TRACE METALS IN FISH EXPOSED TO ASBESTOS RICH SEDIMENTS

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Abstract. Chrysotile asbestos fibers which contain high levels of Ni, Cr, Co, and Mn were introduced into the Sumas River by a landslide. The pH in the streamwater decreased with increasing distance from the landslide input source and in the process the trace metals in the asbestos rich sediments get leached. The findings from this study show that some of the trace metals such as Mn and Ni are elevated in several fish species. The Mn levels in fish muscle tissue and whole fish were found to be significantly higher than levels reported in the literature. Similarly Ni levels in whole fish were high. Threespine stickleback were found to have the highest Mn and Ni levels. Besides the elevated metal levels we have so far observed no other abnormalities in fishes of the system.

1. Introduction

Significant asbestos fiber concentrations have been reported in many rivers (Schreier and Taylor, 1980) and health concerns have been expressed regarding the use of such water for consumption. Recent reviews by Cummins (1984) and Toft *et al.* (1984) indicate that potential cancer risks from consumption of asbestos rich water are small. However, the effect of asbestos fibers on the aquatic biota has not been investigated in any depth. There is a legitimate concern because Proctor and Woodell (1975), Schreier and Timmenga (1986), and Schreier *et al.* (1986) have shown that terrestrial biota is adversely affected by the presence of asbestos rich materials. The medical researchers have focused on the physical properties of fibers as a cause of disease, but plant ecologists have long claimed that trace metals and nutrient imbalances associated with asbestos are the primary causes of stress (Proctor and Woodell, 1975).

Trace metals such as Ni, Co, Cr, and Mn are present in high concentrations in almost all materials rich in chrysotile asbestos (Cralley *et al.*, 1968; Morgan *et al.*, 1973; Teherani, 1985). Trace metal analysis has been used as an indication of asbestos exposure in several studies (Schreier *et al.*, 1986; Schreier and Timmenga, 1986). These metals are present in part as a component of the chromite and magnetite contamination in asbestos (Schwarz and Winer, 1971) but significant amounts are also present in the chrysotile structure as isomorphic substitutions (Morgan *et al.*, 1973).

Chrysotile asbestos is a magnesium silicate mineral that is only stable at very high pH and once exposed to pH < 8.0 the Mg is leached from the fiber structure in the form of $Mg(OH)_2$. During this process trace metals also are released and a fragile silica fiber structure remains (Choi and Smith, 1972; Morgan *et al.*, 1973; Morgan and Holmes, 1986). We expect this process to take place in many river systems but often the concentrations are too low and the analytical techniques too tedious to demonstrate the process.

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In 1975 an asbestos rich serpentinitic bedrock formation was exposed by a major landslide in the headwaters of the Sumas River basin. Since that time large quantities of chrysotile asbestos fibers are introduced into the stream system. We initiated this study to document potential effects of asbestos on the fish population by analyzing trace metal concentrations in both fish muscle tissue and whole fish homogenate.

Very little work has so far been carried out on the potential effects of asbestos on fish and other aquatic biota (Hanson, 1985). Belanger *et al.* (1986, 1987), Batterman and Cook (1981), and Lauth and Schurr (1983) are the only published works involving aquatic organisms and asbestos exposure. In these studies asbestos fiber analyses have been conducted in animal tissue but no trace metal concentrations were determined. We chose to analyze trace metals rather than asbestos fibers in fish because there are considerable difficulties in quantifying asbestos fibers in tissues originating from waters contaminated by asbestos.

The surface conditions and fiber durability of asbestos are important medical considerations and recent work by Morgan *et al.* (1977), Monchaux *et al.* (1981), and Jaurand *et al.* (1984) have shown that the cancer incidence in test animals decreased significantly once the asbestos fibers have been leached with acids prior to administration to the animals. A somewhat similar process is expected to take place in the stream water where fibers are leached and associated trace metals are then released. We hope to determine the potential effects of this process in some of the resident fish in the stream as well as those which remain there up to a year or more as juveniles.

2. Background of Study Site

The Sumas River originates in the Cascade Mountain range in Northwestern Washington State, enters Canada near Everson and flows into the Fraser River near the Vedder Canal (Figure 1). A massive landslide introduced serpentinitic sediments with large quantities of chrysotile asbestos fibers into the stream system. A major inundation of agricultural land occurred in 1975 and a similar event occurred in 1983 and 1984. In each case asbestos rich sediments remained on the land constituting a health hazard to the human population and creating toxic conditions to the terrestrial biota. Since the river carries some of the highest known asbestos fiber concentrations, our recent interest focused on the effect of this material on the fish population. The river is unaffected by industry and the main land use activities are forestry in the headwaters, and intensive dairy agriculture in the lower reaches of the basin. The asbestos fiber point source input occurs some 16 km above the international border and the Canadian section of the stream to the Fraser River confluence is some 25 km long (Figure 1). The maximum annual discharge at the hydrometrics station near the international border usually occurs in January (38.7 $m^3 s^{-1}$, January 6, 1984) and minimum flow levels occur during the summer (0.96 $m^3 s^{-1}$, June 9, 1983). The asbestos fiber and trace metal concentrations decrease with increasing distance downstream from the landslide but at the border both the fiber and metal concentrations are still higher than regional background levels (Schreier, 1987). The stream pH levels decrease from pH 9.0 at the

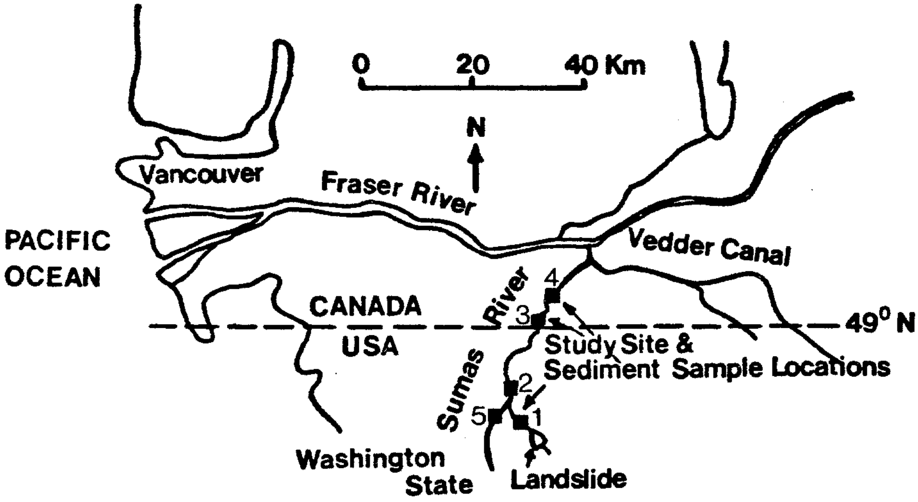

Fig. 1. Overview of study area and location of sampling stations.

asbestos source area to pH 7.1 at the border, and is likely due to agricultural inputs from manure and fertilizers. This decrease in pH is of concern because of the instability of chrysotile in pH < 8.0, where Mg and trace metals are released into the water.

3. Materials and Methods

Fish specimens were collected on two occasions using an electroshocker and a seine net. The first sampling occurred on May 14, 1985, and the second on June 13, 1986. The stream section immediately below the international border was sampled near the hydrometric station and approximately 16 km below the landslide. This site was chosen because: (1) logistical difficulties hindered us from operating across international boundaries, (2) there was no evidence of fish occurrence in the upper reaches of the river near the landslide, and (3) the border site has asbestos levels which were in the 10^9 fiber L^{-1} range which is 1 to 2 orders of magnitude greater than most natural streams. The latter conditions are more conservative and realistic in comparison to the 10^9 to 10^{12} fibers L^{-1} level obtained in the upper reaches of the basin.

The fish were identified, measured and weighed in the laboratory. Epaxial white muscle tissue samples were collected using an acetone-cleaned scalpel. This process could only be carried out on the larger fish and in many cases the tissue of several specimens had to be pooled in order to obtain sufficient sample weight for chemical analysis. All small fish specimens were washed with distilled water and combined into pooled samples for whole fish analysis. A total of 39 samples originating from 259 fish specimens and covering eight different species were analyzed for trace metals (Table I).

Tissue samples and whole fish samples were minced in a stainless steel grinder and samples of the homogeneous mixture were weighed and placed into a freeze dryer for

TABLE I

Fish samples used in study

1985 fish samples				1986 fish samples			
Epaxial white muscle tissue analyses				Epaxial white muscle tissue analysis			
Species[a]	# of fish	$\bar{X}$ fork length	$\bar{X}$ wet weight	Species[a]	# of fish	$\bar{X}$ fork length	$\bar{X}$ wet weight
Prickly Sculpin	3	10.4 cm	14.0 g	Prickly Sculpin	1	16.4 cm	55.5 g
Prickly Sculpin	3	10.9 cm	14.9 g	Prickly Sculpin	2	13.8 cm	31.4 g
Prickly Sculpin	2	10.5 cm	13.5 g	Prickly Sculpin	4	12.2 cm	17.0 g
Northern Squawfish	1	14.8 cm	32.7 g	Northern Squawfish	4	9.1 cm	8.2 g
Northern Squawfish	1	11.8 cm	19.0 g	Coho Salmon (fry)	10	4.6 cm	1.2 g
Coho Salmon (smolts)	2	10.6 cm	9.5 g	Redside Shiner	3	9.0 cm	9.3 g
Coho Salmon (smolts)	2	9.2 cm	8.7 g	Threespine Stickleback	10	6.4 cm	3.5 g
Redside Shiner	3	8.8 cm	10.3 g				
Cutthroat Trout	1	12.9 cm	24.5 g				
Cutthroat Trout	1	11.1 cm	13.6 g				
Whole fish analyses				Whole fish analyses			
Species[a]	# of fish	$\bar{X}$ fork length	$\bar{X}$ wet weight	Species[a]	# of fish	$\bar{X}$ fork length	$\bar{X}$ wet weight
Prickly Sculpin	2	5.6 cm	2.0 g	Prickly Sculpin	4	5.8 cm	2.1 g
Coho & Chum Salmon (fry)	6	4.8 cm	1.2 g	Coho Salmon (fry)	10	4.1 cm	0.8 g
Coho (fry)	7	4.1 cm	0.9 g	Coho Salmon (fry)	10	3.9 cm	0.7 g
Threespine Stickleback	14	4.7 cm	1.8 g	Coho Salmon (fry)	8	4.2 cm	0.9 g
Threespine Stickleback	14	4.7 cm	1.8 g	Threespine Stickleback	10	5.4 cm	2.1 g
Threespine Stickleback	13	4.6 cm	1.7 g	Threespine Stickleback	10	5.3 cm	1.8 g
Threespine Stickleback	13	4.5 cm	1.6 g	Threespine Stickleback	10	5.0 cm	1.5 g
Northern Squawfish	2	4.2 cm	1.0 g	Threespine Stickleback	10	5.0 cm	1.4 g
				Threespine Stickleback	10	4.4 cm	1.4 g
				Threespine Stickleback	10	4.4 cm	1.4 g
				Threespine Stickleback	20	4.0 cm	0.9 g
				Redside Shiner	20	3.1 cm	0.5 g
				Largescale Sucker	1	9.5 cm	10.1 g

[a] Chum salmon = *Oncorhyncus keta*; coho salmon = *Oncorhynchus kisutch*; cutthroat trout = *Salmo clarki*; largescale sucker = *Catostomus macrocheilus*; northern squawfish = *Ptychocheilus oregonensus*; prickly sculpin = *Cottus asper*; redside shiner = *Richardsonius balteatus*; threespine stickleback = *Gasterosteus aculeatus*.

48 hr. Subsequently the samples were subjected to a low temperature plasma oxygen ashing procedure and the light brown ash was dissolved in concentrated HNO_3 on a hot plate. Equal parts of concentrated HCl were added after cooling. After appropriate dilutions with deionized water the samples were analyzed with a Jarrel Ash inductively coupled argon plasma spectrometer (ICAP) and with an atomic absorption spectrometer with a graphite furnace attachment (GTA). A U.S. Environmental Protection Agency (EPA) fish reference sample was used as a standard for the analysis.

TABLE II

Water quality and sediment chemistry at study site

Variables – Streamwater	# of samples	Concentration range 1983–1984
Asbestos fiber concentration	7	8.0×10^7–2.3×10^{11} fibers L^{-1}
Asbestos fiber length	7	1.3 – 2.2 μm
Asbestos fiber width	7	0.10– 0.12 μm
pH	7	7.1 – 8.0
Ca	7	9.1 –20.2 mg L^{-1}
Mg	7	11.7 –19.3 mg L^{-1}
Cr	7	0.3 – 4.0 μg L^{-1}
Mn	7	0.3 –70.0 μg L^{-1}
Ni	7	8.0 –23.6 μg L^{-1}
Variables – Sediments (< 63 μm fraction)		
Ca	6	2.3– 2.7%
Mg	6	5.2– 6.3%
Cr	6	170– 217 μg g^{-1}
Mn	6	1260–1903 μg g^{-1}
Ni	6	416– 564 μg g^{-1}
Co	6	26– 45 μg g^{-1}
Cu	6	29– 58 μg g^{-1}
Zn	6	112– 169 μg g^{-1}

A detailed description of the water quality and sediment conditions in the stream is given by Schreier (1987). Summary data in Table II provide information on the asbestos fiber and trace metal concentrations at sampling station #3 (Figure 1) where the fish were collected. Figure 2 represents an example of the effect of the landslide on the streamwater and sediment chemistry in the downstream direction. Water and sediment samples were collected on seven different occasions during the 1983–1984 hydrologic year and covers both the minimum and maximum annual flow conditions. Water samples were filtered and analyzed for major ions and metals within 48 hr of collection using conventional methods described by Inland Waters Directorate (1979). Total metal values in the water were determined using a Varian Spectrometer with a graphite furnace attachment (GTA). The total elemental composition of the sediments was carried out on the smaller than 63 μm fraction using an aqua regia and hydrofluoric acid digestion

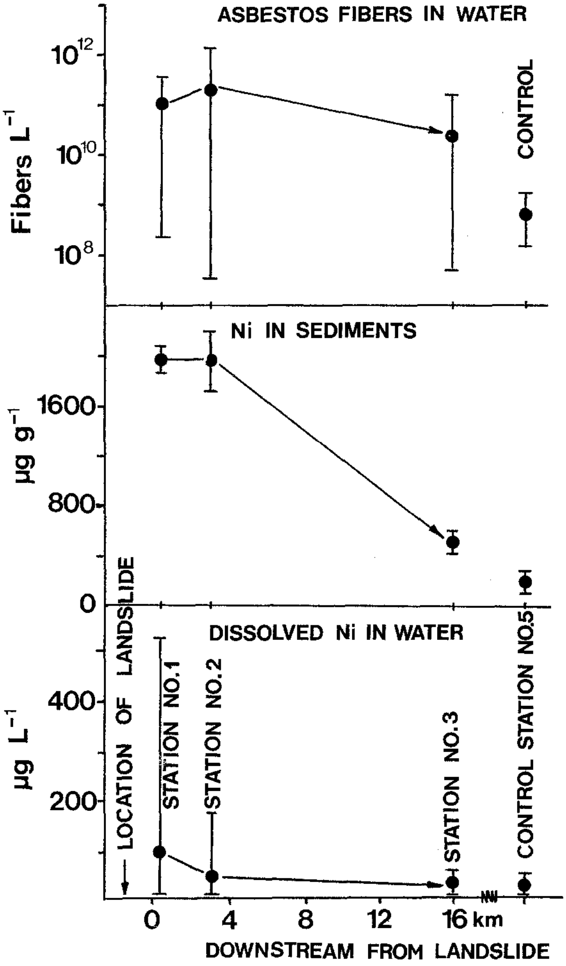

Fig. 2. Asbestos fiber and Ni concentrations in water and sediments downstream of landslide (mean annual value and range over 1983–1984 hydrological cycle, $n = 7$).

method (Price and Whitestide, 1977) and the solutions were analyzed with an inductively coupled argon plasma spectrometer (ICAP).

4. Results and Discussion

The water quality and sediment conditions which prevail at the test site (Station No. 3) are summarized in Table II. Asbestos fiber concentrations are subject to seasonal variations and are very closely correlated with discharge rates (Schreier, 1987). The trace metals associated with the asbestos fibers follow the same trend. Little seasonal variation was observed in the trace metal concentrations in the sediments but Ni, Mn, and Cr values were well above those conditions found in natural streams, unaffected by asbestos fibers.

The site where the fish were collected is some 16 km downstream from the asbestos input area and as shown in the example in Figure 2 the fiber and Ni concentrations decrease with increasing distance from the landslide. Chromium, Co, and Mg which are all present at high levels in the asbestos, followed the same trend, while Cu, Zn, and Mn showed the opposite effect (Table III). Copper and Zn are not present in asbestos and the increase in those cases may be due to agricultural input in the lower portion of the stream. Manganese concentrations are high in asbestos but this metal did not follow the same trend as the other trace metals associated with asbestos. The sediment concentrations increased in the upper section of the river and then decreased (Table III). Agricultural inputs and/or reduced leaching in alkaline media might be a possible explanation for this anomaly. In any case the levels at the fish study site were significantly higher than those found in adjacent rivers and average conditions in world sediments and soils (Table III).

The fish analysis data are summarized in Table IV. Few differences occurred between the 1985 and the 1986 data set. Prickly sculpins showed slightly higher Zn and Mn values in 1985 than in 1986 and in the whole fish analysis threespine sticklebacks had higher Mn and lower Zn values in 1985 than in 1986. The most significant differences occurred between fish species. Threespine stickleback, redside shiner, large-scale sucker, and northern squawfish had the highest concentrations of Mn, Co, and Zn. The same general trend between species was observed for both the tissue and whole fish samples. In the whole fish analysis threespine sticklebacks were found to have significantly higher Cu, Mn, and Ni values than the other fish species.

There was no evidence that metal concentrations were related to fish size within the range analyzed, but our catches were limited to fairly small specimens. What is more significant is the comparison between the elemental content in the fish from the Sumas River with those from the Lower Fraser River to which the Sumas River is a minor tributary. Figure 3 shows a species by species comparison between our muscle tissue metal levels and those reported by Northcote *et al.* (1975) and Singleton (1983) in the Lower Fraser River, to which the Sumas River is a minor tributary. Significantly higher Mn muscle tissue values were observed in the Sumas samples for cutthroat trout, coho and chum salmon combined, and northern squawfish ($\alpha = 0.05$, Mann Whitney U-Test). No literature data were available for comparing redside shiner and threespine stickleback levels which had the highest Mn values in our study. In contrast there was no difference between our Cu tissue levels and those reported in the literature. Since Cu is not associated with asbestos and since the Cu levels in our sediments were low the results suggest that the asbestos rich sediments have contributed to high Mn levels in three and possibly more fish species.

Of the four trace metals associated with asbestos, Mn and Ni were found to be elevated in our fish samples. Chromium values were at or below the levels reported by Jenkins (1980a) in a literature review on the subject. Because Co levels were near the detection limit and there were little published data on Co levels in fish no further

TABLE III

Average elemental composition of sediments in Sumas River Basin

Variables	Stations along the Sumas River downstream			Literature comparison		
	13 km below asbestos input (Station #1)	International border station 16 km below input (Fish sampling site)	31 km below input (15 km above Fraser River Confluence) (Station #4)	Data from lower Fraser estuary (Hall, 1985; Stancil, 1980)	$\overline{X}$ suspended sediment data from major world rivers (Salomons and Forstner, 1984)	$\overline{X}$ elemental soil constituents (Ure and Berrow, 1982)
Mn (μg g^{-1})	1162	1550	1032	333–654	770	760
Ni (μg g^{-1})	2008	486	232	34– 96	52	34
Cr (μg g^{-1})	517	196	253		72	84
Co (μg g^{-1})	92	42	14	9– 26	14	12
Cu (μg g^{-1})	8	34	58	19–124	33	26
Zn (μg g^{-1})	51	134	135	33–140	95	60

TABLE IV

Elemental analysis of muscle tissue and whole fish samples

Species	Mean and range	Ca	Mg	Fe	Cu	Zn	Mn	Co	Cr	Ni	# of composite samples
Epaxial muscle tissue (total elemental concentration in $\mu g\ g^{-1}$ wet weight)											
Coho	$\bar{X}$	366	306	11.7	0.30	2.82	0.49	0.05	0.21	0.09	3
Salmon	R	334–388	267–348	9.8–15.4	0.08–0.50	0.10–4.82	0.26–0.62	0.04–0.06	0.14–0.28	0.01–0.14	
Prickly	$\bar{X}$	385	295	5.1	0.32	3.85	0.33	0.03	0.09	0.13	6
Sculpin	R	128–579	265–323	3.7–7.3	0.05–0.49	0.70–6.65	0.10–0.57	0.02–0.04	0.06–0.11	0.04–0.38	
Northern	$\bar{X}$	977	327	6.4	0.48	8.54	0.68	0.03	0.12	0.08	3
Squawfish	R	556–1318	300–364	4.5–9.0	0.25–0.66	8.54–9.02	0.30–0.97	0.01–0.05	0.09–0.15	0.06–0.10	
Redside	$\bar{X}$	725	351	6.1	0.35	8.56	0.78	0.04	0.09	0.20	2
Shiner	R	556–894	313–389	3.9–8.3	0.01–0.69	7.53–9.58	0.52–1.05	0.03–0.05	0.05–0.14	0.05–0.36	
Threespine Stickleback	$\bar{X}$	581	289	4.8	0.07	0.90	2.06	0.07	0.07	0.15	1
Cutthroat	$\bar{X}$	378	274	6.6	0.33	5.70	0.51	0.09	0.09	0.14	2
Trout	R	331–425	271–277	6.1–7.1	0.30–0.35	3.90–7.50	0.46–0.57	0.09–0.10	0.09–0.10	0.10–0.19	
Whole fish analysis (total elemental concentration in $\mu g\ g^{-1}$ wet weight)											
Coho	$\bar{X}$	1986	306	39.3	0.77	21.3	2.79	0.08	0.22	0.37	5
Salmon	R	1603–2196	198–349	27.5–62.8	0.57–0.88	10.5–27.7	1.37–3.12	0.06–0.10	0.14–0.44	0.26–0.65	
Prickly	$\bar{X}$	5659	371	49.2	0.88	15.1	7.31	0.12	0.23	0.84	2
Sculpin	R	4970–6348	302–441	34.6–63.8	0.86–0.90	11.8–18.3	5.62–9.00	0.08–0.15	0.18–0.29	0.72–0.97	
Northern Squawfish	$\bar{X}$	4617	343	26.8	1.13	32.3	4.23	0.04	0.16	0.43	1
Redside Shiner	$\bar{X}$	5214	384	44.8	0.89	66.0	6.46	0.18	0.17	0.41	1
Threespine	$\bar{X}$	6233	473	75.8	1.59	37.3	19.61	0.22	0.27	1.16	11
Stickleback	R	4746–7117	384–553	59.0–105.0	1.15–2.40	20.7–47.4	14.00–29.9	0.16–0.31	0.20–0.38	0.95–1.66	
Large-scale Sucker	$\bar{X}$	6088	511	60.0	0.72	24.6	5.93	0.59	0.30	0.86	1

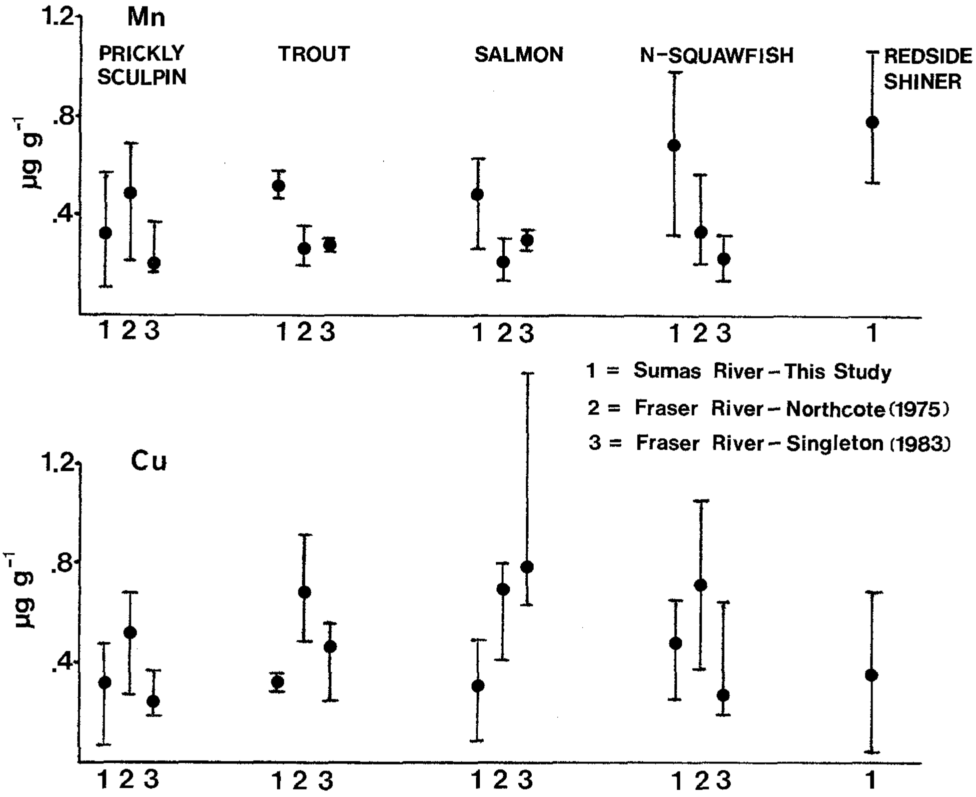

Fig. 3. Differences in Mn and Cu content in Sumas and Fraser River fish muscle tissue (in $\mu g\, g^{-1}$ wet weight).

comparisons were made for these latter two metals. Unfortunately literature on Ni and Mn concentration in fish is scarce so we were unable to make a direct comparison between our fish species and data from the same species elsewhere. In addition there is no consensus on whether we should analyze muscle tissue, liver, kidney, or whole fish samples in order to document metal accumulation (Hakanson, 1984). There is no consensus in the literature as to what causes high metal levels in fish. Metal affinity of tissues is very specific and is dependent on metal chemistry, physiology of organisms, diet, and habitat (Ney and Van Hassel, 1983). Metal level in liver is often considered a good index of accumulation but as shown by Forstner and Wittman (1979), Hakanson (1984), and Hutchinson *et al.* (1975), this varies between fish species and metals being considered. Because of the small size of our fish we were unable to obtain sufficient samples for liver analysis. As a consequence our comparison in Figure 4 shows the Mn and Ni levels in muscle tissue, whole fish, and selected organs as found in the available literature and includes a wide range of freshwater fish species.

The Mn levels in muscle tissue of threespine stickleback and redside shiners were found to be higher than Mn levels in other fish studies. This is also confirmed when we

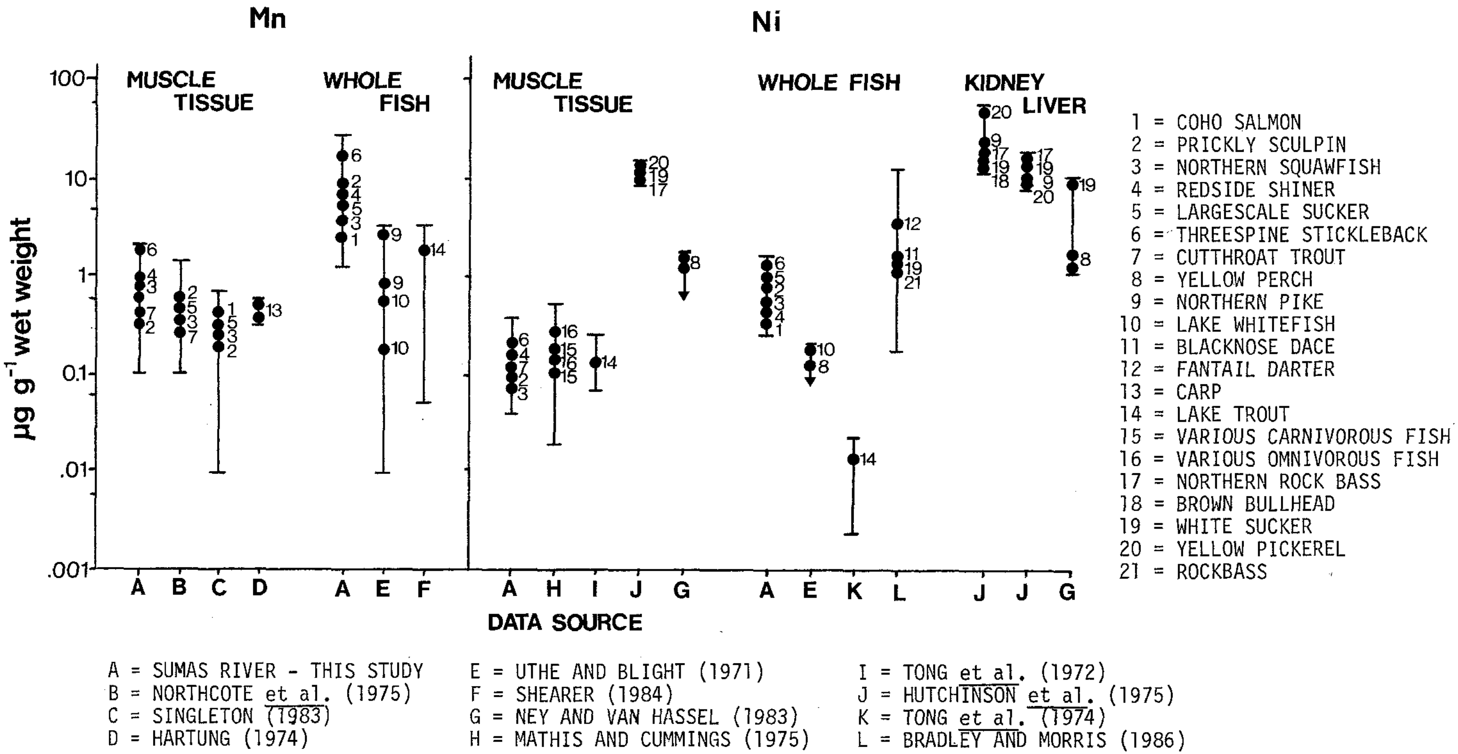

Fig. 4. Comparison of Mn and Ni concentrations in Sumas River fish and fish from literature sources (values in references G and L were converted from μg g^{-1} dry weight to μg g^{-1} wet weight assuming 80% moisture content).

compare our whole fish analysis results with those reported by Uthe and Blight (1971) and Shearer (1984). Although Mn is not one of the most toxic metals in fish, Srivastava and Agrawal (1983) have shown that fish exposure to Mn levels of 900 $\mu g\ L^{-1}$ results in liver problems, changes in fish testis and 30% mortality. Such concentrations are at least one order of magnitude higher than the dissolved Mn levels in the Sumas River water at the study site. Nevertheless, Mn levels in the sediments are in the 1500 $\mu g\ g^{-1}$ range and depending on the form and availability of the metal there is a need for further investigation.

The Ni levels in muscle tissue were found to be comparable to other literature sources (Jenkins, 1980b) and there is no significant difference between our values and those reported by Mathis and Cummings (1973) and Tong *et al.* (1972). In contrast, the data by Hutchinson *et al.* (1975) which cover the Sudbury Ni-mining region, showed Ni tissue levels that were one order of magnitude higher than those found in our study. These authors did in fact report the highest Ni levels in fish kidney and these remain the highest reported Ni levels in the literature. If we compared the whole fish analysis data for Ni it can be seen that overall Ni levels in our study were higher than those reported by Uthe and Blight (1971), and Tong *et al.* (1974) but they do not reach the levels reported by Hutchinson *et al.* (1975) and Ney and Van Hassel (1983). In these latter cases Ni contamination from pollution sources was significantly higher in the sediment than in our study.

5. Conclusions

There is some concern that the presence of asbestos fibers in stream water might have a detrimental effect on the aquatic environment. The fibers are potentially an irritant to fish and some evidence by Belanger *et al.* (1986) suggests that asbestos causes detrimental genetic effects on fish and clams. Asbestos becomes unstable in $pH < 8.0$ and in the process trace metals that are associated with the asbestos fibers are released into the water and sediments. We have documented that this process is taking place in the Sumas River in British Columbia where a landslide introduces large quantities of chrysotile asbestos. No evidence was observed that the fish are adversely affected by the material except that there is a general increase in Mn and Ni concentration in the whole fish samples. The Mn values in muscle tissues of cutthroat trout, salmon, redside shiners, and threespine stickleback were significantly higher than those found in background samples in adjacent streams. Muscle tissue samples do not seem to be diagnostic of Ni accumulation. Our study was limited to small fish and a more detailed study which includes large fish and examination of possible behavioral interactions is needed to observe long term effects.

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