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Acid mine drainage at the Holden deposit, Chelan County, Washington

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INTRODUCTION

The Holden orebody is a relatively low-grade, copper-zinc-silver-gold volcanogenic massive sulfide deposit (VMS) located within the crystalline core of the northern Cascade Range, north-central Washington (see fig. 1) (Nold, 1983; Dragovich and Derkey, 1994). The mine, which began production in 1938, ceased operations in 1957 because of high production costs and a drop in the copper market. While operative, over 212 million pounds of copper, 40 million pounds of zinc, 2 million ounces of silver, and 600 thousand ounces of gold were extracted from 10 million tons of ore (McWilliams, 1958). At present, the mine site consists largely of mill tailings, waste dumps, portal and shafts, dismantled mill, and other abandoned surface structures. Because of the availability of large amounts of pyrite and other sulfide minerals, massive sulfide deposits (such as Holden) have an unusually high acid-generating capacity that allows waters draining this deposit type to dissolve and transport exceedingly high concentrations of metals (Smith and others, 1994). Such discharge represents potential sources of acid and metal-loading in the environment that could adversely impact not only local water quality, but biota as well. (Plumlee and Edelmann, 1995).

The U.S. Geological Survey, in cooperation with the U.S. Forest Service, is currently conducting geochemical studies at the Holden mine as part of an integrated mineral-resource and mineral-environmental assessment of Wenatchee National Forest. Prior to the geochemical investigation, the Forest Service initiated a reclamation-restoration program to remedy obvious environmental concerns associated with the mine complex. Reclamation work focused on reestablishing landscapes and environments that would mirror the surrounding undisturbed terrain. This included stabilizing the tailings piles, rechanneling stream and seasonal runoff, and establishing specialized nurseries to explore revegetation possibilities. These efforts, however, did not address the problem of acid mine drainage. The Holden study was therefore designed, in the context of reconnaissance sampling, to rapidly identify, at a relatively low cost, possible contaminating effects of waters draining sulfide-rich mill tailings, underground workings, and unmined ore zones at the inactive mine site.

LOCAL GEOLOGY AND MINERALIZATION

The Holden mine lies within the crystalline core of the northern Cascade Range, a region characterized by three major northwest-trending tectonostratigraphic terranes: the Nason, the Swakane, and the Chelan Mountains. The Holden orebody is located within the Chelan Block of the Chelan Mountains terrane (Dragovitch and Derkey, 1994). The most detailed account of the local geology and a correlation of the rock units is given in Cater and Crowder (1967).

The Holden deposit is strata-bound within a sequence of steeply dipping, high-grade metamorphic rocks, largely consisting of calc-silicate gneisses and amphibolites of probable late Paleozoic age. These rocks are more or less andesitic in chemical
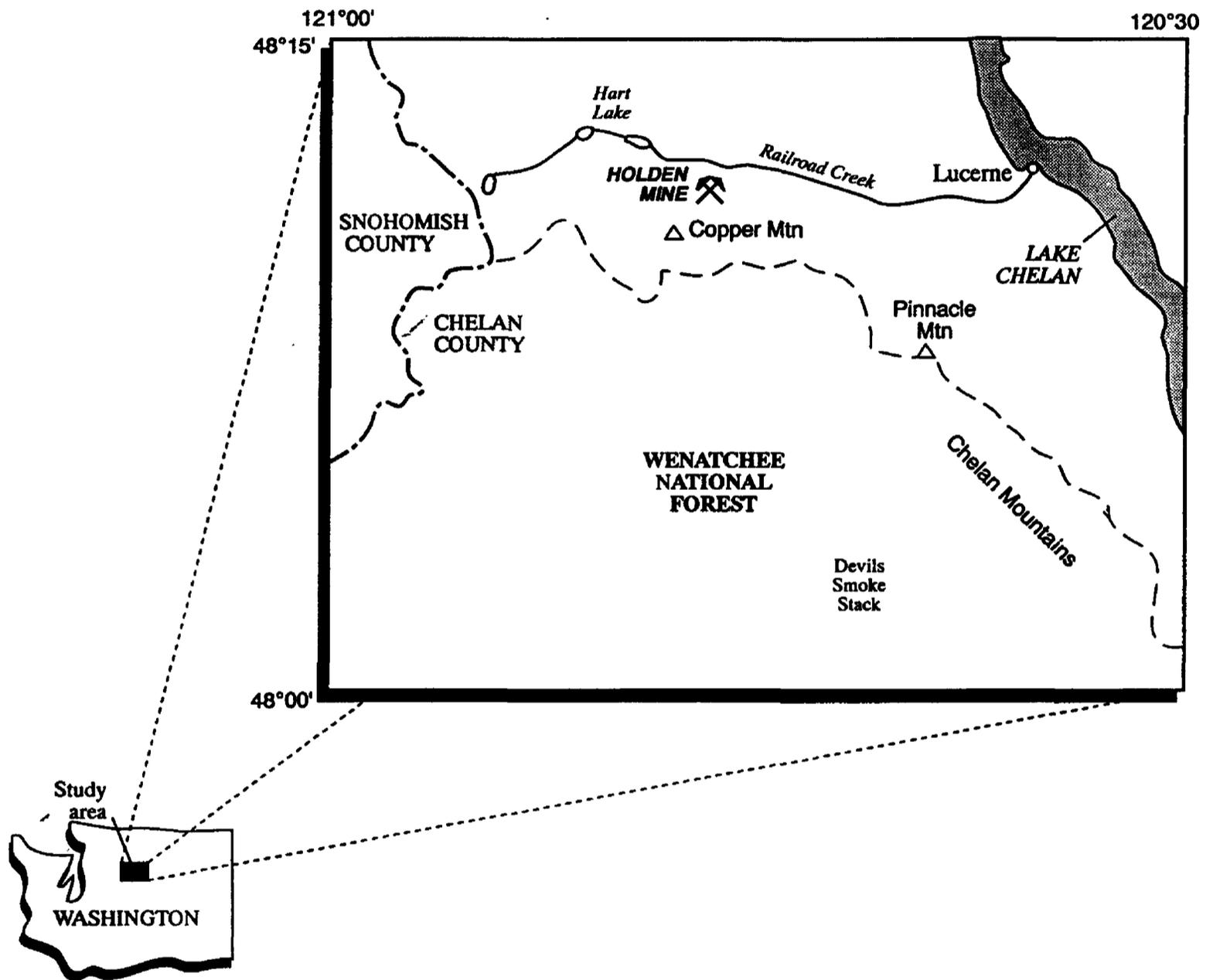
Figure 1. Location of the Holden mine, Chelan County, Washington

make-up, suggesting andesitic volcanics and related volcaniclastic sedimentary rocks as protoliths. The orebody is hosted in a pyritic sericite-quartz schist sequence within the high-grade metamorphic rocks. Exposures of the ore-bearing schist, which ranges from 300 to 600 ft in thickness, can be traced for at least three miles on the surface (Nold, 1983). The deposit is strataform within the schist unit and displays the well-developed metal zoning characteristic of VMS deposits, with a lower or original copper-rich footwall and a zinc-rich upper or original hanging wall. This primary metal zoning has endured the ravages of high-grade metamorphism as well as intense deformation, which has overturned the deposit, resulting in a copper-rich zone structurally above the zinc-rich zone (Nold, 1983). The most prominent ore and ore-related minerals include pyrite, pyrrhotite, chalcopyrite, sphalerite, and native gold. Galena, magnetite, and molybdenite are also present but merely considered accessories. The gold is mostly linked to chalcopyrite in the structural hanging wall, while silver is associated with sphalerite in the structural footwall (McWilliams, 1958). Mineralization is for the most part disseminated, although massive ores are found locally in the zinc-rich zone (Nold, 1983).

FIELD AND LABORATORY METHODS

Sample media for this investigation included waters, effluent related secondary sulfate salts, Fe-oxide and hydrous metal oxide precipitates, and mill tailings.

Water samples were collected from within, above, and below the mine workings in polypropylene bottles previously rinsed with 10% HNO_3. At each site a 60-mL sample, filtered through a 0.45-μm filter and acidified with concentrated HNO_3 to pH <2, as well as an untreated 250 mL sample were collected. Selected anions were determined on the untreated samples by the ion chromatography method of Fishman and Pyen (1979). Major, minor, and trace element contents of the filtered and acidified samples were determined by an unpublished inductively coupled plasma-mass spectrometry (ICP-MS) method (A.L. Meier, written commun. 1994). In addition, water temperature, pH, and conductivity were measured at each sample locality.

Three types of precipitates were collected at the Holden site. These include: (1) a thick flocculent, milky-white precipitate formed in waters that drain the mine portal; (2) salts that have precipitated and formed a thin crusty veneer at the base of the mill tailings; and (3) highly oxidized precipitates that are associated with acidic waters or seepage at the foot of the mill tailings. The flocculent material was collected by simply spooning (plastic spoon) the substance into a 250-mL polypropylene bottle. Salts and oxidized precipitates were gathered from various locales at the base of the tailings with the aid of a plastic knife and stored in plastic zip-lock bags. All samples were analyzed by inductively coupled plasma-atomic emission spectrometry (ICP-AES) using the 40-element procedure of

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Briggs (1990) and the 10-element procedure of Motooka (1990). In addition, conventional x-ray powder diffraction (XRD) techniques (Klug and Alexander, 1974) were employed to determine the mineralogy of the precipitate samples.

A single grab sample of shallow mill tailing material was collected to determine its chemical properties and heavy-metal content. The sample was analyzed by the ICP-AES methods of Briggs (1990) and Motooka (1990), as well as for low-level gold by the graphite furnace atomic absorption spectrophotometry (GF-AAS) method of O'Leary and Meier (1990).

A complete listing of the analytical results, sample locality maps, and description of the analytical techniques are given in Kilburn and others (1994).

RESULTS AND DISCUSSION

Although findings of this study are clearly inhibited by the limitations commonly attached to reconnaissance investigations, sufficient data has been gathered to make some pertinent observations. Two major sources of contaminated water flowing directly into nearby Railroad Creek were identified. The first consists of a series of highly acidic metal-rich seeps located at the base of the mill tailings. Water from these seeps issues directly into Railroad Creek or accumulates in numerous standing pools at the foot of the tailing impoundment. The other is acidic, metal-rich effluent waters emerging from the mine portal. The effluent waters exiting the portal flow downhill almost a quarter of a mile before discharging into Railroad Creek, some 200 ft below the mine entrance. In addition to acid drainage, a less conspicuous source of contamination shows up at the base of the mill tailings in the form of various soluble efflorescent salts derived from sulfide weathering and related acid-generation. These encrustations or secondary sulfate salts often host exceedingly high concentrations of metals and acid in solid form, which, during periods of rain or seasonal snowmelt, readily dissolve, releasing their potential toxic constituents to the environment.

SEEP WATER

The active seep water collected at the base of the mill tailings had a pH of 2.9, conductivity of 1250 µS/cm, and contained 690 mg/L SO_4 , as well as concentrations (in µg/L) of >6,000 Al, 6.8 Cd, 53 Cu, 50,000 Fe, and 3,800 Zn. Quiescent or inactive seep water collected from a standing pool also at the foot of the tailings had a pH of 2.8, conductivity of 1110 µS/cm, and contained 456 mg/L SO_4. Dissolved metals included (in µg/L) 1,500 Al, 21 Cu, 23,000 Fe, and 130 Zn (Kilburn and others, 1994). Applying the pH-metal classification scheme of Ficklin and others (1992), whereby drainage waters can be classified according to pH and the sum of aqueous or dissolved base metals (Cu+Zn+Cd+Co+Ni+Pb), the active seep discharge can be interpretated as a high acid-high metal solution, whereas the dormant pool water can be regarded as high acid-low metal. Its surprising that both water samples failed to reflect the extreme

metal chemistries anticipated, considering the nature of the deposit and placement of the samples (base of the tailings). Of particular interest is the comparatively low concentrations of Cu and Zn carried in solution. The geologic, geochemical, and external controls for high acid-extreme metal drainage discussed by Plumlee and others (1992;1993) are clearly in place at the Holden deposit, where mining of massive sulfide ore has afforded oxygenated waters easy access to tremendous volumes of highly permeable, fine-grained, sulfide-rich tailings. The geochemical metal-partitioning processes of sorption and mineral precipitation do not appear to exert much of a diluting influence or control with regard to the seep waters dissolved metal species (other than Fe). The pH is much too low for significant sorption reactions to take place (Smith and others, 1994), and chemical analysis of highly oxidized precipitates associated with the actively flowing seep waters contained, not surprisingly, 32% Fe, but only moderate concentrations of 160 ppm Cu and 130 ppm Zn (Kilburn and others, 1994).

The preponderance of Fe detected in the waters and oxidized precipitates certainly points to a pyrite dominated system, while the rather conservative Cu and Zn determinations suggest a lack of significant chalcopyrite and sphalerite in the tailings, apparently an indication of efficient milling procedures at the Holden mine. Mill tailings analytical data seems to corroborate this line of reasoning, containing almost 5% Fe, 0.6 ppm Au and only moderate amounts of Cu (110 ppm) and Zn (190 ppm) (Kilburn and others, 1994). The integrity of the tailings sample, however, should be considered somewhat suspect, since it consisted of a single grab sample collected at the surface and cannot be regarded as truly representative. Moreover, surface tailings would be the first material to face dissolution by oxidation and weathering processes, liberating migratory trace metals and concentrating the immobile Au.

From the data at hand, it remains uncertain whether the acidic, metalliferous seep waters pose an environmental threat to Railroad Creek. Water samples taken from Railroad Creek immediately above and below the mill tailings showed very little chemical variance, and compared favorably with background levels noted elsewhere along Railroad Creek and its tributaries. Given the relatively low discharge volumes from the acidic seeps and the high runoff in Railroad Creek, it appears that metal concentrations and pH are immediately diluted to background levels or precipitated. The impact of seasonal variation, however, was not addressed and could be significant.

PORTAL WATERS

Effluent waters emerging from the mine portal rapidly flow downhill nearly a quarter mile before discharging directly into Railroad Creek, some 200 ft below the mine entrance. Suspended in the water or adhering to rocks and debris the length of the mine drainage was a thick, milky-white, flocculent material, later identified as amorphous Al and Fe hydroxides. Water collected at the entrance of the mine (portal) had a pH of 5.2, conductivity

of 760 µS/cm, and contained 410 mg/L SO_4, in addition to concentrations (in µg/L) of 1,000 Al, 21 Cd, 570 Cu, 580 Fe, and 4,800 Zn. As the mine waters descended the hillside prior to entering Railroad Creek, their chemical properties remained remarkably uniform. A sample of the effluent discharge taken just above its confluence with Railroad Creek had a pH of 5.0, conductivity of 800 µS/cm, and carried in solution 406 mg/L SO_4, as well as (in µg/L) 940 Al, 21 Cd, 580 Cu, 370 Fe, and 4,900 Zn (Kilburn, and others, 1994). The drop in dissolved Fe is attributed to the continued hydrolysis of Fe and precipitation of Fe-hydroxides. According to the classification diagram of Ficklin and others (1992), the effluent waters are considered an acid-high metal solution, the high metal rating based almost entirely on the dissolved Zn content. This is not surprising since Zn, which is by and large the primary base metal component in mine drainage streams, is highly mobile and resists sorption reactions within the pH range of the Holden adit waters (Plumlee and others, 1993; Smith and others, 1994).

The flocculated Al and Fe hydroxides, which are amorphous and were identified on the basis of chemical evidence, probably precipitated from solution as a result of pH dependent solubility reactions (Smith and others, 1989). The development of the metal hydroxides and corresponding acidic, metal-rich adit waters is attributed to many complex and sometimes indistinct geochemical and geologic mechanisms (Plumlee and others, 1994). Although these interacting processes or controls are unique to each deposit, certain general inferences can be made regarding the evolution of the Holden adit waters.

Ground water flowing or percolating through faults, fractures, or other permeable zones within the abandoned underground workings, react with and readily oxidize and dissolve pyrite and other sulfides, liberating H^+, Fe^{2+}, SO_4^{2-}, trace metals (such as Zn, Cu, Pb, and Mo) and add high concentrations of acid to the migrating waters. The ensuing acidic waters are highly corrosive and aggressively attack not only sulfide minerals but the enclosing wallrock as well, prompting the acid dissolution of resistive minerals such as the aluminosilicates (releasing in solution such major elements as Al, Ca, K, Na, and Mg) while at the same time consuming acid in the process. Influenced by various buffering reactions such as the wallrock interaction described above and/or dilution by divergent ground and mine waters, the pH of the solution rises to the point that precipitation of the Al and Fe hydroxides and coprecipitation or sorption of dissolved trace metals onto these same hydrous metal oxides occurs (Smith and others, 1994). Chemical analysis of the metal hydroxides merely confirmed their affinity for attracting trace metals, in particular Ag (1.6 ppm), Cu (710 ppm), Mo (19 ppm), Pb (160 ppm), and to a lesser extent Cd (.63 ppm) and Zn (200 ppm) (Kilburn and others, 1994). Although metal mobility and partitioning mechanisms are not sufficiently understood, the reactions are pH dependent, generally escalating quickly as pH

increases toward neutral (Smith and others, 1993).

The influence on Railroad Creek by the mine discharge, like the seep water, is unclear. Certainly multifarious sorption and metal-partioning reactions have played a dominant role in controlling and tempering the mine waters dissolved metal content prior to intersecting Railroad Creek. A survey of the data indicates that the flocculated metal hydroxides act as an effective sink for a number of prominent trace metals (e.g. Cu, Pb, and Mo), yet aqueous concentrations of Zn (and to a lesser extent Cd) remain significant at pH 5.0-5.2, reflecting their high mobility at pH levels where other metals are normally sorbed (Smith and others, 1994). How long the sorbed metals remain immobile within the solid phase is subject to the scavenging metal hydroxides capacity to resist dissolution (Smith and others, 1993). The chemistry of water collected below the confluence of the mine drainage and Railroad Creek is comparable to natural background levels elsewhere in the Holden area, suggesting again, the attenuating effect of the high runoff noted in Railroad Creek and the less than extreme metal content of the mine waters. Seasonal weather variations, however, would no doubt affect metal dispersion.

SECONDARY SULFATE SALTS

As seepage at the base of the mill tailings evaporates during intervals of dry weather, a somewhat less obvious source of contamination is introduced at the Holden site in the form of various soluble salts derived from sulfide weathering and related acid-generation. These encrustations or secondary sulfate salts, which develop as thin coatings on basal tailing material and other debris, store acid and metals in the solid form. During the course of hydrologic events such as rain or seasonal snowmelt, they readily dissolve, releasing their toxic constituents to the environment.

Samples of the sulfate-rich coatings were collected along the periphery of the tailings pile near the base. Ensuing XRD analysis of the salts revealed an extremely complex mineralogy, accommodating numerous interrelated secondary sulfates as well as minor amounts of relict primary minerals. Magnesium and aluminum sulfates were the common constituent in all the samples, with crusts gathered along the western fringe of the tailings pile exhibiting the most complex sulfate suites (including Fe, Cu, and Zn phases). Most of the salts possessed relatively low to moderate metal concentrations, with notable enrichment variations of 3 to 16 ppm Cd, 94 to 5,800 ppm Cu, and 320 to 15,000 ppm Zn. Severe metal accumulations, however, were detected in coatings from the western lip of the tailings pile, with metal levels reaching 13 ppm Ag, 370 ppm Cd, 43,000 ppm Cu, 280 ppm Mo, 120 ppm Pb, and 40,000 ppm Zn (Kilburn and others, 1994). These extreme metal concentrations are associated with the multiple or complex sulfate phases previously alluded to, and include: pickeringite [$MgAl_2(SO_4)_4 \cdot 22H_2O$], halotrichite [$Al_2Fe(SO_4)_4 \cdot 22H_2O$, kalinite [$KAl(SO_4)_2 \cdot 11H_2O$], alunogen [$Al_2(SO_4)_3 \cdot 17H_2O$, siderotil

[$FeSO_4 \cdot 5H_2O$], antlerite [$Cu_3(SO_4)(OH)_4$, gunningite [$ZnSO_4 \cdot H_2O$], chalcanthite [$CuSO_4 \cdot 5H_2O$], and bonattite [$CuSO_4 \cdot 3H_2O$].

The underlying cause with respect to the isolated growth of exotic efflorescent sulfate minerals in company with exceptionally high metal concentrations is obvious; localized access to highly acidic waters transporting in solution high to extreme levels of Fe, Al, Cu, Zn, and other metals. The particulars however, remain problematic. Perhaps there is some sort of "hardpan" effect here, whereby cemented layers have developed in other parts of the tailing pile, greatly reducing the mobility of dissolved metals (Blowes and others, 1991), or maybe the western edge of the impoundment is simply enriched in major and trace metals. A plausible explanation stems from the sample locality itself, positioned immediately below the dismantled mill and related waste dumps, likely areas of sulfide weathering. Acidic, metalliferous runoff from these sites, on their descent to Railroad Creek , would inevitably advance directly over the metal-rich salt flats, culminating at times,in the seasonal precipitation of high-metal sulfates. A stream sediment and corresponding heavy-mineral concentrate sample gathered in support of the regional mineral resource assessment seem to bolster this interpretation (at least the local availability of large amounts of sulfide minerals). The sediment and derivative concentrate, collected adjacent to the high-metal sulfate site, contained by far the most geochemically anomalous suite of metals of any sediment samples taken in the regional mineral study (collectively enriched in Fe, Ag, Au, Cd, Cu, Mo, Pb, and Zn). Optical studies found the concentrate sample rich in sulfides (over 50%), and identified substantial pyrite with minor amounts of chalcopyrite and sphalerite.

In other areas the negative environmental impact (such as periodic fish kills) commonly attributed to the dissolution and aqueous dispersion of highly soluble metal sulfates is well documented (Nimick and Moore, 1991) and should not be minimized. The extreme metal properties identified with some of the Holden areas secondary salts undoubtably earmark sources of pollution and are cause for some concern. Runoff associated with these salts following seasonal storms would probably lead to short-term spikes or increases in the acidity and dissolved metal content of nearby Railroad Creek. If and to what extent this represents an environmental hazard or is in anyway harmful to the creeks biota is unknown at this time.

CONCLUSIONS

Each mine site is unique, yet environmental hazards associated with acid-mine drainage remains a common and disturbing factor in mineral development. Volcanogenic massive sulfide (VMS) deposits such as Holden are inherently rich in pyrite and other sulfides. When oxygen-charged waters come in contact with waste piles, underground workings, and unmined ore zones of such deposits, they often acquire high concentrations of acid and dissolved metals and represent potential sources of acid

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and metal loading in the environment. Within the necessary constraints of reconnaissance sampling, some insight into possible problems associated with acid-mine drainage at the Holden orebody, a relatively low-grade VMS deposit, were realized. The study, because of its restrictive nature, did not address such issues as the dominant controls that influence mine drainage composition or seasonal variation, and are left to more detailed investigations.

Two sources of contaminated water flowing directly into nearby Railroad Creek were identified at the Holden deposit. The first consists of a series of highly acidic, metal-rich seeps located at the base of the mill tailings and the other is acidic, metal-rich effluent water exiting the mine portal. It's unknown whether these contaminated waters have a serious impact on Railroad Creek. The chemical properties of water samples collected below the confluence of the acidic, metalliferous waters and Railroad Creek compared favorably with background levels for the area. Given the relatively low discharge volumes from the seep and adit waters and the high runoff in Railroad Creek, it appears that metal concentrations and pH in the contaminated waters are quickly diluted to background levels.

In addition to acid drainage, a less conspicuous source of contamination is found at the base of the tailings impoundment in the form of various soluble efflorescent sulfate minerals, also byproducts of sulfide weathering. Certain of these secondary sulfates or salts, were found to host exceedingly high concentrations of metals in addition to acid in the solid phase. During the course of hydrologic events, these salts quickly dissolve, releasing their toxic constituents to the environment. The environmental consequences caused by the periodic dissolution of these salts, other than temporary acid and metal spikes in Railroad Creek, is uncertain.

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