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By:

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EFFECTS OF ABANDONED GOLD MINE TAILINGS ON THE ARSENIC CONCENTRATIONS IN WATER AND SEDIMENTS OF JACK OF CLUBS LAKE, B.C.

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ABSTRACT

From 1933 to 1964 gold was extracted by underground mining at the northeast shore of Jack of Clubs Lake. At present, tailings and waste rock 4.5 m thick covers approximately 25 hectares of land adjacent to the lake. Arsenic concentrations (>2,000 $\mu g.g^{-1}$) were found in the tailing materials. Two simultaneous processes are controlling the elevated As concentrations in the lake sediments (up to 1,104 $\mu g.g^{-1}$), deposition of suspended particles enriched with As and subsequent diagenetic cycling of sedimentary As. Arsenic concentrations (as high as 556 $\mu g.L^{-1}$) in water samples adjacent to the tailings indicate mobilization of As. However, dissolved As is readily scavenged or coprecipitated with iron hydroxides. The greatest proportion of As in the sediment cores is associated with iron oxides and sulphides. Under oxic conditions the high concentrations of Fe in the tailings is actually beneficial and very effective at limiting the migration of undesirable elements.

Keywords: arsenic, lakes, mining, water, sediments.

INTRODUCTION

Gold mining has been a major activity in Canada for over a century. Since 1921 Canada has occupied second or third place among the world's gold producing nations (Cranstone, 1985). Like many other industries, gold mining has generated wastes, such as tailings and waste rock, which contain some naturally occurring and potentially toxic elements found in the metal ore, and toxic elements and compounds introduced during the various stages during the extraction of the metals from the ore. Arsenic is widely diffused in nature and occurs in many types of gold ore deposits. The principal As-minerals are arsenopyrite, niccolite, cobaltite, tennantite, enargite, orpiment and realgar (Boyle and Jonasson, 1973).

Arsenic used to be extracted as a byproduct in many gold mines and sold mainly for the manufacture of pesticides (Nriagu and Azcue, 1990). However, the ban of using As-pesticides on food crops diminished considerably the demand for As. Since then, the extraction of As was not profitable, and large quantities of As had been abandoned in the gold mine tailings and waste rock. The mobility and potential toxic effects of As has been reported in gold mining areas in Canada (Azcue and Nriagu, 1993; Mudroch and Clair, 1986), Montana, USA (Moore *et al.*, 1991), Ghana (Manful and Verloo, 1989), Brazil (Malm *et al.*, 1989) and South Africa (Forstner and Wittmann, 1976). At present this problem has been recognized in the majority of active gold mines. However, there are many abandoned gold mine wastes containing large quantities of As. One of such abandoned gold mining sites is located at Wells in the Cariboo region in British Columbia (Canada).

The Cariboo region has been an important mining area since the 1800's. Early in 1861 dozens of gold-bearing creeks were discovered and by 1960 most of the creeks in the Cariboo region had been prospected. The town of Wells (Figure 1), at the northeast shore of Jack of Clubs Lake (JCL) was built on the tailings from the old Lowhee Mine. Starting in 1933 the Gold Quartz Mining Company extracted gold by underground mining rather than panning, dredging, and sluicing until 1964. The mine produced in excess of five million dollars worth of gold (Harris, 1984; Taylor, 1978).

The results presented in this study are a part of a multidisciplinary investigation of the biological and geochemical effects of the abandoned gold-mine tailings in JCL and their impact on the Fraser River system (Mudroch *et al.*, 1993). The main objectives of this study were: 1) to determine the processes affecting the distribution of arsenic in water, suspended and bottom sediments, and sediment interstitial water of JCL, and 2) to evaluate the mobility of arsenic in the gold mine wastes abandoned for about 30 years.

MATERIALS AND METHODS

Study Area

Jack of Clubs Lake (JCL) is located in the Barkerville Terrane, which is mostly underlain by clastic sedimentary rocks, principally Precambrian and Palaeozoic sandstone with minor proportions of limestone and mafic volcanic rocks (Struik, 1988). The tailings from the milling and gold extraction were discharged into the northeast end of JCL, changing the original morphometry of the lake. At present, tailings deposit about 4.5 m thick covers approximately 25 hectares of land adjacent to the lake. The tailings are dissected by Highway 26, Lowhee Creek and Willow River (Figure 1).

Jack of Clubs Lake is 2.4 km long and 0.5 km wide, with a mean depth of 19 m and a maximum depth of 63 m. Its flushing rate is extremely rapid, averaging 0.8 years. Lowhee Creek flows through the tailings before it empties into the northeast end of JCL. During spring runoff the waters of the creek flood an extensive area of the tailings. Most of the sediments that have accumulated near the mouth of the creek are remnants of extensive hydraulic mining activities that occurred during the gold rush (Andrews, 1989). A man-made channel dug through the tailings at the northeast end of the lake drains into the Willow River, the only outlet of the lake, which flows for 130 km before discharging into the Fraser River. Part of the water flowing through this channel originates as groundwater seepage from the tailings and does not reach the lake.

Bowron Lake located inside the Bowron Lake Provincial Park about 30 km east of Wells, was selected as a control or reference lake for the study because there are no reports of mining activities, past or present, along the lake. The bedrock in the watershed of Bowron Lake consists mainly of quartzite, limestone, shale, phyllite, dolomite, and conglomerates.

Sampling and Analytical Methods

Water Samples

The field work was carried out during the first week of August, 1992. Water samples were collected 1 m below the surface and 1 m above the lake bottom, at each of the 10 stations in the lake using a van Dorn bottle (Rosa *et al.*, 1991). Surface water samples from streams and seepages (S1 to S7 in Figure 1) were also collected. The water samples were filtered in the field within a few hours of collection using a Millipore glass filter apparatus with 0.45 μm cellulose acetate filters. Samples were acidified with Ultrex grade HNO_3 to a final concentration of 0.4% and stored at 4°C until analyzed at the Geological Survey of Canada (Finch *et al.*, 1992). At all sites continuous surface-to-bottom depth profiles of pH, specific conductance, dissolved oxygen, and temperature were recorded by a HYDROLAB Profiling System, Model DataSonde$_R$ 3.

Sediment core sections 0 to 5, and 5 to 10 cm were extruded from the core liner under an atmosphere of nitrogen and squeezed separately to press out the interstitial water (as described by Kalil and Goldhaber, 1973). The time between sample collection and termination of squeezing was less than 5 hours. Groundwater was sampled by means of piezometers previously inserted into the tailings. First, the water level in each piezometer well was measured and the standing water was pumped out. The piezometers were allowed to recover overnight, and the next day any water which had accumulated in the piezometers during the night was pumped out at a low flow rate, using prewashed polypropylene tubing. Once the water started to flow, the tubing was attached to an on-line 0.45 μm filter and several mL of water were passed through the filter. A flow-through cell was attached for determination of pH.

Arsenic was determined by formation of its hydrides using $NaBH_4$ as a reductant followed by analysis employing quartz tube AAS. Arsenic was reduced with KI and ascorbic acid prior to analysis so that the concentrations reported represent As(III) and As(V). Two controls, NBS 1643c and an in-house reference standard "tap", were included in each batch of samples processed.

Sediments

Lake sediment samples were taken at 10 stations using a miniponar grab sampler. This sampler encompasses an area of 234 cm^2 and penetrates to an average depth of 5 cm. The sediment was placed in a glass tray, homogenized with a plastic spoon and transferred to prewashed plastic vials. Samples were stored at 4°C in the field and freeze-dried when taken back to the laboratory. Tailings samples were separated into six different particle size fractions: 54-140 μm, 40-54 μm, 27-40 μm, 19-27 μm, 13-19 μm, and <13 μm, by the "Sieve and Sedigraph Method", using a Warman Cyclosizer (WR-200) and sieves (LC-10B) (Duncan and LaHaie, 1979). Carbon analyses were performed on freeze-dried samples using a LECO-12 Carbon Analyzer, and loss on ignition was determined by ashing dry sediment at 450°C to constant weight.

A modified Kajak-Brinkhurst corer was used to collect sediment cores to examine vertical profiles of As concentrations in the sediment. For this purpose, sediment cores were subdivided into 1-cm sections and analyzed by AAS following application of the sequential leach scheme outlined in Table 1. It should be borne in mind that these are operationally defined fractions and that some refractory minerals such as spinels, tourmaline, zircon, etc. are not dissolved effectively by this acid digestion, but the sulphides and oxides of interest in this study are. Arsenic was first separated from potential interferents such as Cu and Ni by coprecipitation with $La(OH)_3$, which was formed by adding $La(NO_3)_3$ and NH_4OH to the extract. Two international lake sediment standard reference samples (TILL-2 and LKSD-4) produced by the GSC (Lynch, 1990) were used to testify the precision attained in the analyses. All calibration solutions for measurement by AAS were made up in the same matrix as the analytes in each of the five leaches.

Suspended sediments were recovered from three locations in the study area (C1, C2 and C3 in Figure 1): (1) at the centre of JCL, three meters below the water surface; (2) from the outflow of the lake (the Willow River); and (3) from the Willow River at the end of its passage through the town of Wells About 2,000 litres of water were pumped into a Westfalia separator at a flow rate of 4 L/min to remove the suspended sediments, which were then freeze-dried, weighed, homogenized by grinding, and analyzed (following the techniques of Mudroch and Duncan, 1986).

RESULTS AND DISCUSSION

Water Quality

Primary production, based on the chlorophyll levels in the lake, at least during our sampling period, was extremely low (ranging from <0.1 to 1.1 $\mu g.L^{-1}$). According to other research results (Azcue *et al.*, 1993) sediment microbial activities were found to be suppressed by tailings deposited in the lake. The low primary production can not be attributed to low or limiting nutrients. It is speculated that the primary production is affected by the tailings deposited in the lake. Both JCL and Bowron Lake are thermally stratified with epilimnion and thermocline thickness between 5 to 7 m, and 2 to 4 m, respectively. The remainder of the water column consists of a cold (7-9°C) hypolimnion. In JCL, conductivity in the water is at minimum near the thermocline and increases in the hypolimnion. The greatest difference in conductivity in the water column from surface to bottom was recorded at stations 6, 5, and 4 which are close to the tailings (Figure 1). This increase is due to the increase of SO_4^{2-} (up to 53%) released from the sediment after oxidation of sulfides. pH in surface water ranges from approximately 7.8 at station #4 to 8.3 at station #1. Bottom water has lower pH at station #4 due to SO_4^{2-}

ions. The oxygen concentration is greater than 7.5 mg.L^{-1} at all sampling stations, with a consistent concentration minimum in the thermocline. This minimum is typical in stratified systems and is attributed to decaying organic matter trapped in the thermocline.

The seepage water at site 05 (Figure 1), situated on the tailings just downslope from the abandoned Cariboo Gold Quartz mine operation, was sampled after a rainy night. It contained the greatest As concentrations (556 μg.L^{-1}), as might be suspected from its pH of 2.7. Sample 05 is highly anomalous in other elements such as: S as SO_4 at 640 mg.L^{-1}; Fe at 163 mg.L^{-1} and Pb at 144 μg.L^{-1}. In the rainy season, seepage from this site would reach JCL via the wide gulley running down the south-east side of the tailings. During this collection period, site #16 represents drainage across the tailings into JCL from the general location of site #05 (Figure 1). This seepage water was similarly high in the elements listed above. The major sources of water (and hence possible contaminants) for JCL which were sampled in this project comprise sites: #16 (from across tailings); #08 (Lowhee Creek having traversed tailings); #15 (drainage through culvert of steep slopes north of Hwy 26); #25 (drainage through culvert from steep slopes on north side of Hwy 26); #01 (stream about 1 m wide at JCL with a rapid flow); and #02 (stream about 0.3 m wide at JCL with a good flow) (Figure 1). Heavy iron oxide staining was visible all along Willow River from JCL to its confluence with Williams Creek; tailings banks are also present in this area of north Wells. Site #12 (S5) is a mixture of seepage and drainage from the mineralized slopes on the far side of the road and the tailings closest to the road. It is clear that the influence of leaching from the main tailings bed and from the general area of the Island Mountain Mine on the other side of the road is quickly dissipated. The concentrations of Fe, SO_4 and As values have fallen to about 100 $\mu g.L^{-1}$, 35-40 $mg.L^{-1}$ and 0.5 $\mu g.L^{-1}$, respectively just before the confluence of Willow River and Williams Creek.

Data for these samples are compared in Table 2 with surface and bottom lake samples. Clearly, in terms of absolute concentration, sites 16 and 08 are the most significant. Arsenic concentrations in the lake itself are low and pH is neutral. Arsenic concentrations in station #4, nearest the tailings, is higher than the other lake samples, however, the difference is more significant for other elements like Pb, 0.11 $\mu g.L^{-1}$ in station #4 and <0.02 $\mu g.L^{-1}$ in station #3. While the chemistry of major ions of JCL water is quite different from that of Bowron Lake (e.g. SO_4 10-20 $mg.L^{-1}$ in JCL vs. 2 $mg.L^{-1}$ in Bowron Lake), As concentrations are considerably similar (averaging 0.26 $\mu g.L^{-1}$ in both lakes).

Results of pore water analyses are presented on Table 2. Concentrations of As in the pore water are approximately two orders of magnitude greater than in lake water. The greatest concentrations of As, were found in the pore water collected from the tailings (163 $\mu g.L^{-1}$). The concentrations of dissolved As are not different between the surface and bottom waters of the lake, thus, it suggests that during the stratification period the As is not being regenerated from the sediments. At some sampling stations the concentration in the bottom water are lower which implies that the sediments are acting as a 'sink' rather than a 'source'. Arsenic is effectively removed from solution as the pH increases and Fe and Mn oxyhydroxides are precipitated. According to these preliminary results, JCL seems to have been only partially impacted by contaminants towards the north end of the lake.

Sediment Quality

The concentrations of total carbon in bottom sediments from JCL ranged from 1.4 to 4.3%. Concentrations of organic carbon in the sediments at station #1 at the southwest end of the lake reached values as high as 4%. Adjacent marshes on the shore most likely contributed organic matter to the bottom sediments in this part of the lake. The lowest concentrations of organic carbon were found in the sediments at station #5, which appeared to be in the area most strongly affected by the tailings on the northeast shore of the lake. The concentrations of inorganic carbon, which probably represents calcite in the sediments, were relatively uniform at all sampling stations with the exception of station #5 where it was about three times greater.

The greatest concentrations of As (up to 1,104 $\mu g.g^{-1}$) were found in the sediments at station #5. The results showed that the northeast end of the lake was the most severely contaminated area and that this area extends about 500 m towards the centre of the lake. The concentrations of As in the tailing materials ranged from

903 to 2,485 $\mu g.g^{-1}$). It is well known that the metal concentrations in the different particle size fractions of the sediments are very important in the evaluation of the transport of metals (Forstner, 1982; Mudroch and Duncan, 1986). Figure 2 shows the concentrations of As and Pb in six particle size fractions of sediments collected in the tailings. The greatest concentration of Pb (3,470 $\mu g.g^{-1}$) was found in the <13 μm size fraction, with decreasing concentrations with increasing size fractions. Consequently, large quantities of Pb bound to the fine silt and clay fractions (<13 μm particle size) may be mobilized by erosion of the fine particles and physico-chemical changes of environmental conditions at the site where the fine-grained particles settle. On the other hand, As concentration was greatest (>2,000 $\mu g.g^{-1}$) in the 45-53 μm size fraction, with decreasing concentrations in the smaller particle size fractions. Thus, As preferentially bound to sediment particles (i.e., silt) tends to be less mobilized by erosional processes.

The greatest concentrations of As (1,216 $\mu g.g^{-1}$) in the suspended sediments were found in the samples collected from the Willow River at the outflow from JCL and decreased with increasing distance from the lake. The concentrations of As (245 $\mu g.g^{-1}$) in the suspended sediments in the middle of the lake (station #8) were similar to those in the bottom sediments (277 $\mu g.g^{-1}$) from the same area in the lake. The elevated levels of As in the tailing materials and in the suspended matter at the outflow of the lake confirmed that under strong erosion conditions (rainy season) particulate-As could be the main pathway of transport of As out of the tailings.

The variation in elemental concentration with depth in the five phases extracted from sediment cores from JCL (station #10) and Bowron Lake are shown in Figure 3. Core 10 was collected at a water depth of ca. 40 m about 1100 m from the tailings, and presumably during spring run-off and times of flooding would receive both material flushed mechanically from the tailings and water carrying high contents of dissolved salts. There is a large difference in the magnitude of As concentrations between JCL and Bowron Lake. In sediment cores from Bowron Lake, As is in the range of 12-23 $\mu g.g^{-1}$. While in JCL cores As values lie in the ranges of 93-574 $\mu g.g^{-1}$. By far the dominant phases of As in Bowron Lake sediment cores are phases 2 and 3 where amorphous Fe and Mn (oxides and oxyhydroxides) and crystalline Fe are preferentially extracted. However, As in JCL is held mainly in phases 3 and 4 (largely sulphides), with 2 being the next most significant. These profiles follow closely those for Fe, with marked depletion at the sediment-water interface. These observations are consistent with the theory that As cycling in freshwater is mainly controlled by coprecipitation with hydrous oxides in the water column and further reduction in the deep sediments (Aggett and O'Brien, 1985); Belzile and Tessier, 1990; Peterson and Carpenter, 1986).

Our interpretation of these data must, of course, be regarded as tentative owing to the absence of information on subjects such as sediment stratigraphy, the hydrological history of JCL, and dates of core horizons. A peak in the concentrations of As at the 4 to 6 cm depth in the sediment collected at station #10 in JCL, most likely reflects the inputs of this element from past gold mining activities at the Cariboo Gold mine around 1940. This assumes the annual sedimentation rate of 1 to 2 mm in the lake. The greatest proportion of As in the sediment cores is associated with iron oxides and sulphides, implying mechanical dispersion from the tailings. Arsenic in true solution appears to be readily scavenged by amorphous iron hydroxides in the lake sediment or by coprecipitation with iron hydroxide on surface media (rocks, tailings themselves) which is prevalent in the area surrounding JCL. Therefore, the high concentration of Fe in the tailings originating from pyrite (associated with Au) is actually beneficial and very effective at limiting the migration of undesirable elements. However, under anoxic conditions, large quantities of As associated with Fe oxyhydroxides may be released into the hypolimnion.

Figure 2 shows the historical changes of the percentages of As bound to different chemical fractions of sediments from JCL (station #10) and Bowron Lake. The significant changes in the As associated with Fe hydroxides and sulphides at the water-sediment interface, reflect the sensitivity of As to changes in the redox conditions. On the other hand the results from Bowron Lake support the assumption that As is less mobile in natural than in polluted lakes (Reuther, 1992). The forms of As and their distribution sequence in JCL are generally consistent with observations in other polluted sediments (Azcue and Nriagu, 1993; Moore *et al.*, 1988).

Conclusions

Based on the previous results several conclusions can be made, bearing in mind, that these results were collected during the summer period. Consequently, for a more precise interpretation seasonal data would be required. The concentrations of As in several water samples indicate the present mobilization of As at low pH, for example, 556 $\mu g.L^{-1}$ of As at pH 2.7 in the seep near the old mine. This indicates that the northeast part of JCL has been impacted by the tailings deposited on the shore to much greater extent than the southwestern part of the lake. The concentration profiles of As in the sediments in JCL suggest that two simultaneous processes are controlling the enrich surficial As concentrations, first mechanical and hydromorphic dispersion of As from its source, such as arsenopyrite, in the tailings, followed by a diagenetic cycling of As near the water-sediment interface.

The concentrations of As in sediment pore water in JCL was about two orders of magnitude greater than that in the lake water. However, there is no migration of As from the sediments into the overlying water, as evidenced by equal concentrations of As in the surface and bottom water in the lake. This indicates that under oxic conditions the bottom sediments act as a sink rather than a source of As. On the other hand, relatively great concentrations of As in the pore water may affect the health of the sediment biota.

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Table 1. Sequential extraction scheme

Extractant	**Phase dissolved**
1M NaOAc. HOAc pH5 [1g/20 ml; 2 x 6 h constant shaking] (two 6h extractions; app. 2d)	1-Carbonates, adsorbed metals, exchangeable metals
0.25 M $NH_2OH.HCl$ in 0.25M HCl [At 60°C; first leach for 2 h, second leach for 0.5 h] (2 and 0.5 hour extractions; approx. half a day)	2-Amorphous oxyhydrates of Fe and Mn. Hydroxylamine hydrochloride is preferred to oxalate reagent as its attack on organic matter is minimal.
1 M $NH_2OH.HCl$ in 25% HOAc [At 90°C; first leach for 3 h, second leach for 1 1/2 h] (3 and 1.5 hours extractions; approx. one day)	3-Crystalline Fe oxides (e.g. hematite, goethite, magnetite, maghemite, lepidocrocite)
$KClO_3/HCl$ for 0.5 h followed by 4 M HNO_3 at 90°C (30 and 20 min; approx. half a day)	4-Sulphides, organic matter
$HF/HClO_4/HNO_3/HCl$ evaporation (approx. 1 day)	5-Silicates, residual crystalline fraction

Table 2. Arsenic concentrations in water ($\mu g.L^{-1}$) and solid phase ($\mu g.g^{-1}$) [Detection limit = 0.2 $\mu g.L^{-1}$].

	Average	n	s.d.	Maximum	Minimum
Jack of Clubs Lake:					
Dissolved water:					
JCL - surface	0.25	22	0.09	0.30	<0.2
JCL - bottom	0.30	15	0.07	0.41	<0.2
Streams	17.5	41	93	556	<0.2
Pore water:					
0-5 cm	76	8	13	91	78
5-10 cm	158	10	107	361	51
tailings	163	12	57	281	102
Bottom sediments					
Jack of Clubs Lake	342	10	296	1,104 (JCL #5)	80 (JCL #9)
Tailings	1529	9	696	2,485	903
Suspended matter					
JCL- station #8	245	3	26	283	224
Willow R. (outflow)	1216	3	76	1311	1124
Willow R.(downstream)	343	3	43	394	289
Bowron Lake:					
Dissolved water	0.26	6	0.14	0.42	<0.2
Pore water	22	4	2.0	24	21
Bottom sediments	19	6	3.7	23	16

Figure 1 "In drafting

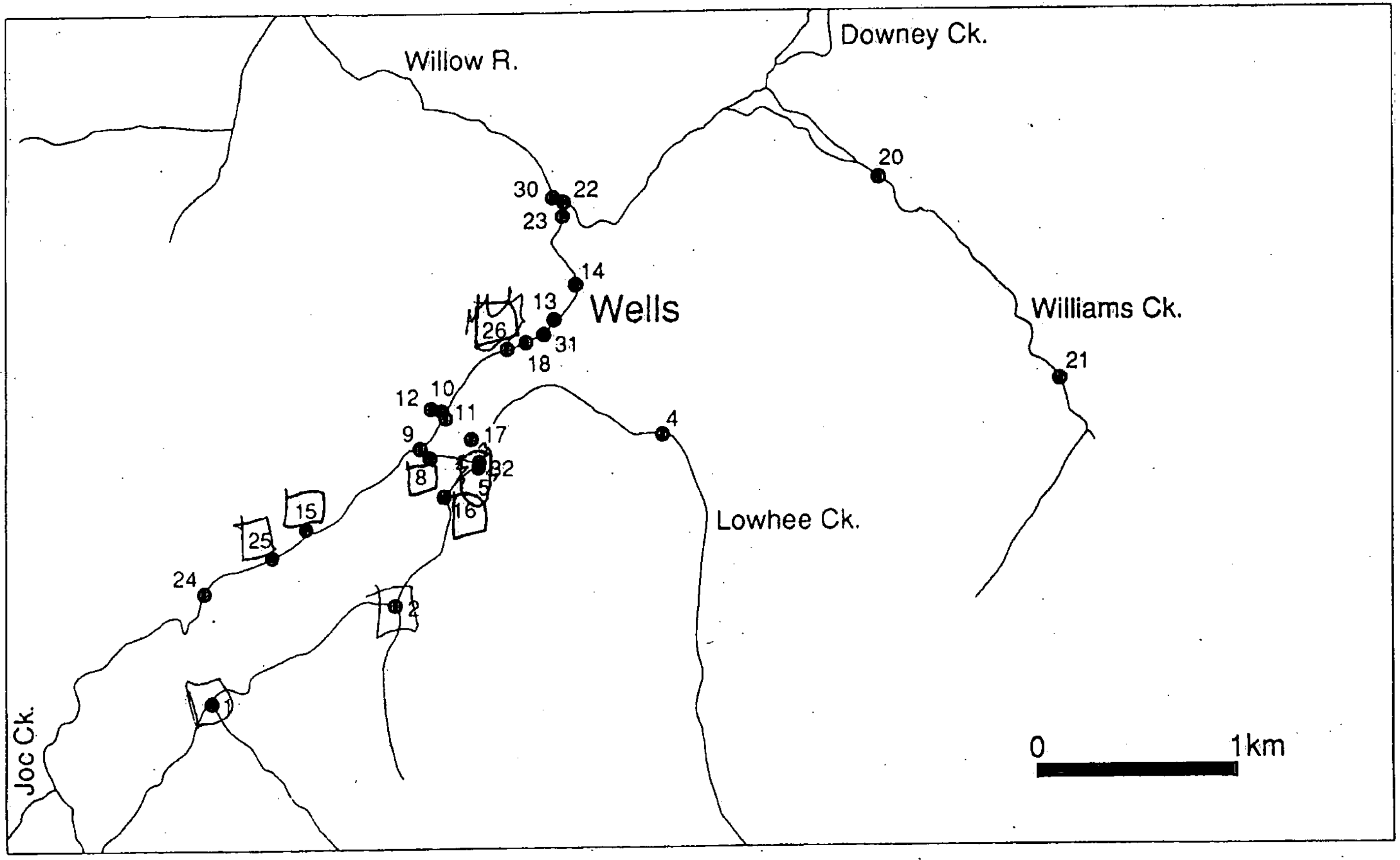
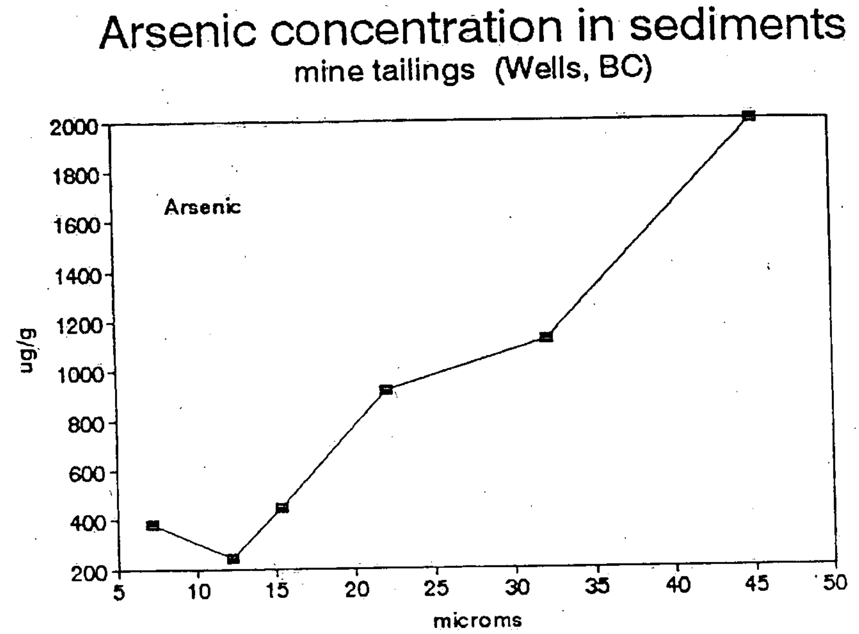


Figure ~~2.2~~ 1. Site locations for stream and seepage samples

Figure 1 ⟹ General Map + Study area with sampling stations

Figure ~~3.18~~ 2. Concentrations of As and Pb in six particle size fractions of the tailings separated by a cyclosizer



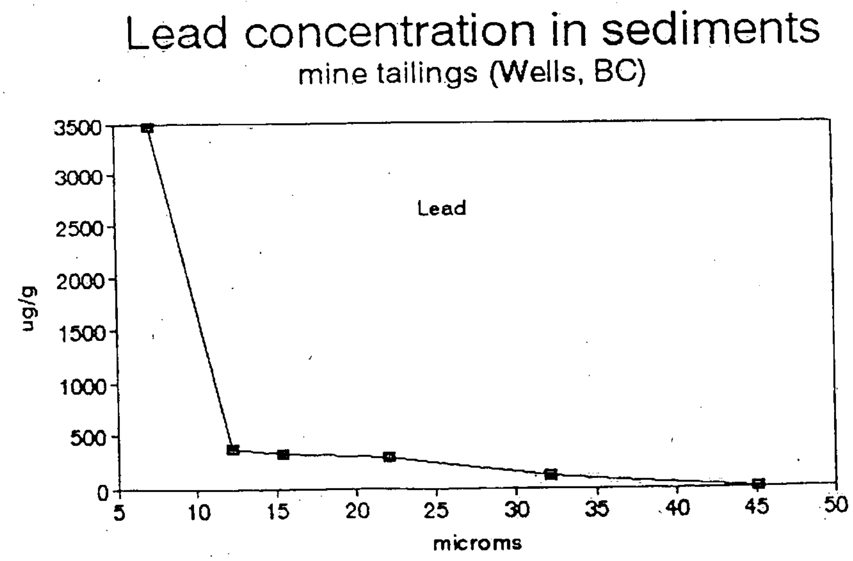
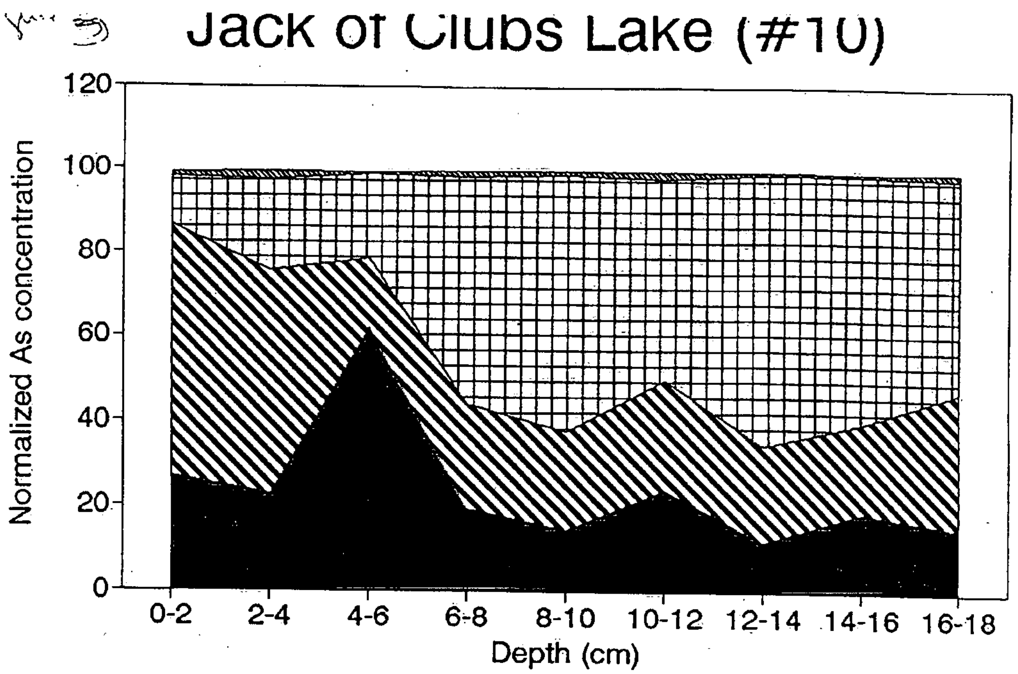
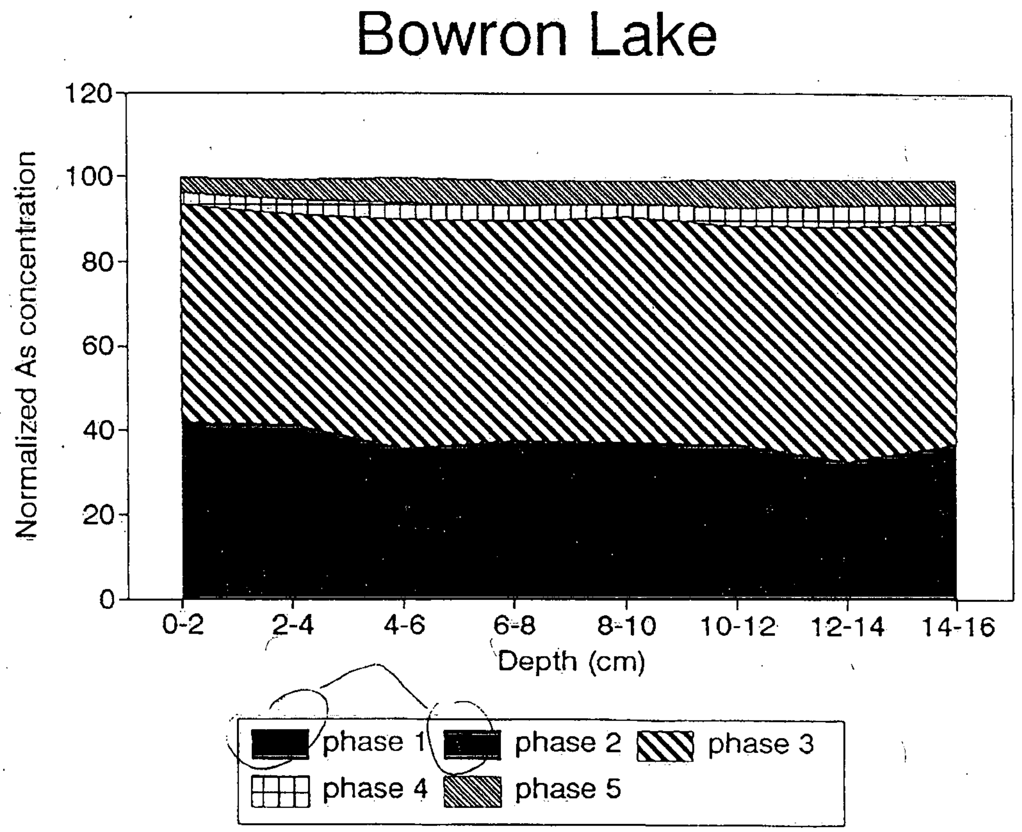


Figure in drafts

Figure 3


Jack of Clubs Lake (#10)


Bowron Lake

Legend explain phases

Figure drafts

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