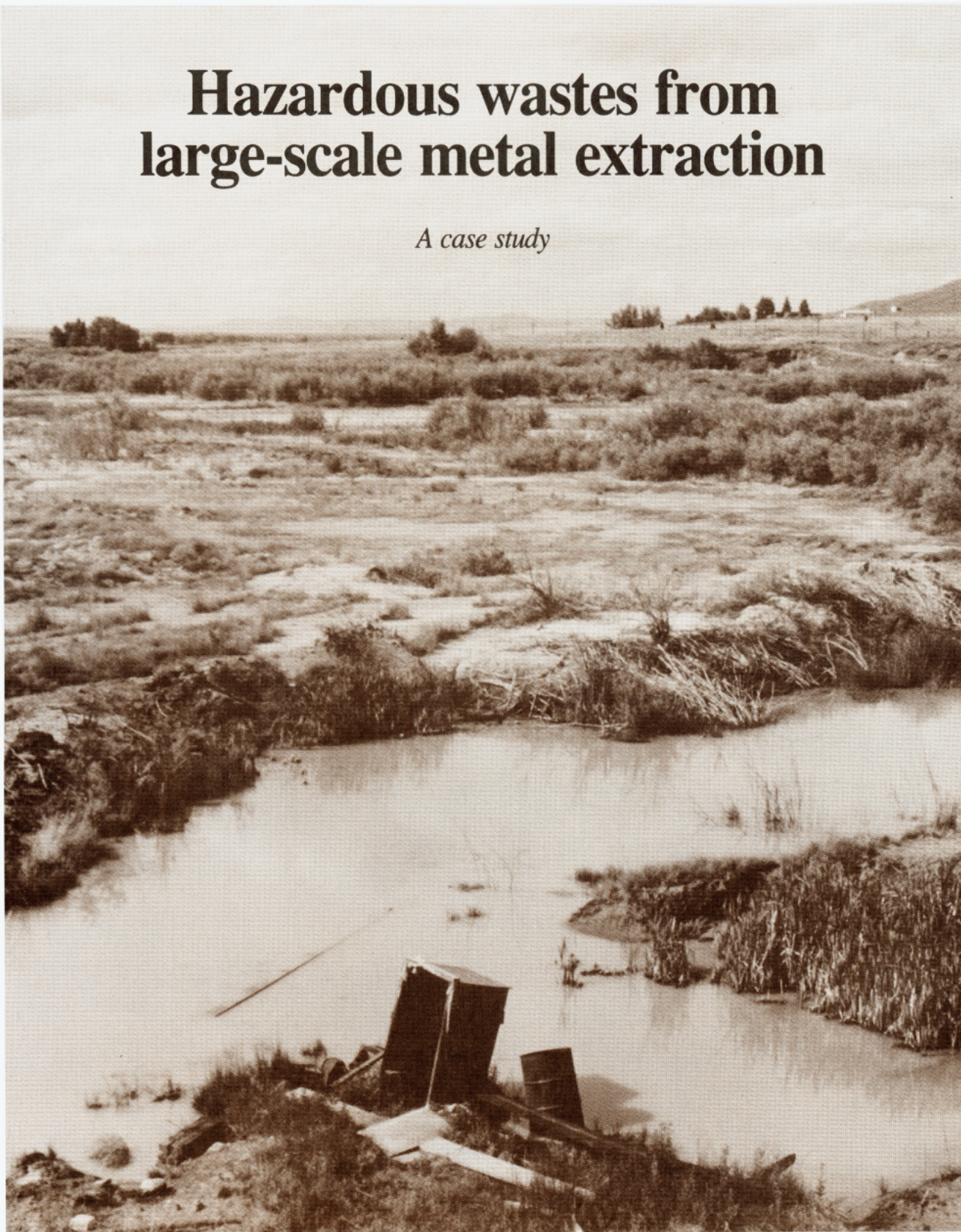


# Hazardous wastes from large-scale metal extraction

*A case study*



*Seepage of metal-rich water from tailings ponds in the Clark Fork River Basin, a Superfund Complex*



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Large-scale extraction of metals presents hazardous waste management problems worldwide. Projections show that by the year 2000 mining will have directly disturbed about 240,000 km<sup>2</sup> of the Earth's surface (1), an area about the size of Oregon; copper mining alone is projected to totally disrupt about 1600 km<sup>2</sup> of land. Milling and smelting presently release an estimated 7 to 70 × 10<sup>3</sup> metric tons (MT) of metals to the aquatic environment (1, 2). Metal production has increased over the last century although the grade of ore has declined (3), and the richest metal reserves are now in less developed countries (4) where legislative protection from environmental damage often is limited.

Coping with hazardous waste problems is an immense challenge (5). In the United States, EPA has identified over 31,000 hazardous waste sites (6); about 1200 of these need immediate response, and only a small proportion have been cleaned up (6, 7). The largest complex of "Superfund sites" to be remediated is in western Montana, in the Clark Fork River Basin. Produced by more than 125 years of copper and silver mining and smelting activities, the Clark Fork Complex encompasses contaminated lands covering an area 1/5 the size of Rhode Island. Many sites throughout the world are similarly disturbed by metal extraction (8, 9), and understanding past abuses helps explain the costs of such actions.

Although the managerial and political challenges facing remediation of contaminated sites have been reviewed (10, 11), the technical difficulties posed by large-scale metal extraction have not been adequately considered. To illustrate the number of problems that can develop, we discuss the processes that must be further understood for effective remediation. We consider whether contamination of soils, air, and water threatens ecological and human health, emphasizing the Clark Fork Complex because of the detailed work conducted there and the magnitude of the associated problems. We conclude by pointing out the difficulties in remediating large-scale hazardous waste problems and the importance and cost effectiveness of careful waste management and waste reduction during future production.

### Origin, types of waste

Hazardous waste problems associated with mineral extraction are determined

by the ore and the processes employed to extract metals. Many ore bodies consist of high-grade metal sulfide veins enclosed in lower grade altered rock (12-15). Sulfur content commonly exceeds 30%, with pyrite ( $\text{FeS}_2$ ) being the most common sulfide in metal ores and a primary component (0.5-4%) of the wall rock that encloses ores (12). Many of the important processes that affect the fate of contaminants like antimony, arsenic, cadmium, copper, lead, and zinc involve the often-complicated oxidation-reduction reactions of sulfides and oxygen (16).

In this paper we characterize waste from such ores as primary, secondary, or tertiary contamination. The variety of wastes produced during mining, milling, and smelting (Figure 1) and deposited near their origin are sources of primary contaminants. As these contaminants are transported away from the site by rivers or through the atmosphere they generate secondary contamination in soils, ground water, rivers, and air. Deposits of these by-products can be distributed over vast areas (2, 9) and, if remobilized, can result in tertiary contamination (17, 18). Interaction among these categories of wastes results in complex problems in geochemistry, hydrology, ecology, and epidemiology.

### Primary, secondary contamination

Primary contamination, commonly in an ill-defined patchwork of waste rock, mill tailings, furnace slag, and flue dust,

is spread over the countryside near centers of mining and smelting (2, 9, 19). Different types of waste have vastly different contaminant concentrations (Table 1). Waste rock is probably the least contaminated material. In the Clark Fork Complex, the 300 million m<sup>3</sup> of rock removed from the principal open pit mine (the Berkeley Pit) and tens of millions of cubic meters from underground workings (20) cover approximately 10 km<sup>2</sup> of land.

As ore is separated by milling and flotation, commonly 90% of it is discarded as tailings (21, 22). When the remaining concentrate is refined by smelting, flue dust and slag are produced (2). Such residues can contain thousands of times the natural levels of arsenic, cadmium, copper, lead, and zinc (2, 9, Table 1). Identifying all the deposits of these wastes is often difficult because of the lack of historic records and the mixtures of waste types (23).

Some of the largest deposits occur in tailings ponds. In the Clark Fork Complex, ponds cover at least 35 km<sup>2</sup> and hold more than 200 million m<sup>3</sup> of tailings (19). Based on average concentrations of metals in the tailings (Table 1), approximately 9,000 MT arsenic, 200 MT cadmium, 90,000 MT copper, 20,000 MT lead, 200 MT silver, and 50,000 MT zinc could be present in the ponds. Similarly contaminated tailings ponds cover about 21 km<sup>2</sup> in Bingham Canyon, Utah (24), and 20 km<sup>2</sup> in Copper Cliff, Ontario (2). Such areas

FIGURE 1  
Types of contamination resulting from large-scale metal extraction

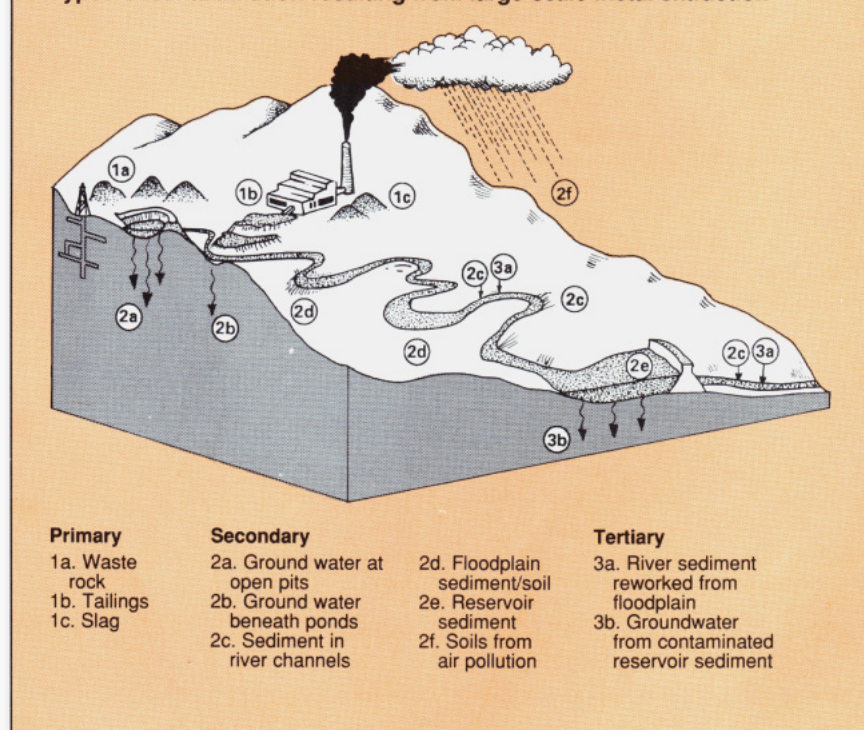




TABLE 1  
Metal concentrations (ppm) in various waste deposits from copper mining and smelting<sup>a</sup>

Element	Heap-roasting slag	Mill tailings	Flue residue	Background
As	1,070	2,960	10,400	10.0
Cd	13.4	8.0	—	0.22
Cu	7,000	6,730	37,100	45.0
Pb	1,030	2,740	—	20.0
Zn	18,000	11,000	—	95.0

<sup>a</sup>Data from References 19 and 56.

throughout the world represent the largest primary contamination sources associated with metal extraction (9).

#### Atmospheric secondary contamination

Nonferrous smelter operations throughout the world result in widespread dispersion of secondary contamination (2), emitting  $3.7 \times 10^6$  MT of air pollutants per year (21). The oldest smelting process, "heap roasting" (intermixing ore and timbers and then burning the wood), released massive amounts of sulfur dioxide and metals to the atmosphere (2). In the late 1880s, when heap roasting was prevalent at Butte, Montana, in the Clark Fork Complex, the resulting fumes were quite noxious (25):

"... Ore was being roasted outside in the grounds of the reduction works, the fumes rising in clouds of cobalt blue, fading into gray, as it settled over the town like a pall. . . . The driver reined his horse as we entered the cloud of stifling sulphur and cautiously guided them up the hill. A policeman, with a sponge over his mouth and nose, to protect him from the fumes, led us to a little hotel in Broadway, for we could not see across the street."

When large-scale smelting techniques were developed, contamination became larger scale. Within months of beginning production in a new copper smelter in 1902 at Anaconda, Montana, arsenic poisoning occurred in cattle, sheep, and horses over an area of 260 km<sup>2</sup> (26). One ranch 20 km downwind of the smelter lost 1000 cattle, 800 sheep, and 20 horses during the first year of smelter operation. Even after construction of a flue system to settle the solids in the smoke, releases of 27,000 kg/day arsenic, 2300 kg/day copper, 2200 kg/day lead, 2500 kg/day zinc, and 2000 kg/day antimony from the stack were documented (26). Today soil contamination visibly affects vegetation and cropland over an area of at least 300 km<sup>2</sup> surrounding the Anaconda smelter site (19).

Contaminated soils and farm animals are common legacies of large-scale smelting operations (21, 27–29). About

104 km<sup>2</sup> surrounding the Sudbury, Ontario, copper smelters are denuded of vegetation (2), as are nearly 70 km<sup>2</sup> surrounding the Copper Hill smelter in Tennessee. In Japan, about 67 km<sup>2</sup> of agricultural soils suffer extreme pollution, greater than allowable limits for rice production. Estimates of agricultural land lost in the United Kingdom since the Romans began metal extraction total 4,000 km<sup>2</sup> (9).

#### Secondary contamination in acid waters

Facilitated by bacterial decomposition, acid is produced when metal sulfide ores react with oxygen-rich water, forming metal ions, sulfate, and hydrogen ions. The resulting acid mine water is a common component of contamination in mine workings and waste rock piles. During underground and surface mining in the Clark Fork Complex, acidic, metal-contaminated groundwater was pumped from the workings. When open-pit mining ended in 1982, pumping was discontinued and water began

filling underground shafts and tunnels and the 390-m deep Berkeley pit. These waters contain metals and sulfate concentrations thousands of times those found in uncontaminated water. The pit probably contains 70 MT arsenic, 20,000 MT each copper and zinc, and 100,000 MT sulfur (from data in References 19 and 30). Groundwater flow raises the water level 22 m per year (19). The hydrology is not well known, but contaminated water could flow into an adjacent alluvial aquifer or overtop the pit rim and flow directly onto the ground surface by the turn of the century, compounding existing contamination problems. The massive scale of acid mine water problems in the Clark Fork Complex is common at other large-scale mining operations in temperate and humid areas.

Groundwater contamination is also associated with tailings ponds. Complex hydrology and chemistry are typical of such environments. Recently constructed ponds in the Clark Fork Complex are full of water, pH is near neutral, and sufficient organic matter is available to establish anaerobic conditions. Sulfides produced in these sediments immobilize cadmium, copper, lead, and zinc, but contaminants in more soluble reduced forms, such as arsenic, migrate into groundwater. In older, dry ponds organic material is limited and small inputs of water oxidize sulfides, so that pH is reduced and metals are carried into the alluvial aquifer. The Clark Fork experience suggests that determining the distribution, fate, or movement of contaminants from



Open-pit mine in the Clark Fork Complex that is expected to overflow near turn of century



diverse expanses of tailings ponds can be extremely difficult.

### Distal contamination from particulates

River transport of contamination from areas subject to long-term deposition of contaminants may be much more extensive than suggested in previous studies. In the Clark Fork Complex, metals enter streams and rivers from primary sources as solutes and particulates and contaminate sediments in the river and reservoirs far downstream from the primary sources (31–35). Concentrations of metals in sediment at the confluence of the headwater tributaries are 20 to more than 100 times higher than in uncontaminated tributaries. Downstream concentrations follow an exponential decline (data in References 31, 32, 34) when viewed over several hundred kilometers. At 380 km, concentrations still exceed those in the least enriched tributaries by 10 times or more. If the exponential function is extrapolated further downstream it suggests that detectable enrichment of most metals would extend more than 500 km.

Until the early 1900s, much of the waste material from milling and smelting in the Clark Fork Complex was sluiced onto surrounding land or into streams. Two tributaries in the headwaters transported the bulk of these wastes. These streams, although only 0.4% of

the total discharge of the Clark Fork River, have supplied the majority of contaminants to the drainage. The addition of huge amounts of sediment to the river system plugged stream beds, causing flooding (36) and deposition of contaminants on the surrounding floodplain. Vast areas of the floodplain became contaminated wastelands (slickens), where no plants survived and the ground was encrusted with bright blue and green copper sulfates. Not much has changed in 70 years. Slickens can still be seen along the Clark Fork River for over 100 km from its origin; highly contaminated banks also have been found 200 km downstream (31, 32). It is estimated that more than 2 million m<sup>3</sup> of contaminated sediments are stored in the floodplain of the Clark Fork and its tributaries (19, 37). This type of secondary contamination provides a huge continuing nonpoint source of metals as the river meanders through its floodplain.

Unrestrained discharges of metal-contaminated sediment continue at some modern metal extraction sites. For example, large-scale metal extraction operations on the Papua New Guinea island of Bougainville have released over  $600 \times 10^6$  MT of tailings directly into the Kawerong River and continue to add  $130 \times 10^5$  MT daily (22). These wastes cover about 18 km<sup>2</sup> in the river system and the Jaba Delta (30 km distant).

There are four dams on the Clark Fork River. Elevated concentrations of some contaminants have been detected in sediments of all the reservoirs (Figure 2, 35). Although the dams inhibit particulate transport, they do not appear to affect the downstream trend of contamination in the river. Reservoir sediments also act as a toxicant sink and can be a source of contamination of ground waters.

Tertiary contamination was discovered at Milltown Reservoir in the Clark Fork Complex (17), more than 200 km from the mines and smelters at Butte and Anaconda. Filled with contaminated sediment, the reservoir retains approximately 100 MT cadmium, 1600 MT each arsenic and lead, 13,000 MT copper, and 25,000 MT zinc. Community water wells adjacent to the reservoir contain arsenic concentrations higher than the EPA drinking-water standards and have been closed and replaced with another water source. Oxidation-reduction processes release arsenic from the reservoir sediments (17), contaminating nearly 3 km<sup>2</sup> of the aquifer. This type of tertiary contamination may be common at large-scale metal extraction sites throughout the world.

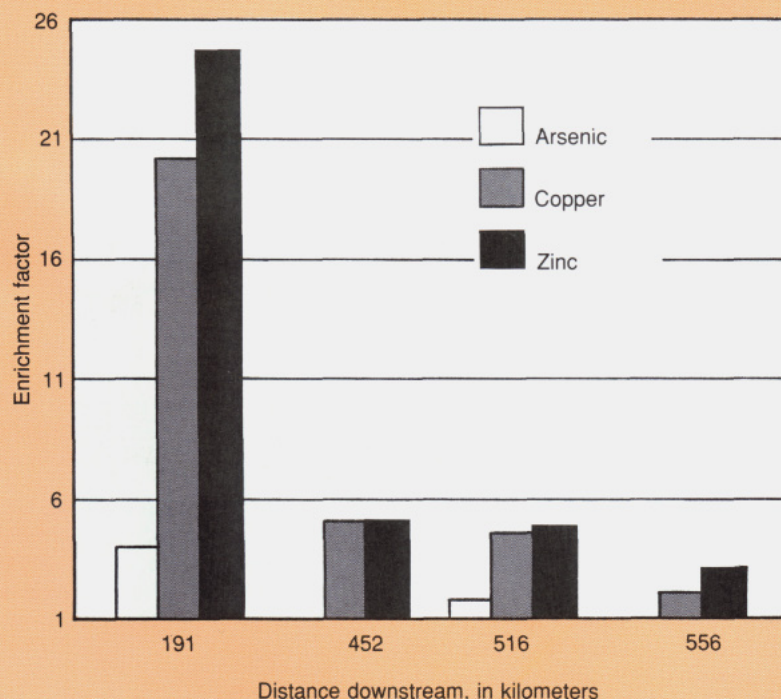
### Effects on ecosystems

High concentrations of copper, zinc, cadmium, lead, and arsenic in rivers resulting from metal extraction have adversely affected fisheries in more than 21,000 km of rivers (21). Rivers in Montana of similar size and comparable habitat to the Clark Fork typically support 3000 to 5000 trout per kilometer. Trout densities in the Clark Fork exceed one-tenth of this in only one small reach of river; in some reaches densities are only 80 trout per kilometer or lower (19, 38).

Biota in the Clark Fork are subjected to chronic metal exposures via contaminated sediments accompanied by periodic episodes of extreme contamination that occur during high flow events. The chronic exposures extend at least 380 km downstream, as evidenced by contamination in the tissues of benthic animals from the food web of trout (Table 2). The episodic events appear to be restricted to the upper 100 km of river and can be accompanied by extensive fish kills in the upper 10 to 20 km of the river. The number of taxa in the benthic community of the river also is reduced in the upper 100 to 200 km (39, 40), and the surviving species are those typically found in metal-contaminated streams.

The ecological effects of pollutants often result from complex interactions of toxins with other environmental processes (41). Better understanding of the relative effects of the different types of

FIGURE 2  
Enrichment factors for metals in sediments in the reservoirs on the Clark Fork River<sup>a</sup>



<sup>a</sup> Enrichment factor is the mean concentration in sediment divided by background concentration from uncontaminated tributaries.



exposure, along with better knowledge of in-stream toxic effects, immigration from clean tributaries, and spatial variations in metal bioavailability seem to be critical prerequisites for preserving or enhancing stressed fisheries in rivers like the Clark Fork.

### Effects on human health

Areas of active and historic mineral extraction, in general, are associated with elevated rates of death from disease in humans (42). Several of the contaminants typically associated with metal extraction activities are hazards to human health, including arsenic, cadmium, and lead (43-45). Comparisons with published national mortality statistics can provide useful preliminary indications of health risk anomalies in specific locations, although they cannot prove cause and effect.

For example, mortality from serious disease has been unusually high in the Clark Fork Complex. Studies comparing 3000 U.S. counties between 1959 and 1972 placed Silver Bow County, which encompasses the Butte mining area, among the 100 counties with the highest death rates for people aged 35 to 74 (42). In a National Institutes of Health ranking of 480 U.S. cities, Butte had the highest mortality ratio of any city in the nation for deaths from all diseases in 1950-51 and 1959-61, and the fifth highest in 1969-71 (the mortality ratio is observed deaths compared to deaths expected per capita from national statistics; 46). Elevated incidence of cardiovascular and kidney disease in Butte contributed to the elevated mortality ratio for all diseases (see Reference 43 for Butte's ranking among the 480 cities). However, Butte ranked even higher for incidence of mortality from diseases other than cardiovascular and kidney (between first and third in the nation in the three time periods).

National Cancer Institute comparisons of cancer rates by county also showed elevated incidence of some cancers in both men and women in the Clark Fork Basin (47). Some decline in mortality may have occurred between 1950 and 1979; more recent data are unavailable. The average age-adjusted mortality rate due to trachea, bronchus, and lung cancers among white males in Montana, Idaho, Wyoming, and North Dakota between 1950 and 1969 was  $25 \pm 4$  deaths per 100,000 people. Deaths from these diseases occurred at more than twice that rate in the counties containing primary contamination (48). During this period, 20.5% of the total number of such cancer deaths in Montana occurred in these counties, among 6-7% of the state's population (49). Rates for eight types of cancer in women were signifi-

TABLE 2

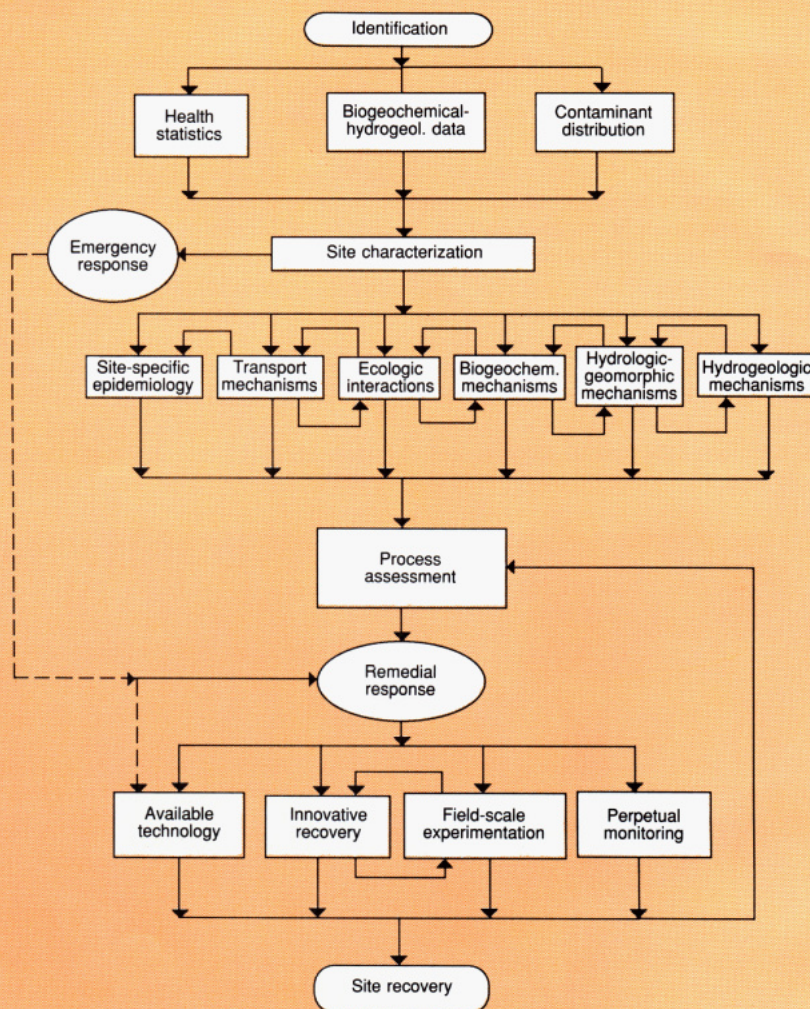
**Copper and cadmium in caddisfly larvae (*Hydropsyche* sp.) in 1986 and 1987 from the Clark Fork River**

River km <sup>a</sup>	Copper ( $\mu\text{g/g dry wt}$ )	Cadmium ( $\mu\text{g/g dry wt}$ )
3-59	$173 \pm 5$	$2.8 \pm 0.7$
61-106	$66 \pm 19$	$4.5 \pm 2.7$
107-163	$59 \pm 17$	$1.6 \pm 1.0$
163-200	$43 \pm 1$	$2.0 \pm 0$
200-381	$29 \pm 8$	$0.7 \pm 0.2$
Reference tributary	15	0.2

<sup>a</sup>Data aggregated from several stations; each aggregate is between two major tributaries.

FIGURE 3

**Remediation design for large-scale metal extraction sites<sup>a</sup>**



<sup>a</sup> Flow of specific information is not dependent on completion of other elements before continuing through the diagram; need for emergency response indicated by dashed line.

cantly greater than expected from national statistics in 1970-79; trachea, bronchus, and lung cancer as causes of death in the Clark Fork Basin were in the 96th percentile nationally.

The ultimate challenge at a hazardous waste complex is to determine if the contamination of soil, air, groundwater, and surface water threaten human health. Comparisons with available national statistics must be followed by spe-

cific studies separating risks that are occupational or related to confounding exposure such as smoking, from risks related to specific waste-associated sources of contamination.

### Strategies for remediation

The misconception that all hazardous waste sites represent single, confined problems is implicit in many discussions of remediation (6, 18, 50-54). This is



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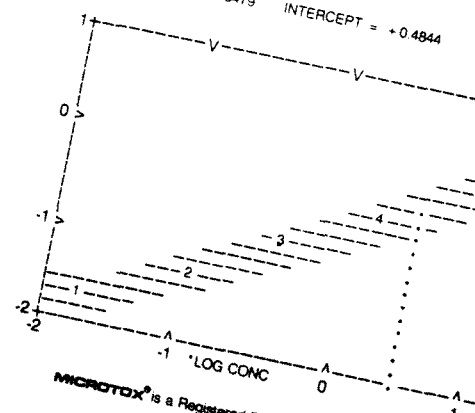
FILE NAME: test 34.05  
Sample Description: "test34"  
Procedure: Quicktest  
Initial Concentration: 90 Percent  
Ionic Adjustment: MOAS

DIL #	CONC	It	NL Dili Ass
1	0.176	80.0	
2	0.352	70.0	
3	0.703	60.0	
4	1.406	50.0	
5	2.813	40.0	
6	5.625	30.0	
7	11.250	20.0	
8	22.500	10.0	

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EC 50 = 1.623 ( 1.397 TO 1.886 )  
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not true for many metal extraction sites. A detailed common recipe for remediation of such sites is not realistic where the number of problems is large and the extent and complexity of contamination is great. Instead, successful remediation will depend on appreciating the complexities and progressively developing appropriate solutions (Figure 3).

Our review illustrates the challenge of even the first step in the process: detailed and accurate site characterization (18). Health statistics and data from the complete biogeologic and hydrogeologic system must be coupled with mapping of primary, secondary, and tertiary contaminants to determine the character and extent of metal contamination. Understanding the processes of transporting and fixing metals is essential for cost-effective and creative solutions. Efforts must involve coordinated, systematic, carefully prioritized, integrated research in all aspects of contaminant transfer. Such studies must be multidisciplinary, rigorously peer reviewed, systematic in their accumulation of knowledge, reflective of related work, and guaranteed of some continuity in support.

When a critical process is sufficiently understood, remediation of associated problems can begin. Remediation options, however, must go beyond application of "proven and effective technologies" (55). In fact, the implication that all problems at metal extraction sites are amenable to such applications is naive. The need for new or improved approaches based upon both better engineering and better process understanding is well established (50). When an immediate response is required to mitigate health risks or severe environmental effects but the best long-term solution is unclear, clean-up efforts could be approached as full-scale, real-time experiments matched with follow-up studies (51-54). Ultimately, remedial efforts should emphasize removal of metals, not just isolation of contaminants from the ecosystem.

Metal contaminants cannot be dissociated into other elemental components, as organic contaminants can; they will change form but they will not degrade through time (56). Metal recovery has the added benefit of producing a marketable product. Unless excess metals are permanently removed from large-scale metal extraction sites, most remedies will be temporary.

It is also important to accept that some of our environmental mistakes are so serious that they cannot be repaired. The near impossibility of attaining predevelopment status in the Clark Fork Complex is a clear example of this unpleasant reality. In such cases, the only reasonable remediation response may be perpetual monitoring, or perhaps the

creation of a "National Environmental Disaster Monument" that could illustrate the costs of past mistakes to future generations.

Finally, careful waste management and reduction during production of future metal reserves worldwide is imperative. On every continent, sites of mining, smelting, or other large-scale waste-generating activities may be found with potential for creating highly contaminated primary waste deposits; secondary and tertiary contamination in soil, groundwater, and surface water; and deleterious consequences for human health and ecosystems. The immense costs associated with historic sites of such contamination in the western United States clearly point out the benefits of avoiding such problems elsewhere.

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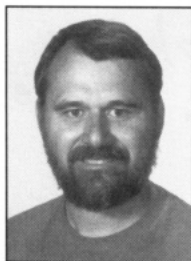
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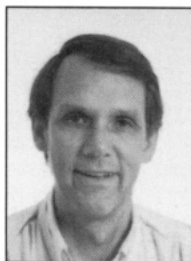
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