Distribution of Trace Metals in Fine-grained Bed Sediments and Benthic Insects in the Clark Fork River, Montana

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Abstract

The downstream distribution of Cu, Cd, and Pb in fine-grained sediments and benthic insect larvae of the Clark Fork River, Montana is characterized. This river has been heavily con laminated as a result of past mining and smelling operations near its headwaters. Concentrations of all metals in bed sediments displayed a simple exponential downstream decrease through the upper 181 km of the river. The trend suggested metal contamination originated from source(s) in the headwaters, with physical dilution occurring downstream. Additional data suggested floodplain sediments also were contaminated by the original source(s). Secondary inputs from cutbanks in the floodplains may have extended the downstream influence of the contamination. The exponential model predicted that sediment contamination should extend at least 550 km downstream, a result that was verified with data from a separate, independent study. Metal contamination, as observed in all taxa of insect larvae collected from the upper Clark Fork. Concentrations in the insect larvae were highest in the upper 100 km of the river, but downstream trends were more complex than those of the sediments. Some differences in trends occurred among taxa and metals. Areas in the river of enhanced or reduced metal contamination also were apparent. Metal contamination, however, was still evident at 381 km, the most downstream station sampled. Metal concentrations in sediments and insects decreased at the confluences of uncontaminated tributaries, but the influence of tributaries on metal contamination in the Clark Fork River was localized, extending for only 1-2 kin below the confluences.

Introduction

Copper mining and smelling operations near the headwaters of the Clark Fork River, Montana have resulted in large-scale contamination of both bed and floodplain sediments of the river (Andrews. 1987; Rice and Ray. 1985; Johns and Moore. 1985; Luoma et a1., 1989). It is estimated that over 100 million metric tonnes of mine and smelter wastes were released via Silver Bow and Warm Springs Creek to the Clark Fork River (Andrews, 1987) and that more than 2 million cubic meters of contaminated sediment have been deposited in the floodplain of the Clark Fork (Moore and Luoma, 1990). Tailings ponds constructed in the headwaters in the mid-1950s greatly reduced the contaminant input to the river. Although smelting operations ceased in the 1980s and mining activities are reduced, contaminated sediments stored in the floodplain provide a continuing source of metals to the river (Johns and Moore, 1985; Axtmann and Luoma, 1990; Moore and Luoma, 1990a).

Reduced trout populations in the Clark Fork as well as occasional fish kills during high-flow events suggest that metals in the Clark Fork may have toxic effects (Phillips. 1987; Moore and Luoma. 1990a). The purpose of this paper is to compare the downstream distribution of Cu, Cd, and Pb in fine-grained bed sediments of the Clark Fork with metal concentrations in benthic insects (which have direct contact with the sediments), and to discuss metal availability to the benthic food web in light of these distributions.

Methods and Materials

Sediment

In order to characterize the large-scale downstream distribution of metals in the sediments of Clark Fork River, fine-grained bed and floodplain sediments were collected during low flow in August 1986 and 1987 along the upper 381 km of river (all distances measured as kilometers downstream from the confluence of Warm Springs Creek) (Fig.1). Fine-grained bed sediments were collected in 1986 and 1987 from 17 sites at an average interval of 11 km throughout the upper 181 km of the river, and in 1986 at three sites in the lower 200 km of the river (Fig.1). Bed sediments were also collected from five major tributaries within 25 km of their confluence with the Clark Fork River (Fig.1).

In 1989, intensive sampling of bed sediment around the mouths of two tributaries, Rock Creek and Flint Creek, was conducted to investigate small-scale effects of tributaries on the distribution of metals in the Clark Fork. Rock Creek was chosen because it had extremely low sediment metal concentrations and might have a diluting effect on metal concentrations in the Clark Fork. Flint Creek sediments were enriched in Pb relative to Clark Fork sediments at the confluence of the rivers, and data from l986-1987 suggested there might be a Pb signal from Flint Creek in the Clark Fork sediments (Axtmann and Luoma, 1990). Triplicate samples were collected in the Clark Fork at each of three sites within approximately 1.5 km both above and below the confluences of the tributaries. Triplicate samples were also collected in the mouths of both tributaries and at sites on the Clark Fork up to 11 km above and below the confluences.

Bed sediments were sampled from the surface of deposits of fine-grained material that collected in slack waters at the river's edge using a polypropylene scoop. The sediments were immediately sieved in ambient river water through a 60 um nylon-mesh sieve. In 1986 and 1987 the sediments were dried, ground and digested in a hot, concentrated HNO3 reflux (Luoma and Bryan, 1981). In 1989 the sediments were digested with a concentrated aqua-regia-HF microwave digestion (Brook and Moore, 1988). Results from the two methods are comparable (Axtmann and Luoma, 1990; Moore and Luoma, 1990b). Copper, Cd and Pb were determined on 1986-1987 samples by flame atomic absorption spectrometry (AAS) and on 1989 samples by inductively coupled argon plasma emission spectrometry (ICAPES). Recoveries for routinely conducted analyses of NBS Reference Material 1645 (river sediment) were consistent and ranged from 70 to 97%. Both Cu and Pb concentrations of analyzed standards (106 + 4 and 686+18, respectively) fell within the 95% confidence intervals of the concentrations reported by NBS (109 +19 and 714+28, respectively). Recoveries of Cd from standards in 1986-1987 were slightly greater than in 1989 (8.2+0.2 and 7 .2 +0.8, respectively). Cadmium concentrations in all three years were consistently below the acceptable range reported by NBS (10.2+1.5).

Benthic Insects

The larvae of six taxa of benthic insects from two orders (Trichoptera and Plecoptera) were collected from the Clark Fork River and five of its major tributaries. Some taxa were rare or absent at some stations, and some taxa were preferentially collected for specific studies; thus not all taxa were represented in all samples. Data from two taxa, the filter-feeding Hydropsyche spp. (order Trichoptera) and the predaceous stonefly Claassenia sabulosa (order Plecoptera) are emphasized in this paper because they were present at most stations and represented the variety of bioaccumulation patterns observed.

Insect larvae were collected with kick nets from riffle areas at the same stations that sediments were collected in August 1986 and 1989 (Fig.1). Insects were sorted on site by taxon, then held in plastic bags filled with ambient river water in an ice cooler for 4-6 hours to allow the insects time to clear their digestive tracts. The water in the bags was then drained and the insects were frozen. Samples were thawed in the laboratory and thoroughly rinsed with distilled water to remove particulates from exterior surfaces. Identifications of specimens were verified and individuals from the same taxonomic group were sorted by size to examine possible size-related differences in metal concentrations. The body length of each Claassenia sabulosa was measured from the head to the last abdominal segment. Different sizes of Hydropsyche spp. were qualitatively separated by eye. Individuals of the same taxon and of similar size were composited into samples to attain a minimum total dry weight of 50 mg. Samples were dried at 800 C, weighed, and then digested by hot 16N HNO3 reflux. The acid was evaporated after the sample solution turned clear then residue was reconstituted in 25% HCI. Metals were determined by AAS on samples collected in 1986 and by ICAPES on samples collected in 1989. Concentrations of Cu and Cd in biological standards (NBS standard reference material 577a, bovine liver) prepared and analyzed by the same method were within the range of certified values. The reliability of the method could not be verified for Pb because concentrations in NBS reference material were below analytical detection limits.

The large-scale distribution of metal contamination in insects through the 381 km study reach was evaluated by aggregating data collected in 1986 from upstream (0-60 km), mid-river (106 -164 km) and downstream (191 -381 km) reaches. Effects of tributaries on small-scale variation in insect metal concentrations in the Clark Fork were analyzed with data collected from the same sites as sediments around the mouths of Hint Creek and Rock Creek in 1989.

Data collected in August 1986 were analyzed for correlations between insect and sediment metal concentrations. Data were log transformed for the correlation analysis. Statistical significance of a correlation (product-moment correlation coefficient, r, was set at $p < 0.05$).

Studies of the contribution of the content of the gut to whole body metal burdens of insects in the Clark Fork also were initiated in 1989. Pleronarcys californica, a detritus-feeding stonefly, was employed in this study because its digestive tract could be easily removed. Individuals of this species were collected from the Clark Fork at 168 km, from Flint Creek and from Rock Creek. Samples were collected and prepared as described above. Specimens were secured on a paraffin surface the under a microscope (magnification 8-40 x). A dorsal, longitudinal incision was made to open the body cavity and expose the gut. The gut Wall was incised and the contents were carefully removed. The gut content and the remains of the body were analyzed separately.

Results and Discussion

Sediment

Metal concentrations in tributary sediments generally reflected the history of metal extraction in the watershed (Axtmann and Luoma,1990). Sediments collected in Flint Creek, which has an extensive history of mining in its watershed, were enriched in both Pb and Cd (although Cd concentrations in Flint Creek were not as of enriched as in the Clark Fork). Less historic mining activity has occurred in Rock Creek and the Bitterroot River basins than in other tributaries. These systems had the lowest metal concentrations in sediments. Low concentrations also were observed at the mouth of the Blackfoot

River, more than 150 km downstream from the inputs of small-scale mining activities (Moore et al., 1990). The mean of the concentrations from the three latter systems was taken as an operational reference concentration for each metal (indicative of pre-mining metal concentrations) with which to evaluate metal enrichment in the Clark Fork (Fig.2).

In contrast, analysis of closely spaced bed sediment samples around the mouth of Flint Creek and Rock Creek in 1989 indicate that tributary input has only a minor and localized effect on Clark Fork sediment metal concentrations. The 1989 study showed that Cu concentrations were low in the sediments of both Rock Creek a Flint Creek, relative to Clark Fork sediment metal concentrations. When Cu concentrations from samples collected in 1989 are plotted with the regression line and 95% confidence intervals for the combined 1986 and 1987 data set, it is clear that Cu concentrations are locally depressed below the confluence of both Rock Creek (Fig. 3a) and Flint Creek, but that they quickly return to their pre-tributary concentrations. Lead concentrations, on the other hand, were higher in Flint Creek sediments than in the Clark Fork sediments, but much lower in Rock Creek. Lead concentrations show a small, and again localized, increase below Flint Creek (Fig. 3b), and a small, but local decrease below Rock Creek. These results further suggest that erosion of contaminated sediments from cutbanks below the confluence of tributaries counteracts the diluting or enhancing effect of sediments from the tributaries.

Benthic Insects

Enriched metal concentrations were observed in all six taxa of benthic insects collected from the upper Clark Fork River, compared to concentrations in animals from tributaries. However, trends in metal contamination of benthic insects in the river were complicated by differences among taxa and metals, and by the scarcity of some taxa, especially in the most contaminated reaches of the river. Hydropsychid caddisfly larvae are the most widely dispersed of the benthos targeted for collection in this study. Metal contamination of caddis fly larvae in the Clark Fork River was evident for 381 km downstream when concentrations in specimens from the Clark Fork were compared with animals from reference tributaries (Table 1). When stations were aggregated by river reach, metal concentrations in the caddis fly, Hydropsyche spp., were highest in the uppermost Clark Fork, and lowest downstream. The elevated concentrations of Cu, Cd, and Pb in the reach between 191 and 381 km downstream of the Warm Springs tailings ponds were significantly higher than in the reference tributaries ($p < 0.05$; ANOVA, except where an individual value from a reference site was compared to the aggregated numbers from the Clark Fork by t-test- Sokal and Rolf, 1969, p. 168-169). Metal concentrations in Hydropsyche spp. correlated positively with the exponential decrease observed in the metal concentrations of fine bed sediments (coefficients of determination, r2 = 0.74, 0.150, 0.54 for Cu, Cd, and Pb, respectively; $p < 0.05$). One notable difference in trends occurred for Cd between 20 and 100 km below the Warm Springs Ponds where (concentrations in Hydropsyche spp. increased despite significant decreases in sediment Cd concentrations (compare Fig. 2 and Fig. 4).

Trends in metal concentrations of the predaceous stonefly, C. sabulosa, were obscured partly because this species was either rare or absent at stations in the upper 60 km, the most contaminated reach of the river. Below 60 km, downstream trends in metal concentrations in this species were complex (Fig. 4), although concentrations were higher than found in reference tributaries. Cadmium in C. sabulosa correlated significantly with sediment Cd concentrations (r2 = 0.48; $p < 0.05$), largely as a result of the difference between the average Cd concentrations of insects collected above 180 km and below 200 km. Correlations for Cu and Pb were insignificant Copper concentrations in C. sabulosa in the Clark Fork River varied over a narrow range of 44 to 75 ug g-l, but these concentrations were 1.4- 2 times greater than in specimens of this species collected from tributaries. Lead concentrations, which were generally low in C. sabulosa, decreased sharply below the most upriver station where the species was found, however no consistent trend was evident further downstream. Concentrations of some metals were lower at stations immediately below Rock Creek (160 km; Cu and Pb) and the Blackfoot River (181 km; Cu and Cd) than at stations located further downstream.

Intensive sampling around the confluences of Flint Creek and Rock Creek in 1989 confirmed the localized influence of tributaries on metal concentrations in insects suggested by the 1986 data and the sediment data collected in 1989, Concentrations in insects in Flint Creek and Rock Creek were low relative to concentrations in the Clark Fork with the exception of Pb in Flint Creek. Lead concentrations in Hydropsyche spp. immediately below the confluence increased with inputs of Pb-enriched sediment from Flint Creek, and Cu decreased in response to an apparent dilution of sediment Cu (Fig.5). Both Pb and Cu concentrations in Hydropsyche spp. decreased 0.1 km below the confluence of Rock Creek (Fig.5, Cadmium concentrations also were lowest 0.1 km below both tributaries (data not shown). Downstream from the confluences of both streams, concentrations in insects returned to values similar to those observed above the confluences.

Although contamination in the undigested content of the gut was important in Pteronarcys, the results also indicated that the body (minus the gut) contained substantial quantities of metals. Thus whole body metal concentrations do not simply reflect gut contamination. More important, whole body metal concentrations and metal concentrations of the body without the gut both generally reflected the relative differences in metal contamination among the three sites studied, so negative or positive influences of the gut content did not change interpretations of metal distributions in the river. Gut contamination in other taxa in the Clark Fork has not been determined, thus it is not known how typical the results for P. californica are. Smock (1983b) analyzed the metal contents in the gut material of 40 taxa divided into five different feeding categories. Although detritivores were not singled out as a separate category, the results from his study showed that the proportions of metals in the gut contents of insects that ingest detritus and plant material were similar to filter feeders. Hydropsyche spp. was included with several other taxa in this later group. In contrast, predators had significantly lower proportions of metal in their gut content.

Conclusions

Extensive metal contamination resulting from historic discharges of mining and smelting wastes was evident in both fine-grained bed sediments and in benthic insects of the Clark Fork River. Elevated sediment metal concentrations extended at least 550 km downstream from the original source of contamination in headwaters, and contamination of biota extended at least 380 km downstream.

Data from both bed sediment and benthic insect larvae indicate that metal contamination was greatest in the most upstream reaches of the river and decreased downstream. Although bed sediment metal concentrations follow a distinct exponential decline with distance away from the source, trends in insect metal concentration were more complex. Areas of enhanced and reduced metal concentrations occurred on spatial scales ranging from less than 5 km to 10's of km were evident.

The small body of available evidence suggests that the whole body trace metal concentrations of aquatic insects reflect in at least a relative sense levels of biologically available metals. The complex trends in the Clark Fork River, suggest that the bioavailability of metals is spatially heterogeneous and element-specific. Local conditions may create islands of reduced or enhanced metal bioavailability, especially where there are sharp physical and/or chemical boundaries such as at the confluences of tributaries. In other cases the causes of the differences in bioavailability are less clear. Cadmium, for example, appeared to exhibit greater bioavailability to caddisfly larvae in the upper 100 km of the river than indicated by the sediments, in the absence of any recognized chemical or physical change in the river.

Simple relations between environmental contaminant concentrations and animal contaminant concentrations are rarely observed in nature because of the complex interaction of geochemical and biological factors (Luoma, 1989). Extractions of sediments with strong acids may not be sensitive to geochemical conditions that affect metal bioavailability (Luoma and Bryan, 1978; 1982; Langston, 1986). Differences in food selection may contribute to differences in whole body metal concentrations among species, with concentrations being higher in species that ingest sediment and detritus than in predators (Smock, 1983b). The difference in Pb concentrations between Hydropsyche spp., a filter feeder, and Claassenia sabulosa, a predator, (Fig. 4) is consistent with Smock's (1983b) conclusions. The absence of C. sabulosa from the most contaminated reach of the river may also have weakened statistical correlations between sediment and metal concentrations in this species.

The effect of tributary input on both benthic insect larvae and sediment metal concentrations was localized, suggesting tributaries are also a source of variability in trends. Inputs of tributaries with relatively low sediment metal concentrations appeared to be responsible for localized decreases in metal burdens in insect larvae, presumably because exposure was reduced at the confluence of the tributaries either by physical mixing (Clark Fork sediments and/or water were diluted with relatively pristine material from the tributaries) or by a physicochemically induced decrease in the biological availability of metals. Flint Creek had an analogous, but opposite effect on Pb concentrations, by being an additional source of Pb-enriched sediments to the Clark Fork. Drift of insects from the tributaries into the main stem of the river could also have influenced metal concentrations in insect samples collected below the confluences.

Concentrations of metals in both benthic insects and bed sediments returned to their pre-tributary concentrations within 1 or 2 km below both Rock Creek and Flint Creek. Two possible explanations for such a localized effect are that tributary sediment loads are small compared to the Clark Fork and/or input of contaminated material from cutbanks below the tributary confluence quickly overshadow any diluting or enhancing effect of the tributaries.

Phillips (1985) reported enhanced fish populations directly below the confluence of Rock Creek. This study suggests that areas below the mouths of uncontaminated tributaries may provide small, localized refuges from severe metal contamination for biota of the Clark Fork River. How biological communities respond to such patches of reduced metal exposure merits study.

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Figure 1: Map of study area, showing the location of sediment sampling stations on the Clark Fork River and its tributaries for 1986 and 1987.

Figure 2: Metal concentrations in fine-grained (< 60 mm) bed sediments in the Clark Fork River. Closed symbols are 1986 data, open symbols are 1987 data. Distance downstream is measured as river km downstream of the confluence of Warm Springs Creek. Mean metal concentrations (MRC) of sediment from uncontaminated tributaries are indicated on the right hand ordinate.

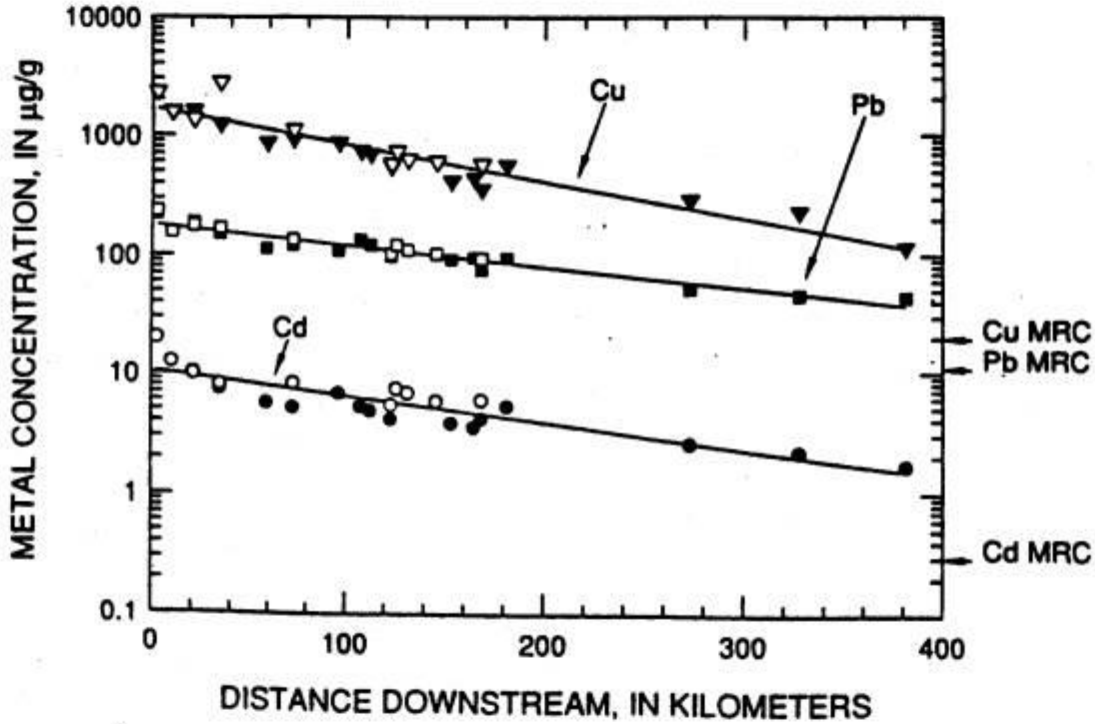

Figure 3a & b: Mean (+/- 1 stdv) of Pb and Cu concentrations in bed sediments collected above and below the confluences of Flint Creek and Rock Creek, respectively, in 1989 plotted with the regression line (solid) and the 95& confidence intervals (dashed lines) for the regression of metal concentration on river km for the combined data for 1986 and 1987. The metal concentrations in sediments collected from each tributary are indicated by an arrow.

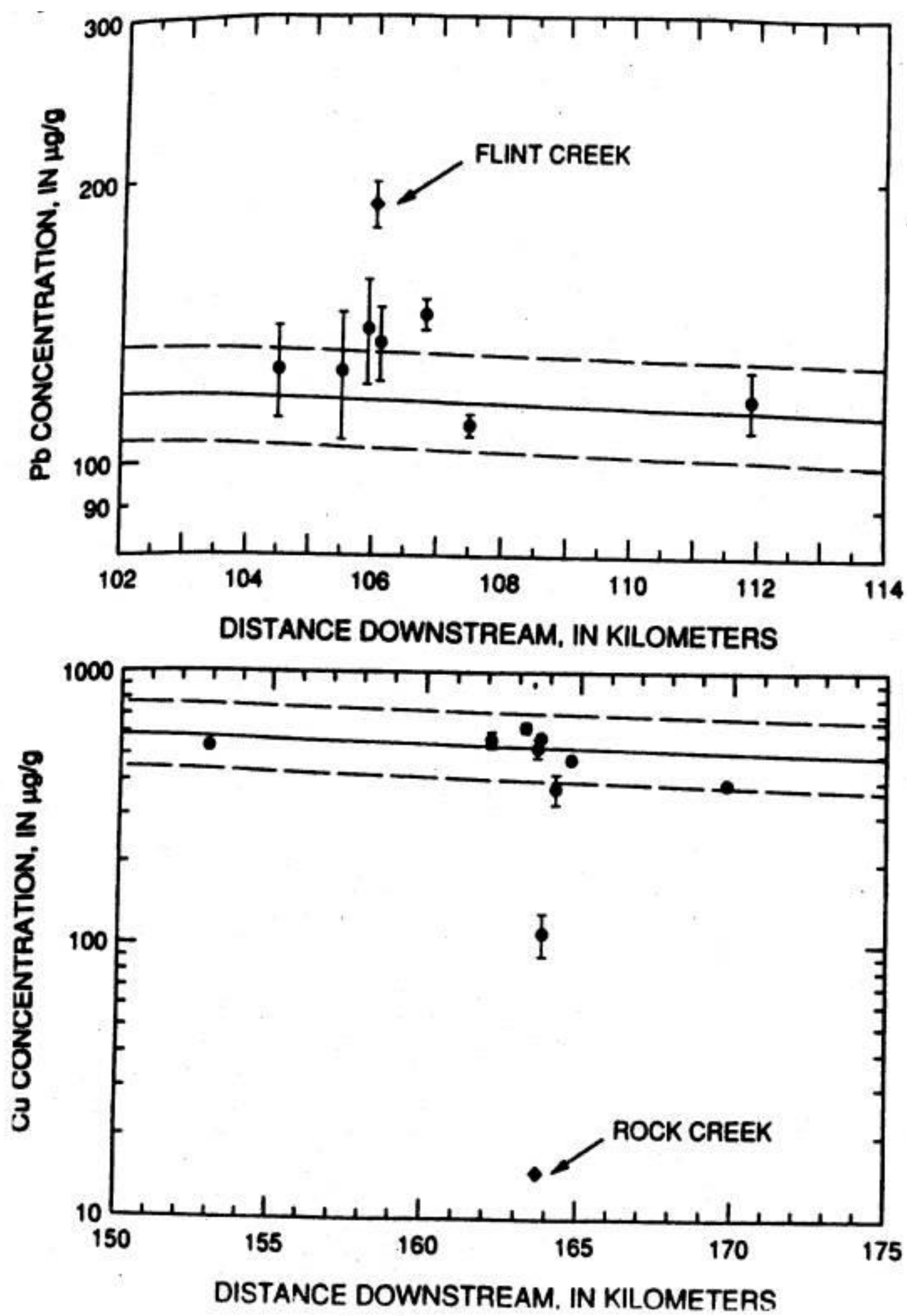

Table 1: Grand mean (data from several stations) (+/- 1 sem) of trace metal concentrations (mg/g) in caddisfly larvae (*Hydropsyche* spp.) collected in 1986 in the Clark Fork River upstream of the confluence of the Little Blackfoot River (0-60 km), between the confluences of Flint Creek and Rock Creek (107-164 km), and between the confluences of the Blackfoot and Flathead Rivers (191-381 km) compared to metal concentrations of specimens from tributaries. Tributary concentrations are for samples (number of stations shown in parentheses) from the Little Blackfoot River, Rock Creek, and the Blackfoot River.

km	number of stations	Cu	Cd	Pb
0 - 60	3	204±18	2.8 ± 0.6	12.8±1.5
106 - 164	3	72 ± 8	2.2± 0.6	8.1 ±1.3
191 - 381	3	27 ±5	0.7 ± 0.2	3.1 ±0.9
tributaries		9 ± 3 (4)	0.2 (1)	0.2 (1)

Figure 4: Downstream profiles of the mean metal concentrations in *Hydropsyche* spp. (closed symbols) and *Claassenia sabulosa* (open symbols) collected in 1986.

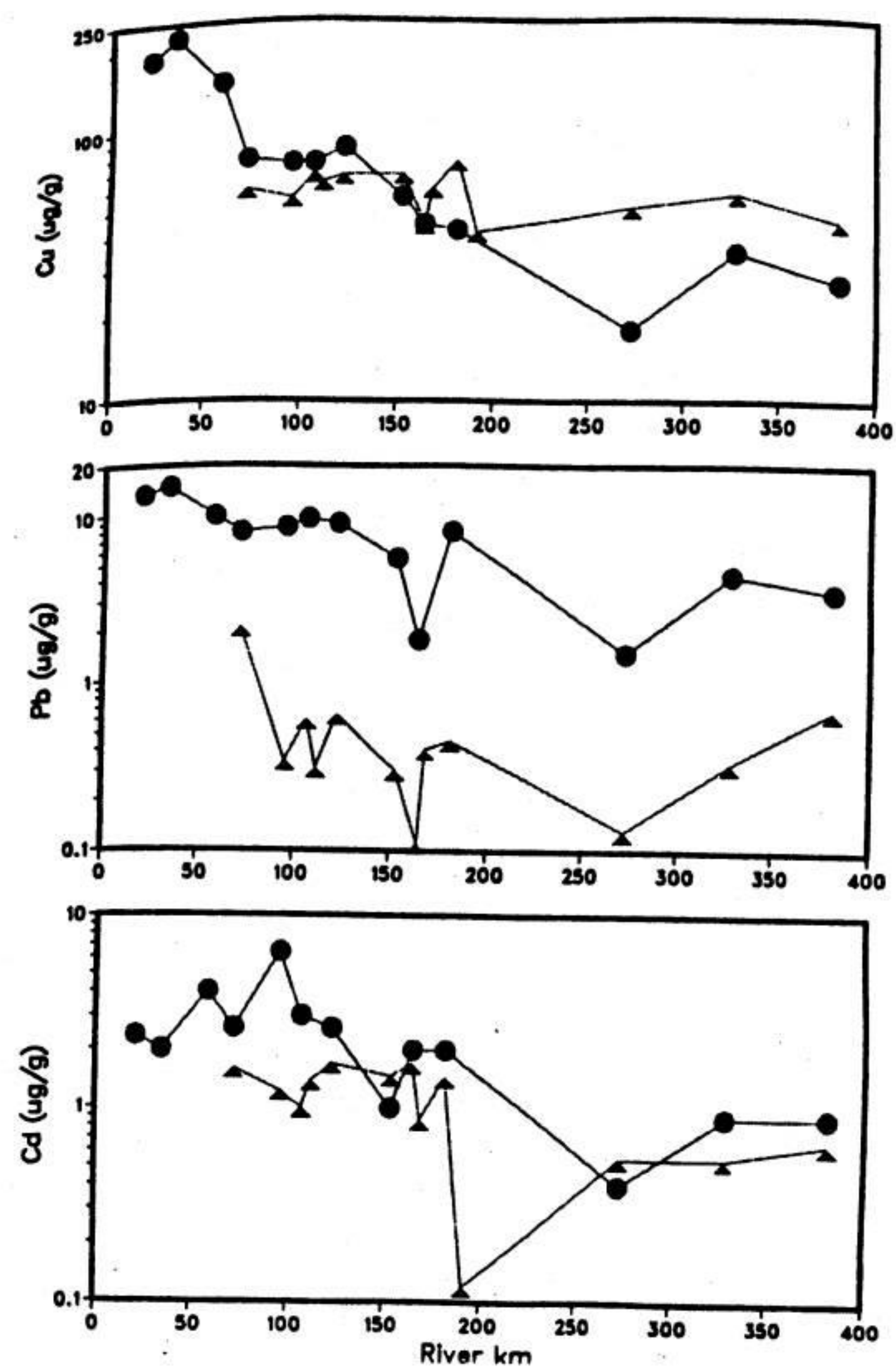

Figure 5: Mean (+/- stdv) of Cu and Pb concentrations of *Hydropsyche* spp. (closed symbols) collected above and below the confluences of Flint Creek and Rock Creek in 1989. Open symbols are the metal concentrations of specimens collected from each tributary (sample sizes range from 1 to 5).

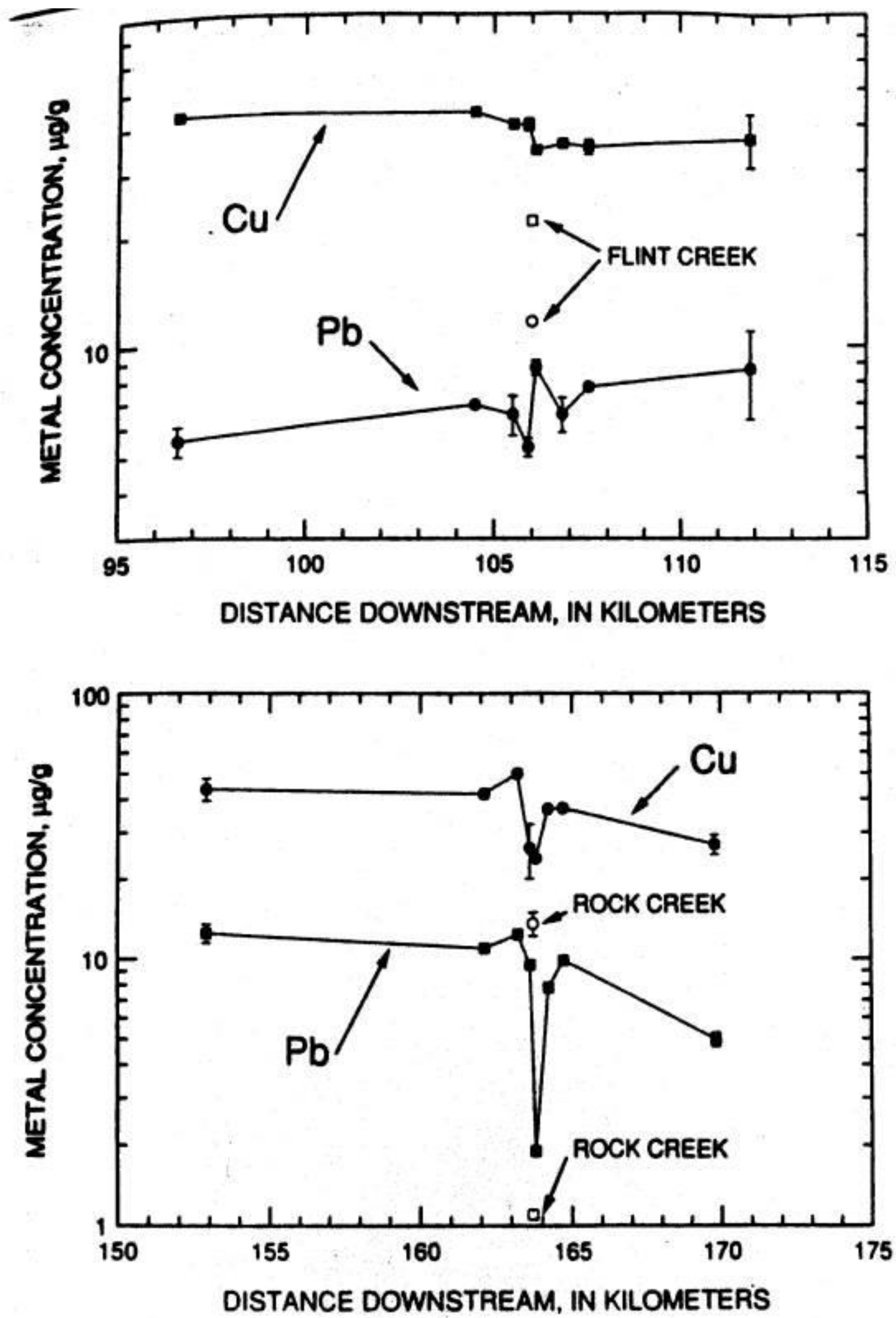

Table 2

Table 2. **Metal in the gut content of the stonefly *Pteronarcys californica* a percent of the total metal mass (mg) of the entire body. Values are the mea ± 1 SEM of 1 to 5 composite samples. Lead in Rock Creek samples was belo analytical detection limits.**

Station	Al	Cd	Cu	Fe	Pb	Zn
Flint Creek	86±2	21	35±8	84±2	50±6	38±8
Rock Creek	90±2	bd	26±1	90±1	bd	31±2
Clark Fork	89±2	57±3	34±2	90±1	60±2	30±1