in the top 1 to 2 cm of the fine-sediment bottoms and creates a flow of water from which it filters particulates. Thus, fine sediment is likely ingested by these amphipods as well as by predators during capture of this very abundant benthic invertebrate. Their relatively high densities, wide distribution (see Figure 13), and observed occurrence in fish stomachs

Figure 13. Distribution of Benthic Amphipod Habitat and Densities in the Columbia River Estuary (after Higley et al. 1976).



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The areal distributions are based on several transects and interpolations using sediment texture-amphipod correlations. The lowest densities (stippled pattern) coincide with the main tidal channels where the highest flow velocities occur. The maximum extent of saltwater intrusion (bottom of main channel) is in the vicinity of Harrington Pt. during low river flow and exceptionally high tide. The near-surface or shallow water areas over much of the estuary located more than 10 km upstream from the mouth are dominated by fresh water. Bottom salinities (>10 m deep) in the vicinity of Station C are about 15 ‰ at the bottom and less at the surface. Salinities upstream of Astoria are generally less than 5 ‰ at all depths. Thus, the estuary is often characterized as a freshwater-dominated estuary. Large circled letters show trawl locations.

(Higley and Holton 1975) provide fairly direct evidence that *Corophium* play a major role in an estuarine benthic biota sediment-food chain pathway.

The above considerations place more emphasis on estuarine sediment transport processes, especially suspended sediment and particle-size relationships.

Suspended Sediment Particle Size. Particle-size distribution of suspended sediment in the estuary was studied by Hubbell et al. (1971). They evaluated suspended sediment concentration and size distribution as a function of tidal stage and location in the estuary under both high- and low-river-discharge conditions. The suspended sediment study sites were very similar in location to the biological collection stations (see locations C, A, and H, Figure 13). Here they found an average distribution of 72 wt% in the 62- μm to 4- μm size class (silt) and a corresponding 26% in the <4- μm or clay-size class. They also noted the presence of a turbidity maximum in the estuary that appeared to move back and forth with tidal stage and river discharge. The turbidity maximum could have maximized the probability of sediment ingestion by the biota examined in this study. Estuarine circulation, tidal dynamics, and bathymetry act in concert to circulate the silt and clay-size sediment within the estuary, maximizing the opportunity for sediment-bound contaminants to enter the food chain via suspended or resuspended sediment ingestion.

Coastal Zone. Particle diameters of suspended sediment, or "lithogenous particulates," are predominantly <4 μm beyond the river mouth in the near-surface oceanic layer defined by a salinity of <32.5 o/oo (Conomos and Gross 1972). The silt component is apparently carried 20 to 30 km offshore, settles, and is then transported to the north along the seabed. Littoral drift eventually transports the silt into Quinault Canyon located 150 km north of the mouth and >40 km offshore (Cutshall et al. 1986). Thus, particle size separation occurs in the transition zone from estuarine to oceanic conditions. The significance is that filter-feeding biota in the coastal zone adjacent to and northward of the river mouth are exposed to the sediment size-fraction with the highest potential contaminant concentrations. Even though significant dilution by mixing with seawater occurs, the filter feeders concentrate the particulates from the water. This is equivalent to sediment ingestion by biota that live in mud bottoms where clay-size sediments accumulate.

The coastal zone filter-feeder/fine sediment pathway is perhaps of most significance for human exposure considerations. For example, oysters, marine mussels, and clams from the bays immediately north of the river mouth are consumed. In Willapa Bay oysters and marine mussels near the mouth, ^{65}Zn specific activities were nearly the same as or within a factor of 2 of those observed for sediment and biota in the Columbia River estuary during the post-shutdown period (Johnson 1979; Seymour and Nelson 1973). Additionally, concentrations in Willapa Bay oysters during 1962-1963 (avg. 80 pCi/g) were over twice as high as in white fish (fish with the highest ^{65}Zn concentrations) from the lower Hanford Reach (Foster 1964). The fact that river water from the Hanford Reach undergoes over a ten-fold dilution by mixing with tributaries and seawater before reaching Willapa Bay is further evidence for incidental ingestion of particle-bound contaminants (i.e., filtering of particulates from the water. Thus human consumption of marine mussels and oysters from the coastal zone near the river mouth may be as

important as consumption of upper river sport fish for assessment of relative risks from upper drainage basin contaminant loading.

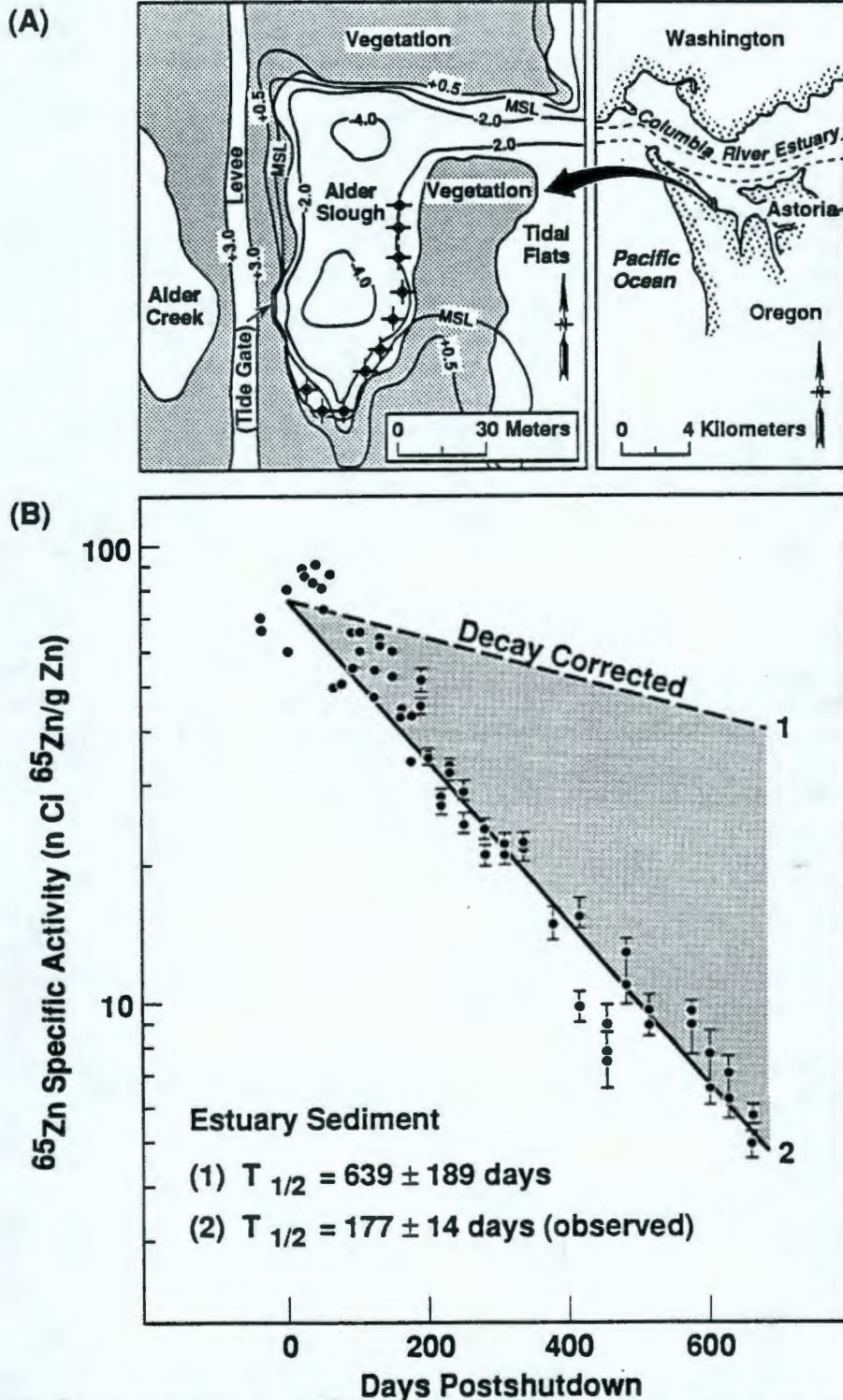
Estuarine Sediment Mass Balance. Approximately 30% of the fine sediment transported into the estuary by the Columbia River is retained (Hubbell and Glenn 1973). Significant accumulations of this fine sediment fraction occur only in protected areas where river and tidal currents are minimal (e.g., certain areas of Young's Bay, Baker Bay). The broad expanse of the estuary and low suspended sediment load do not favor an extensive or rapid buildup of fine-textured bed sediment. The significance of a slow, fine-sediment accumulation rate on contaminant availability in bed sediments of the river-reservoir system, as well as in the estuary, are discussed in the following section.

Bed Sediment Dynamics. The rate of decline in radionuclide concentrations with time post-shutdown in surficial bed sediments provides a measure of the turnover or "flushing" time in this most active portion of the sedimentary column. Radionuclide-depth profiles for sediment cores in key depositional sites yield information on deposition rates, sediment mixing, and event history. Because interpretation of surficial sediment results and core data require different approaches, they are discussed separately as follows.

Surficial sediments. ^{65}Zn results expressed as the ratio of radioactive to stable zinc for surface sediments (top 1 cm) collected versus time in the estuary are shown in Figure 14. Both the estuary and McNary Dam surface sediment composites yield apparent half-times of decline of about 600 to 700 days (corrected for physical decay). These rates appear to be somewhat longer than the apparent washout rate observed for suspended sediment (see Figure 12) and exhibited by the estuarine biota. Since the input rate of radioactivity declined in the years before final shutdown, deeper sediments with higher concentrations may have been mixed with the newly deposited sediments at these sites. This condition may have produced an apparent decline that was somewhat longer than that observed for suspended sediment in transport by the river. Because half-times of decline summarized in Figure 12 are based on biota collected within the more dynamic areas of the estuary (i.e., where water velocities preclude fine sediment deposition, [see Figure 6]), the shorter half-times are reasonable. Biological data for more restricted or protected areas with fine sediment bottoms (e.g., Young's Bay) were not available to test this inference.

Sedimentation rates. Johnson (1979) used radionuclide ratios of two isotopes of europium (^{152}Eu and ^{154}Eu) and $^{60}\text{Co}/^{152}\text{Eu}$ to estimate sedimentation rates behind McNary Dam. Based on these estimates and mass balance considerations, he concluded that the river supplied only enough sediment to maintain 2 to 4 cm/yr of fine sediment over approximately the lower one-third of the reservoir (i.e., where fine-textured sediment accumulated). Beasley and Jennings (1984) later confirmed this low sedimentation rate based on several cores from the lower section of the reservoir. Considering that the long-term annual average suspended sediment concentration of the Columbia River below McNary Dam is on the order of 30 mg/L (Weyl 1970), the slow sedimentation rates are reasonable. Fine sediment accumulation behind the three reservoirs downstream of McNary Dam should be even less. For example, coring attempts in these downstream reservoirs resulted in very little success (Beasley and Jennings 1984). This is consistent with seismic profiling data from behind the dams (Conomos and Gross 1972) indicating very little net

Figure 14. Surficial Sediment ^{65}Zn Specific Activity Versus Time Post-Shutdown. 1971-1972.



Sample locations (⊕) from a protected mud flat at the Alder Slough/Young's Bay study site are shown in inset (A). Samples of the top 1 cm were composited from 10 locations on each sampling date. Water freely exchanged with the Columbia River in response to tidal action (Renfro, 1972). Quiescent conditions at high tide allow fine sediment to accumulate. Line 2 is the variance-weighted linear regression of the observed specific activities on time. Computed half-times and associated error terms from the slope are as shown in the legend. Line 1 is based on the slope of line 2 corrected for radioactive decay ($T_{1/2} = 245$ days). Line 1 represents the rate of decline for sediment-bound contaminants not subject to physical decay or degradation following termination of chronic input to the river system.

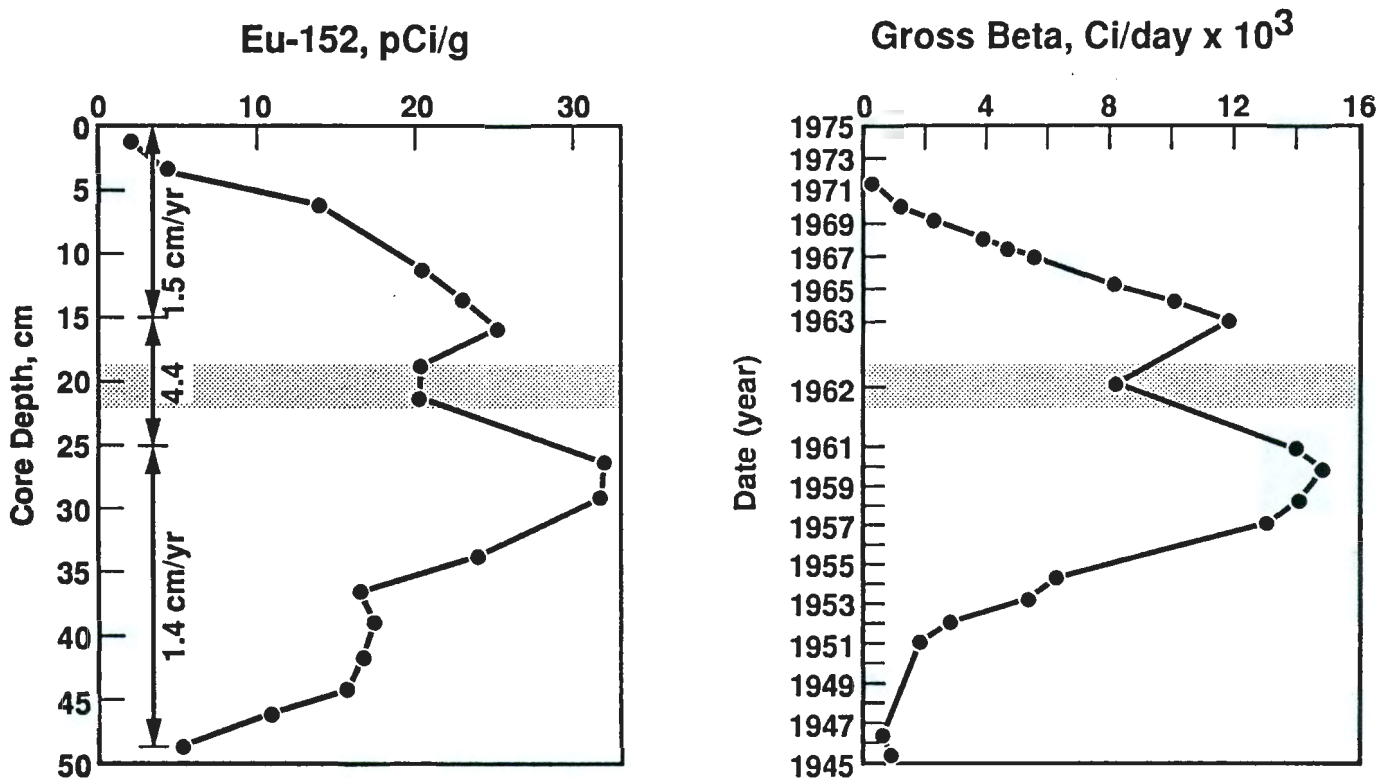
sediment accumulation. Such slow sedimentation rates would allow an extended opportunity for resuspension and downstream transport because burial would be delayed. The significance for the lower river system is that while contaminant storage by bed sediments behind the reservoirs may be small, that which is temporarily stored may be highly susceptible to mobilization during hydraulic perturbations (e.g., high annual runoff, floods, flow manipulation, or drawdown). The fine silt and clay-size fractions are the most susceptible to such perturbations.

Correlation with Radionuclide Discharge History. Event markers are useful for evaluating sediment mixing and deposition rates. Radionuclide-sediment depth profiles should reflect the reactor effluent discharge history (see Figure 2). However, radioactive decay of some radionuclides may distort the depth patterns. This can be remedied by "dating" the core and making the appropriate decay correction versus depth. Decay-corrected ^{152}Eu (half-life of 13.6 yr) versus depth is shown in Figure 15. The date assignments and sedimentation rates shown were based on $^{60}\text{Co}/^{152}\text{Eu}$ ratios for each depth increment or core section (Johnson 1979). The assigned dates or ages were then used to decay correct the ^{152}Eu to the time of shutdown (January 1971). The reactor release rate (annual average total beta activity in kCi/day) was replotted on the same depth-time scale for comparison with the decay-corrected ^{152}Eu . The close correspondence between the reactor discharge history and sediment concentration versus depth indicates there was a relatively undisturbed depositional environment at this location in the reservoir and that the core obtained is a reasonably complete record. In addition, if major scouring occurred at this site, sections would be missing, and thus the input rates and profile would not match. The inflection due to changes in operating conditions (see Figure 2) can be used as a discrete "event" marker to check the accuracy of the age assignment. For example, the inflection that occurred in 1962 because of changes in coolant water pretreatment coincides with the assigned age for this depth increment. The sediment depth above this level divided by the time difference (1975 to 1962) yields an average sedimentation rate of $(20 \text{ cm}/13 \text{ yr}) = 1.5 \text{ cm/yr}$. This is in close agreement with the Beasley and Jennings (1984) estimate for this portion of the reservoir. The short period of apparently more rapid sedimentation shown in the reconstruction on Figure 15 is unexplained.

One other feature of the input rate versus depth profile comparison shown in Figure 15 is the "lag" in decline during the accumulation period following shutdown in 1971 and 1975 when the core was collected. Even though input terminated, the presence of radiotracer persisted up to 4 yr following shutdown. Apparently, scour or resuspension from upstream sources continued to redistribute a portion of the bed sediment inventory. Such effects are even more pronounced in the higher energy environment of the estuary as suggested by the ^{152}Eu depth profiles for cores in Young's Bay (Figure 16).

Figure 16 shows the vertical and areal distribution of ^{152}Eu concentrations based on 70 grab samples and 10 cores collected in June 1974. Highest concentrations in surficial sediment are associated with areas of fine sediment accumulation, as described previously for the heavy metal distributions. The reactor effluent discharge history is not obvious in any of the cores as observed in the McNary Dam core (see Figure 15). Perhaps the most significant observation is that even $4\frac{1}{2}$ yr after shutdown, surface concentrations of this radionuclide, which was uniquely of Hanford Site

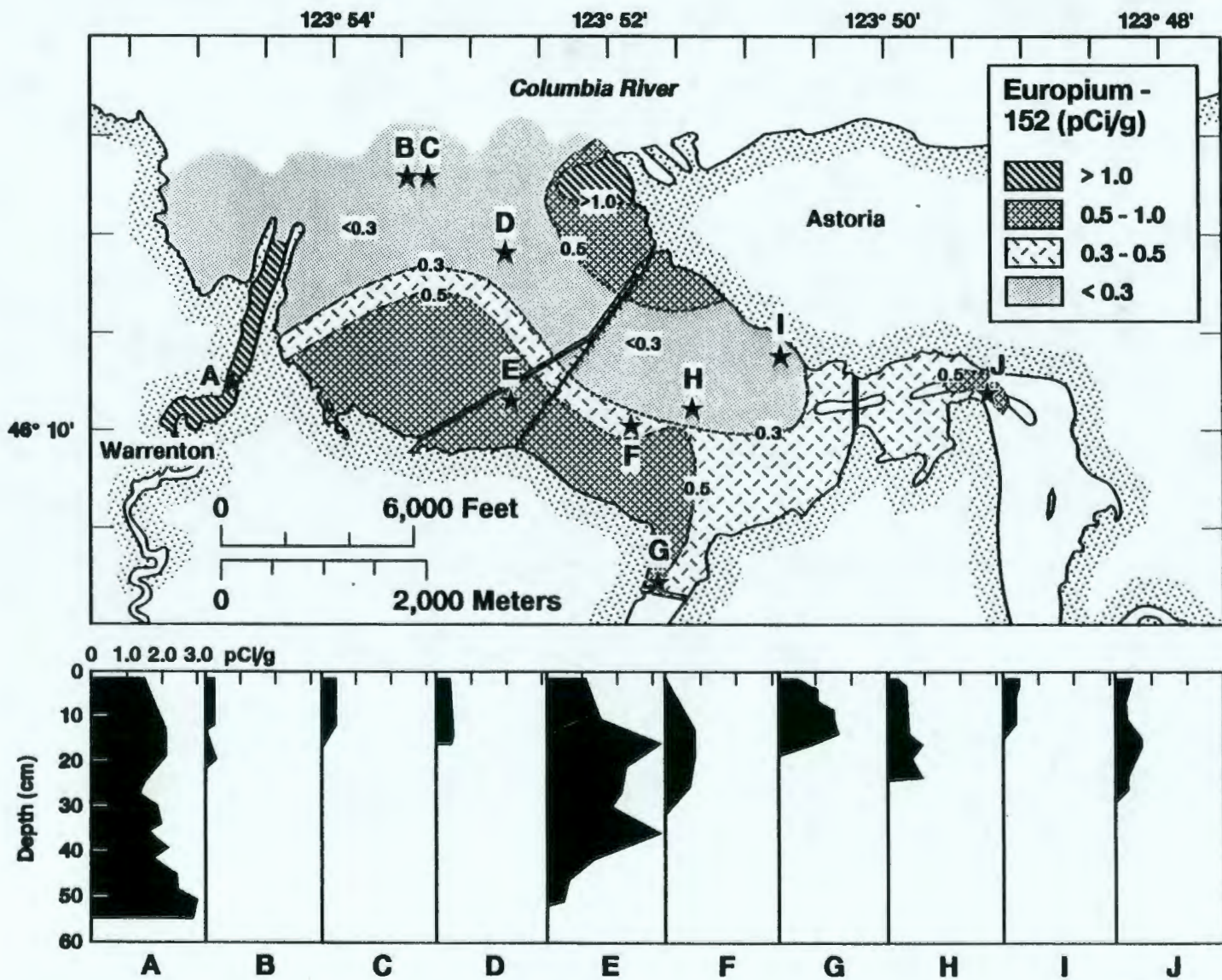
Figure 15. Correlation of ^{152}Eu in a McNary Dam Sediment Core with Effluent Discharge History.



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Shaded horizontal band indicates a period of reduced radionuclide discharge because of a change in operating conditions that occurred during 1962 (see Figure 2).

Figure 16. Areal and Vertical Distribution of ^{152}Eu ($T_{1/2} = 13.6$ year) in Young's Bay Sediment, June 1974.



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Surficial sediment concentration contours are based on 70 grab sample locations. The areal distribution pattern of ^{152}Eu approximates the sediment texture pattern shown in Figure 5a, illustrating the importance of texture in controlling the concentrations of particle-seeking contaminants in bed sediments. Similar areal distribution patterns were noted for the mining related heavy metals, zinc, cadmium, and lead. Vertical sediment texture profiles for cores A through J above are the same as shown at the top of Figure 5b.

origin, are only moderately attenuated. For example, the apparent sedimentation rate at core site E (protected area behind the solid fill causeway) is 4 cm/yr (Johnson and Cutshall 1975). Yet the ^{152}Eu concentration declined by only a factor of about 3 during the 4½-yr period. The other vertical patterns show very little indication of any dramatic decline in concentration with depth or time after shutdown. As already discussed, physical and biological mixing processes as well as anthropogenic disturbances occur in this bay. Thus, bottom sediment mixing as well as resuspension and redistribution could account for the apparent persistence of the radioactive tag long after termination of the source. In view of the high densities of benthic amphipods in this bay (see Figure 13) and their importance in the estuarine food chain, the slow attenuation rate or turnover in the near-surface sediments make this benthic habitat vulnerable to potential contamination (i.e., extended recovery times).

To summarize, contaminant concentrations in surficial sediment decline slowly following terminated or reduced input, especially in the estuary. This may be caused in part by mixing processes that tend to maintain contaminants in surface sediment. Finally, regardless of whether post-shutdown resuspension/supply occurred from upstream, from within the estuary itself, or both, the bed sediment and the suspended sediment results discussed (Johnson 1991) all indicate a relatively slow decline of sediment-bound contaminant concentrations following termination of the input source. This extends the natural purging time for sediment-bound contaminants, especially in the estuarine segment of this large aquatic ecosystem. This is particularly important because of the many species of juvenile fish, including chinook salmon smolts, that feed predominantly on filter-feeding and sediment-ingesting benthic invertebrates throughout the estuary.

SUMMARY AND DISCUSSION

Application of the results of this study to sediment-contaminant exposure pathway analysis and development of sediment quality criteria are discussed as follows.

Particle Size. Particle size was seen to control the initial sediment contaminant concentration as well as the transport, bed sediment distribution patterns, and potential incorporation into the food chain. The <4- μm size class exhibited the highest metal concentrations and has been shown to be a significant fraction of the total suspended load transported by the river and through the estuary to the coastal zone (Glenn 1973, Hubbell et al. 1971; Conomos and Gross 1972). The results of this study suggest that fine silt and clay (<4- μm size class) play an important role in the incorporation of surface-active contaminants in the food chain of many species of fish and invertebrates in the Columbia River and adjacent coastal zone.

Heavy metal, particle size, and radionuclide data suggest that contaminant concentrations in the Columbia River system can be approximated with a simple spherical particle-sediment concentration or specific surface model. Based on the above observations and the role of sediment in estuarine and coastal zone food chain transfer of contaminants implied by the radiotracer biodynamics, sediment quality surveys are recommended to use the <4- μm size class for assessment purposes. Where sufficient material for analysis in this size class is not available, extrapolation from the median

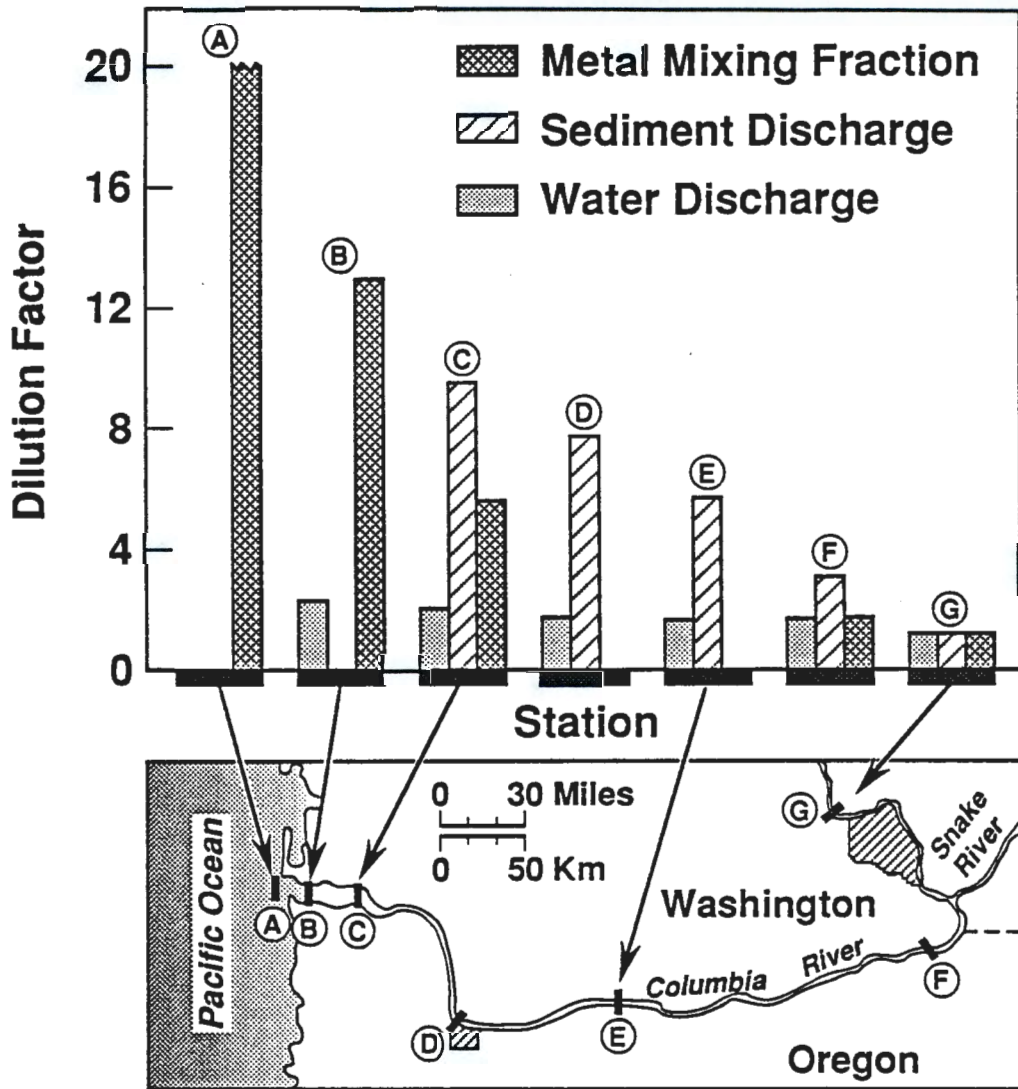
diameter of the unfractionated sample (Equation 3) is suggested. However, currently available or recently developed analytical methods, e.g., x-ray fluorescence and laser ablation inductively coupled plasma spectrometry for metals and microimmunoassay methods for organics make it feasible to analyze milligram amounts of solids. Thus, microtechniques combined with settling column separations (Guy 1969) should make analysis of the clay-size fraction possible in most cases. If not, centrifugation can be used to accumulate suspended sediment for size separation and subsequent analysis.

Sediment Dilution. Heavy metal mixing calculations as well as radiotracer results show the effect of downstream dilution by tributary sediments on bed sediment contaminant concentrations. These data are consistent with suspended sediment transport measurements made between Pasco, Washington, and Vancouver, Washington (Haushild et al. 1966). Previous sediment transport results are combined with results of this study in Figure 17. The Snake and Willamette Rivers contribute most of the suspended sediment to the lower Columbia River. However, this generalization may be altered during singular events such as the Vanport flood of 1948, Christmas flood of 1964, and ash fall from Mt. St. Helens via the Cowlitz River, when major sediment loads are added to the lower river and estuary. Nevertheless, the "average" conditions depicted in Figure 17 show the disproportionate effect of sediment dilution as compared to solute dilution. Sediment dilution results in significant attenuation of sediment-bound contaminants of upstream origin. For example, concentrations of metals and any other sediment-associated contaminants from the upper drainage basin will be diluted by 10 to 20 fold or more by the time river-borne sediment is deposited in or passes through the estuary. Additional sediment dilution near the mouth and on the nearby continental shelf occurs because of offshore mixing with suspended sediment contributed by adjacent coastal streams (Kulm et al. 1975, Cutshall et al. 1986). The downstream dilution or attenuation also means that slightly elevated contaminants detected in sediments of the lower river and estuary could be much higher in the upper drainage basin where dilution by tributary sediment is minimal. This may be an important consideration in the interpretation of results from lower river sediment quality surveys currently underway.

In the case of heavy metals, the effects of sediment dilution on concentrations in sediment can be mistakenly taken to show that pollutants are transported for only short distances. For example, the attenuation of the particle size dependence for zinc with distance downstream (see Figure 4) might suggest that zinc from the upper river does not reach the lower river in significant amounts. However, ^{65}Zn and ^{152}Eu , uniquely introduced at the Hanford Site, found in the lower river and nearly 200 km seaward beyond the river mouth in bed sediments (Gross and Nelson 1966, Cutshall et al. 1972, Cutshall et al. 1986) demonstrate the mobility and long-range dispersal of contaminants in fine silt and clay-size sediment from the upper Columbia River drainage basin.

Washout Dynamics. Post-shutdown time-series radiotracer results reported in this study for the lower river and estuary indicated decay-corrected half-times of decline on the order of 400 days for biota and suspended sediment and >600 days for surficial bed sediment. The longer rate of decline in surficial bed sediment is attributed to benthic mixing and resuspension/redistribution in protected areas where fine sediment accumulates. The significance of these observations is illustrated in

Figure 17. Downstream Dilution of Columbia River Sediment Between Priest Rapids Dam and the Mouth.



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All dilution factors are normalized to the Priest Rapids Dam site. Main stem Columbia River water dilution (bars designated as water discharge in the legend) by mixing with tributary water is shown for comparison with sediment dilution. Dilution factors based on metal mixing fractions were calculated using equation 2 and are listed in Table 5. Dilution factors based on sediment discharge were calculated from suspended sediment transport data reported in Haushild et al. (1966).

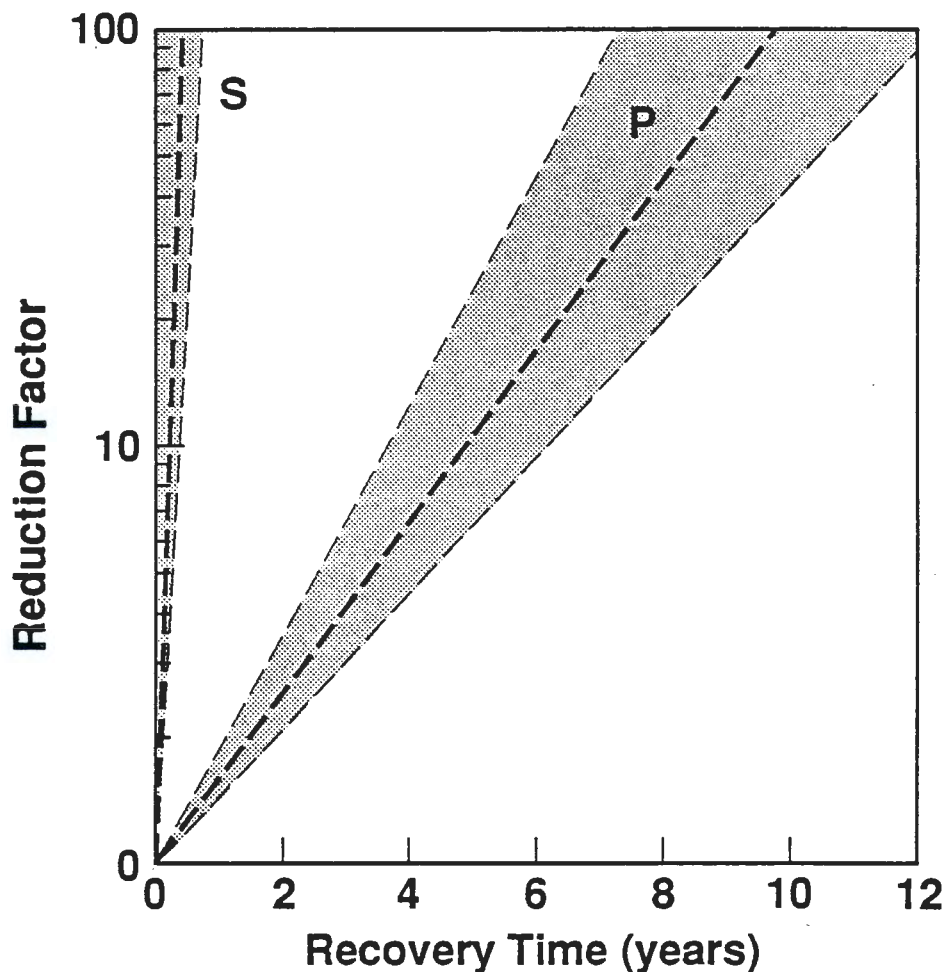
Figure 18 where the above half-times are used to estimate recovery time (i.e., return to background conditions following termination of a chronic input). This plot suggests, for example, that a 10-fold reduction in the concentration of a contaminant in surface sediments could require over 6 yr after termination of the source. The estimated recovery times could be higher or lower depending on environmental conditions during any given year as compared to conditions that existed in 1971 and 1972. Nevertheless, the general trend and implications are clearly illustrated by this application of the post-shutdown radiotracer data. When a large river-reservoir system has been subjected to chronic loading of "particle-seeking" contaminants, several years may be required for restoration by "natural" processes.

Water and Sediment Quality Implications. Results of this study are significant to multiple-use water resource planning in the Columbia River basin. For example, flow augmentation to speed the downstream migration of young salmon accelerates resuspension of fine sediment and associated contaminants. If significantly elevated surface sediment contaminant concentrations in the upper reservoirs still exist today, especially below the confluence of the Snake River with the Columbia River, the contaminants could be mobilized and become incorporated in estuarine food chains. Results of sediment quality surveys currently underway need to be factored into flow regulation planning.

Disturbance of previously deposited fine sediment containing elevated contaminant concentrations is another potential impact. Streamside construction activities or dredging dislodge fine sediment. Periphyton (gelatinous algal and bacterial coatings) trap fine sediment either on exposed surfaces or within the stream bank gravels. This phenomenon occurs in the Hanford Reach of the Columbia River where gravel and boulders accumulate extensive sediment-periphyton "mats" a few millimeters in thickness (Cushing et al. 1981). During diurnal and seasonally high water, particle transport into the stream bank may also occur. Colloids and clay-size sediment may become trapped by "filtering" (Domenico and Schwartz 1990) or by incorporation in periphyton or bacterial surface coatings on sands and gravels in the stream bank storage zone. Localized disturbances caused by dredging or contaminated soil/sediment excavation at or below the water table near the shoreline would release ultra-fine sediment ($<4 \mu\text{m}$) containing potentially high concentrations of adsorbed contaminants. The Hanford Reach of the Columbia River may be especially vulnerable to such effects. For example, this is the last freeflowing stretch of the river in which chinook salmon (*Oncorhynchus tshawytscha*) still spawn. Disturbances caused by dredging or other activities near the stream bank could expose young salmon fry to sediment-bound contaminants either by direct contact with sediment particles or by incorporation of sediment-bound contaminants in their food sources. Recent shoreline sediment reconnaissance sampling results in the Hanford Reach indicated above-background concentrations of heavy metals that are consistent with historical inputs from upstream refinery operations (Peterson and Johnson 1992).

Ecological Risk Assessment Considerations. Risk assessments begin with defining the perceived hazard, i.e. characterizing the source, receiving environment and endpoints or the ecological values deemed at risk and worthy of protection (Suter and Loar 1991). Then exposure and effects assessments are performed. The exposure phase involves estimating the distribution of contaminant and concentrations to which the endpoint species are exposed.

Figure 18. Recovery Times for Particle-Bound Contaminants (P) and Water (S) Following a Chronic Input to the Lower Columbia River and Estuary.



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The line shown for particulates (P) is based on decay-corrected rates of decline for ⁶⁵Zn in water, sediment, and biota. The range shown reflects the differences in observed half-times of decline, i.e., 400 days for water/suspended sediment and estuarine biota, and 600 days for surficial sediment. The line for solutes (S) is based on a mean flow rate for the lower river system of 200,000 cfs and the river-reservoir volume under mean flow conditions, i.e., solving the relationship: $C/Co = \exp[-(Q/V)*t]$ for time "t" for various reduction factors R, where $R = 1/[C/Co]$; Q = mean river flow rate, and V = river-reservoir volume from Lake Wallula behind McNary Dam down to but not including the estuary. Particle-seeking contaminants introduced upstream of Lake Roosevelt would probably show similar recovery times in the upper river system. However, the combined effect of storage/resuspension in the river above Priest Rapids Dam as well as in lower river system, would be longer recovery times (in response to reduced upriver input) for the lower river and estuary than shown above.

Effects assessments rely on lethal and sublethal tests with site-specific biota and or extrapolation from other ecologically equivalent test species from other locations.

While the above approach may seem straightforward, application to a system the size of the Columbia River, estuary, and adjacent coastal zone is a formidable task. A species/habitat of interest approach, however, can simplify the analysis by focusing on a few critical or indicator species and habitats. Indeed, it may be the only practical solution in such a large and dynamic system. The philosophy underlying the "species of interest" approach is that protection of the habitat of a specific species will in turn protect the ecosystem at large (Warren 1971). Two such sites for assessing the health of the Columbia River system are considered here: (1) chinook salmon spawning beds in the Hanford Reach and (2) benthic amphipod habitat in Young's Bay and vicinity near Astoria, Oregon. Because salmon that spawn in the Hanford Reach pass through the reservoirs, lower river, and estuary as outmigrating smolts and as returning adults, they are large-scale, integrating biomonitors of environmental conditions in this large, interconnected fluvial-marine ecosystem. Additionally, some of the outmigrating chinook salmon smolts use the brackish water benthic habitat in Young's Bay to make the freshwater-to-marine transition (Higley et al. 1976). They may be particularly sensitive to contaminants or other stresses during this physiologic adaptation phase (Higley et al. 1976).

In view of the varied conditions to which they are exposed and with allowance for harvesting and natural changes in oceanic conditions, chinook salmon returns or redd counts in the Hanford Reach may be useful as long-term indicators of the overall health of the river.

Exposure Estimates. Estimating the contaminant concentration to which the target population is exposed is often based on mathematical transport models. The many factors controlling solute-particle or mass transfer, however, complicate this approach. An alternative is to use the tracers evaluated in this study to estimate the mass transfer of contaminant from water to sediment.

Mass Transfer Coefficient. Normalizing the contaminant concentration in downstream sediment by the mass input rate or loading (MT/yr) becomes, in effect, a "time-integrated" mass transfer coefficient. For example, because of the slow sedimentation rate at depositional sites, a surficial sediment sample of a few centimeters depth represents an accumulation period on the order of a year or more. The sample thus represents the average conditions for this period of time. The primary assumptions are (1) loading remained relatively constant over the period of interest and (2) post depositional changes in sorptive status of the contaminant are minimal. An example set of such computations is shown in Table 7.

The transfer coefficients listed in Table 7 provide an estimate of the incremental increase in contaminant concentration of the $<4\text{-}\mu\text{m}$ size class resulting from steady-state input of 1 MT/yr. The systematic decrease in K_R with downstream distance is attributed to the sediment dilution effect previously discussed. The larger apparent sediment-transfer for lead than zinc is attributed to differences in their affinity for particulates. For example, the apparent distribution coefficients (based on the Northport

Table 7. Water-to-Sediment Heavy Metal Transfer Coefficients. $K_R = C/L$, where $C = \mu\text{g/g}$ and $L = \text{MT/year}$. Loading Assignments: 100; 4,000; and 45 MT/yr for Lead, Zinc, and Cadmium, Respectively. Background-Corrected Metal Concentrations Are for Either the $<4\text{-}\mu\text{m}$ Size Fraction from Settling Column Separations or Were Adjusted to a Median Diameter of $4 \mu\text{m}$ (See Equation 3).

Depositional Regime	Sample Site Location	Pb		Zn		Cd	
		C	K_R	C	K_R	C	K_R
Upper River	Lake Roosevelt	^a 550 ±30%	6	N/A	N/A	N/A	N/A
Mid River	Priest Rapids	^b 400	4	2500	0.6	37	0.8
	Lake Wallula	^b 244	2	1570	0.4	30	0.7
		^c 100 ±27%	1	850 ±13%	0.2	11 ±24%	0.3
Coastal Zone	Young's Bay	^b 36	0.4	590	0.2	*	*
	Astoria Canyon	^b 13	0.1	80	0.02	*	*

^a Mean of six sample locations ($570 \pm 170 \mu\text{g/g}$; Whetten et al. 1969) in lower Lake Roosevelt minus $20 \mu\text{g/g}$ to allow for natural background (sample from Lower Arrow Lake on Columbia main stem above Trail, British Columbia; Johnson et al. 1989). Whetten et al. reported bulk sediment texture was $<4 \mu\text{m}$ based on particle size analysis of splits of the same Lake Roosevelt samples analyzed for lead and other metals (Table 3 and Figure 10 in Whetten et al. 1969).

^b Based on adjusted concentrations of Table 4 values. Adjusted to a median particle diameter of $4 \mu\text{m}$ using equation 3 after subtraction of a background of $20 \mu\text{g/g}$. Median particle diameters were $16 \mu\text{m}$, $16 \mu\text{m}$, and $4 \mu\text{m}$ for Lake Wallula, Young's Bay, and Astoria Canyon samples, respectively. Adjusted cadmium concentration was calculated in the same manner but with a background subtraction of $0.6 \mu\text{g/g}$ (sample from Lower Arrow Lake, British Columbia, Johnson et al. 1989).

Zinc concentrations based on direct analysis of the $<4\text{-}\mu\text{m}$ size fraction with a background subtraction of $200 \mu\text{g/g}$ (average of the $<4\text{-}\mu\text{m}$ size fraction of Snake River and Willamette River samples).

^c Based on analysis of the $<4 \mu\text{m}$ size fraction of archived, air-dried core tops (0-2 cm) collected 1-2 mi upstream from McNary Dam and near the center of the reservoir. Values shown are the mean \pm %CV (one-sigma, $N = 12$). The settling column pipet method of Guy (1969) was used for the size separations. The separates were dried in 50 mL polyethylene bottles and the residue digested in 1 mL each of nitric and hydrofluoric acid for 12 hr at 60°C . Digests were diluted to 25 mL in dilute nitric and analyzed by inductively coupled plasma emission spectroscopy in August 1993. Standard Reference Materials (SRM 2704 and SRM 1646; river and estuary sediments) were run with the sample set; recoveries ranged from 84% to 100% (Mean = 93%). Background levels were subtracted as described in note ^b.

filtered water data) are 1×10^5 and 3×10^5 for zinc and lead, respectively. Distribution coefficients of this magnitude are characteristic of a wide range of contaminants (e.g., polychlorinated biphenyls, insecticides, and plutonium; Olsen et al. 1982). It is thus proposed, as a first approximation, that the transfer values listed in Table 7 may be used to estimate sediment concentrations caused by upstream loading of a wide range of contaminants. Refinements can be made by conducting natural water uptake experiments with the contaminant of interest using zinc and or lead as controls, i.e., normalizing the laboratory distribution coefficient for the contaminant of

interest to zinc or lead. Mathematically, this can be expressed as follows:

$$K_R^x = [K_d^x / K_d^{Zn}] * K_R^{Zn} \quad 6)$$

where

- x = Contaminant of interest
- K_d = Laboratory distribution coefficient
- K_R = Transfer coefficient for the appropriate river segment (Table 7).

An independent check on the K_R values based on zinc (Table 7) can be made using ^{65}Zn data. Because this radiotracer was uniquely of Hanford Site origin, no background correction is required. This may be an important consideration because not all downstream heavy metal sources may have been taken into account. If additional sources existed, the actual dilution or attenuation based on metal content may be underestimated. However, because there is only one source of the ^{65}Zn , it is not subject to this potential uncertainty.

To compare the radiotracer and stable zinc transfer coefficients, mass units must be eliminated. For this purpose, we redefine K_R as follows:

$$\begin{aligned} *K_R &= C/L = [(\mu\text{g/g})]/[(\text{MT/yr}) * (10^{12} \mu\text{g/MT})] \quad \text{for Zn} \\ &= C/L = [(\text{pCi/g})]/[(\text{Ci/yr}) * (10^{12} \text{pCi/Ci})] \quad \text{for Zn-65} \end{aligned}$$

The mass (MT) and radioactivity (Ci) units cancel for which the resulting units for $*K_R$ are yr/g in both cases. Multiplying the coefficients in Table 8 by the annual average radionuclide or metal input rates in pCi/yr or $\mu\text{g/yr}$ respectively yields estimated sediment concentrations.

Results of the comparison summarized in Table 8 indicate good agreement for the McNary Dam site while the estuary comparison is less definitive. In the latter case, the mid estuary ^{65}Zn data was based on regression of sediment concentration versus mean particle diameter from several transects across the estuary ranging from the lower to upper estuary (Hubbel and Glenn 1973). Thus, the value obtained is interpreted to be an estimate of average conditions or mid-estuary. Even with these uncertainties, the general trend is consistent with results in Table 7 (i.e., showing the effects of attenuation with distance downstream and in the estuary). The general agreement between the two independent estimates provides additional support for the mass transfer coefficients for estimating sediment exposure levels caused by chronic inputs. Table 8 also suggests that if there is additional zinc input (other than natural background) from downstream sources, it must have a negligible effect. It should also be noted that ^{60}Co yields a similar transfer coefficient based on sediment samples from the lower portion of Lake Wallula (see footnotes to Table 8 for data sources and related details):

$$*K_R = [(14 \text{ pCi/g}) * (32/4)] / [(310 \text{ Ci/yr} * 10^{12} \text{ pCi/Ci})] = 3.6 \times 10^{-13} \text{ yr/g}$$

This is reasonable because ^{60}Co and ^{65}Zn exhibit similar ionic and sorptive characteristics in effluent and Columbia River water (Perkins et al. 1966). Furthermore, because heavy metal and radionuclide points of entry to the river are separated by over 600 km, the overall concordance noted above is all the more remarkable. This suggests there must be a high degree of uniformity in the processes controlling the transport and fate of fine-sediment associated

Table 8. Comparison of Zinc and ⁶⁵Zn Mass Transfer Coefficients for Selected Depositional Regimes.

Regime	Location	*K _p , yr/g	
		Zinc	⁶⁵ Zn
Mid river	McNary Dam (lower pool)	4 x 10 ⁻¹³	^a 2.5 x 10 ⁻¹³
Coastal Zone	Young's Bay	2 x 10 ⁻¹³	N/A
	Mid-estuary	N/A	^b 3 x 10 ⁻¹⁴
	Astoria Canyon	2 x 10 ⁻¹⁴	N/A

^aBased on a mean ⁶⁵Zn concentration (470 pCi/g) for 26 locations in lower reservoir collected in 1963-1964 (from Table 4 in Haushild et al. 1975); concentration adjusted to a particle size of 4 μm with equation 3 and an average mean particle diameter for the lower reservoir of 32 μm (from Whetten et al. 1969). An annual average ⁶⁵Zn release rate to the river of 15,000 Ci for 1964 (Hall 1968) was used for the loading estimate.

^bBased on (1) ⁶⁵Zn concentration for 4 μm median particle diameter from regression of grain size and ⁶⁵Zn content for several mid-estuary samples collected in 1965 (Hubbel and Glenn 1973) and (2) annual average loading of 12,500 Ci/yr (Hall 1968).

contaminants in the Columbia River. Reference monitoring and assessment sites at selected downstream locations may thus be representative of long stretches of river. Consistent with this inference is the homogeneous character of fine-fraction sediment properties and radionuclide-particle size relationships in the Columbia River (Haushild et al. 1975).

Effects Screening. The following illustration is limited to chronic inputs of heavy metals from mining and refinery operations, the major known source of contaminant loading that occurred during the early to mid 1970's. Evaluation of this past occurrence should illustrate application of the contaminant-sediment exposure method and will hopefully serve as a paradigm for similar considerations. The Hanford Reach is the impact zone of interest.

Metals to be considered for this illustration include zinc, lead, cadmium, and mercury at approximate annual average input rates (rounded to one significant figure) of 4,000, 100, 40; and 2 MT/yr, respectively. Corresponding fine fraction (<4 μm) sediment concentration estimates for the Hanford Reach based on transfer coefficients are shown in Table 9.

Based on downstream trends in transfer coefficients shown in Tables 7 and 8, and the sediment dilution considerations summarized in Figure 17, the Young's Bay fine sediment (<4 μm) metal concentrations are estimated to be 1/10th of the concentrations shown in Table 9. Concentrations observed in Young's Bay sediment in 1974, at those locations where fine sediment accumulates, are consistent with this prediction (see Figure 5, core profiles A and E).

No freshwater sediment toxicity data for the Columbia River exists. However, Ontario Provincial freshwater sediment quality criteria have been proposed for selected constituents. Metals of interest for this study are shown in Table 10 along with world average shale values and the EPA chronic

Table 9. Illustration of Sediment Exposure Levels Calculation for the Hanford Reach (K_p for Priest Rapids Dam, Table 7).

Metal	¹ MT/yr	K_p	² μ /g
Zinc	4,000	0.6	2400
Lead	100	4	400
Cadmium	40	0.8	32
Mercury	2	*(4)	8

* K_p for Lead used based on assumed similarity in particle affinity.

¹ PPe-1978 estimated Canadian input to upper Columbia River.

² μ g/g = MT/yr (column 1) x K_p (column 2).

Table 10. Reference Freshwater Sediment and Water Quality Criteria.

Metal	¹ Sediment Quality (μ g/g)	² World Avg. Shale (μ g/g)	³ Water Quality (μ g/L)
Zinc	120 ⁴ (100)	95	82
Cadmium	0.6	0.3	0.9
Lead	31 (50)	20	2.2
Mercury	0.2	0.4	0.012

¹ Ontario Provincial freshwater sediment quality guidelines (lowest effect levels); acid extractable.

² World average shale values from Bowen (1966); total digestion.

³ From Johnson et al. (1989); freshwater water quality chronic criteria were calculated for a total hardness (CaCO_3) of 74 mg/L, EPA, Quality Criteria for Water, 1986 and 1987 update #2, EPA-440/5-86-001. Sediment quality are for lowest effect levels, Ontario Provincial freshwater sediment guidelines.

⁴ Wisconsin State Sediment Quality Standards: Elder 1994, *Water Resources Bull.* V. 30 (2):254.

freshwater quality criteria. The sediment criteria or guidelines shown are the lowest levels at which effects were observed. The sediment quality guidelines may be somewhat conservative because they are very close to natural background levels. Note, the world average shale values are total, i.e., include a matrix bound fraction that may not be biologically available. Nevertheless, historical heavy metal concentrations in the <4- μ m size fraction (e.g., Table 9 and Figure 4) were far in excess of either of these sediment reference levels.

Estimates of average release rates that would theoretically just meet the Ontario Provincial sediment quality guidelines can be obtained by dividing the guideline values listed in Table 10 by the appropriate mass transfer coefficients (K_p , Table 9). This yields 200, 0.75, 7.8, and 0.05 MT/yr for zinc, cadmium, lead, and mercury, respectively. The corresponding release rates based on the EPA chronic water quality criteria (column 3, Table 10), assuming complete mixing and an average flow rate of 100,000 cfs, are 6,900; 76; 185; and 1 MT/yr for zinc, cadmium, lead, and mercury, respectively. Thus, a sediment-benthic biota exposure pathway, as applied under the conditions described above, is 20 to 100 times more restrictive for

Thus, a sediment-benthic biota exposure pathway, as applied under the conditions described above, is 20 to 100 times more restrictive for controlling releases of these heavy metals than the EPA chronic water quality criteria. This further emphasizes the significance of contaminant-sediment exposure pathways in the Columbia River system.

Fortunately, by 1986, heavy metal releases from the Cominco, Ltd. refinery declined to about 15% of the 1977 levels used in the Table 9 estimates. During this same period, dramatic increases in chinook salmon spawning occurred in the Hanford Reach. This reach supports ~60% of the chinook salmon run on the Columbia River (Dauble and Watson 1990); this is also a significant fraction of the total U.S. chinook salmon fishery. What role, if any, contaminants from upstream sources had on spawning success in the Hanford Reach is unknown. There are many variables that influence salmon returns. For example, during the last 3 to 4 yr, the chinook salmon returns to the Hanford Reach have declined. However, the pattern of decline is similar to the one for the Sacramento River, California. This suggests unfavorable oceanic conditions existed during the latter period. Be that as it may, favorable physical and water quality conditions must have prevailed in the Hanford Reach during the dramatic increase in returns during the 1980's. These favorable conditions may be useful as benchmarks for protecting this high-value, natural resource. The significantly reduced heavy metal and related refinery waste loading from upstream sources, and associated sediment-bound contaminant concentrations, may be one attribute in a profile of favorable physical and chemical conditions.

Modes of exposure and toxic response of benthic biota to particulate-phase contaminants in the Hanford Reach are unknown. However, as indicated previously, the periphyton coatings that occur on surfaces in this dynamic, fluvial environment are known to accumulate sediment fines. It is possible that eggs and newly hatched fry and fingerlings could be exposed to fine sediment by direct contact and/or via ingestion of prey species that graze on the periphyton-sediment particle aggregates. These exposure modes could be evaluated with partial life-cycle testing in flow-through microcosms or simulated shoreline environments using water and natural substrate from the river, and varying amounts of contaminated sediments (e.g., <4- μm size sediment from lower Lake Roosevelt). Likewise, benthic amphipods from the lower river and estuary could be used for site-specific toxicity testing. Hatching success, behavioral, physiologic, and or biochemical indicators could be used as indicators of response to varying contaminant-sediment concentrations. In view of the potential significance of a sediment exposure pathway, as indicated in this paper, and the value of the resource, appropriate ecotoxicity testing and development of site-specific sediment quality criteria are warranted. Additionally, analysis of metals in <4- μm size separates of bed sediments should be included in ongoing monitoring activities upstream of the salmon spawning areas to follow the long-term trends in mining related metal inputs.

CONCLUSIONS AND RECOMMENDATIONS

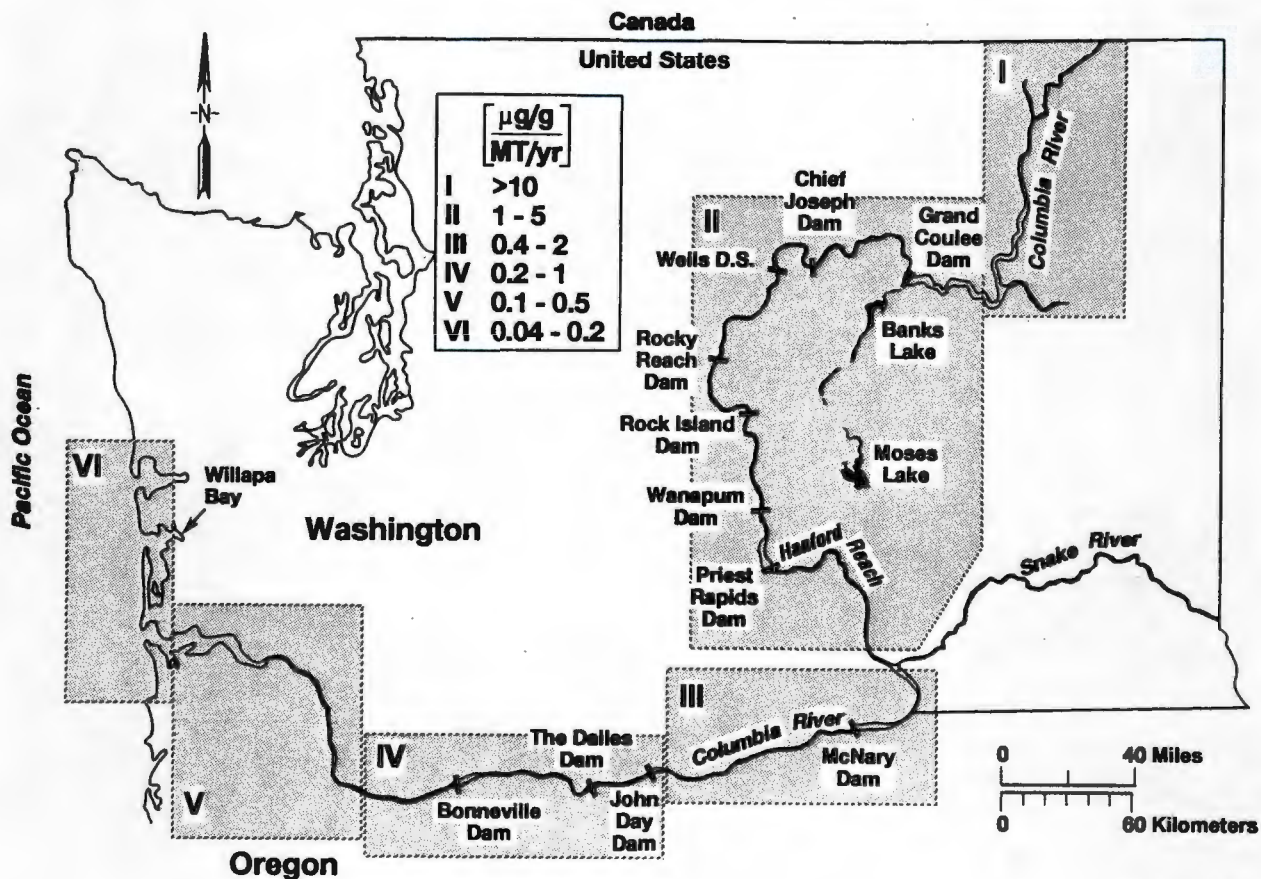
Suspended sediment and hydrologic conditions play a significant role in the long-range dispersal of particle-seeking contaminants discharged to the upper Columbia River. The very low suspended load and relative abundance of the clay-size (<4 μm) fraction in suspension, coupled with its high sorption

potential, account for the relatively high particulate-contaminant concentrations in this size fraction. While the total mass of clay-size sediment in the system is small, fine silt and clay do accumulate slowly behind certain reservoirs, in sloughs and side channels of the river, and in protected areas of the estuary. Some of these fine sediment depositional sites are also critical habitats for outmigrating salmon smolts as well as other fish and aquatic birds. Fine silt and clay also become attached to periphyton coatings on sand, gravel, and boulders. Incidental ingestion of sediment, as suggested by this study, is an important exposure pathway in the shallow benthic habitats noted previously. The most significant potential human exposure pathway for particle-bound contaminants involves consumption of the filter-feeding shellfish, such as marine mussels and oysters, in the adjacent coastal zone (Essig et al. 1973).

It is suggested that the $<4\text{-}\mu\text{m}$ size fraction be used in risk assessments involving surface-active contaminants and that analysis of this size fraction be included in sediment quality surveys. Impact assessments and sediment quality surveys should view the river reservoir complex and coastal zone as one interconnected system. Six assessment zones (Figure 19) ranging from Lake Roosevelt to the coast are proposed for this purpose. The six zones are based on sediment exposure potential (mass transfer coefficients), presence of critical habitats, and sediment dilution by tributaries. Potential impacts in each zone from chronic inputs of reactive contaminants in the upper drainage basin can be assessed by using the indicated mass transfer coefficient assignments for each zone of interest and the upstream contaminant loading. Effects screening can be done based on the estimated sediment-contaminant concentrations and appropriate sediment quality criteria. Based on effects screening from this study, a total loading of particle-seeking contaminants from all sources of less than 10 MT/yr of lead, or its ecotoxic equivalent, is suggested as a long-term goal for releases to either Zone I or II, Figure 19 (mixtures of contaminants should be allocated in proportion to loading/limit ratios, the sum of which should be ≤ 1).

Finally, lower river sediment and water quality surveys (Koberstein 1992) and impact assessments of waste disposal and sediment disturbances in the upper drainage basin should be integrated to (1) optimize monitoring and environmental protection programs, (2) ensure that appropriate data are obtained for evaluating the relative risks of individual projects or activities, and (3) address cumulative impacts versus distance along the main stem of the Columbia River. The primary water quality objective should be the protection of the ecological health of the singular most important water resource in the Pacific Northwest. It is hoped that the present work will provide some perspectives for development of an integrated, basin-wide approach to water resource protection and management in the Columbia River drainage basin.

Figure 19. Proposed Assessment Zones (Based on Sediment Exposure Potential, Habitat, and Resource Protection Goals).



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The mass transfer coefficients, in units of $\mu\text{g/g/MT/yr}$ ($<4 \mu\text{m}$ size fraction) for each zone are based on a combination of judgment and the range of calculated values shown in Tables 7 and 8, together with sediment dilution factors (Figure 17). The contaminant loading point of entry for application of the numerical values shown in the legend should be upstream of the confluence with the Snake River. Zone I is the highest exposure potential because of minimal sediment dilution in this reach and the existence of both a human exposure pathway (sport fish consumption) as well as potential sediment toxicity to biota. Zone II, which includes the irrigation impoundments supplied by water from the Grand Coulee pumping station, is nearly equivalent to Zone I. Some sediment dilution apparently occurs, however, because of shoreline erosion that adds clay minerals to the lower reservoir system. The bed sediment for several miles upstream from the dam is all in the $<4 \mu\text{m}$ size class, suggesting that particulates in water diverted into the irrigation system are of a similar particle size. Zone II extends from Grand Coulee Dam down to just above the confluence with the Snake River because of minimal sediment dilution in this reach. Critical habitats and exposure pathways are (1) the salmon spawning beds below Priest Rapids Dam and (2) consumption of sport fish from the reservoirs and irrigation impoundments. Zones III, IV, and V have progressively lower sediment exposure potential, and there are no free-flowing sections or salmon spawning beds. Zone V includes the benthic amphipod habitat in fine sediment in Young's Bay. The principal pathway of concern in Zone VI is human consumption of marine mussels and oysters (Willapa Bay). Loading limitations based on sediment exposure levels for Zones I and/or II, and the Ontario Provincial sediment quality (lowest effect) levels for zinc, cadmium, mercury, and lead (or equivalent toxicity or other contaminants) would probably provide adequate protection for aquatic biota in the other zones. Human exposure potential, however, could be nearly as great in Zone VI as in Lake Roosevelt because of the coastal zone/shellfish pathway.

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Tony Knepp	H6-06	x			
Karl Fecht	H6-06	x			
Steve Reidel	H6-06	x			
Kent Reynolds	H6-06	x			
Vern Johnson(30)	H6-06	x			
Kevin Lindsey(15)	H6-06	x			
John Auten	H6-06	x			
Doug Hildebrand	A5-55	x			
Mat Johansen	A5-19	x			
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Central Files (2)	L8-04	x			
EPIC (2)	H6-08	x			