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Chapter 25

Attenuation of Acid Rock Drainage in a Natural Wetland System

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At an abandoned mine site on Mount Washington in central Vancouver Island, British Columbia, natural attenuation of acid rock drainage is occurring in a small pond 50 m downstream from a shallow open pit. During the relatively dry summer months, water at the outlet of the pond system usually exhibits a pH value about one-half unit higher than that at the inlet. Dissolved concentrations of Cu and Zn and sulfate are concomitantly reduced to values lower by factors of 7, 15 and 3, respectively. Analyses of sediment cores from the pond reveal two distinct geochemical settings. Under oxidizing, deltaic conditions at the pond inlet, elemental copper and ferric hydroxide precipitate. Apparently, mixing of surface and ground waters provides the appropriate chemistry for the precipitation reactions to proceed. Near the pond outlet, reducing conditions predominate. Iron monosulfides and framboidal pyrite form *in situ*. Sulfur isotope analyses confirm that sulfate reduction is occurring in the pond sediments. In addition to formation of authigenic minerals, sorption of trace elements onto the iron hydroxides and sulfides is likely an important attenuation mechanism.

Acid rock drainage derived from oxidation of sulfide minerals is one of the most serious and costly environmental problems facing the mining industry today. Whereas recent research has greatly improved our knowledge on various aspects of acid mine drainage, abatement technology is still developing (e.g. 1). Though many active schemes for treating acid rock drainage are readily available, they are generally costly and require ongoing maintenance. In addition, sludges produced during conventional treatment (e.g. lime addition) often require costly storage and disposal. Passive or "walk-away" solutions are more desirable because of the possibility of eliminating long-term maintenance costs.

The natural capacity of wetlands to attenuate contaminant transport has been recognized for a long time and has been successfully utilized to treat municipal and

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industrial wastes (e.g. 2). However, the efficiency of passive wetland treatment to ameliorate metal-contaminated mine effluents is still largely unknown. For passive treatment to be effective, the long-term stability of natural wetlands, which tend gradually to fill with sediments, will have to be considered. Though recent research has clearly demonstrated the applicability of the concept under controlled conditions, using engineered wetlands (e.g. 3-5), there are only a few studies that provide information on the metal amelioration effect in natural systems (e.g. 6). Furthermore, the inorganic or biogeochemical mechanisms of metal attenuation in engineered or natural wetlands generally have not been described in detail.

The purpose of this paper is two-fold: 1) to document the attenuation of acid mine drainage products in a natural wetland system; and, 2) to investigate possible geochemical reactions involved in the metal attenuation process.

Site Location and Description

The abandoned Mount Washington Mine is located on the north side of Mount Washington (Figure 1) at an elevation of about 1320 m above mean sea level. The mine operated for about two years, starting in December, 1964. The ore consisted mainly of stockworks of chalcopyrite in fracture and breccia zones close to the contact of a Tertiary dioritic laccolith with Cretaceous meta-sediments (7). The abandoned mine site contains two remnant open pits, designated respectively as the North and the South Pit, each with associated waste rock dumps. The North Pit renders acidic drainage enriched in copper which apparently has had a detrimental effect on the salmonoid population downstream, notably in the Tsolum River (8, 9).

Figure 1b shows a map of the North Pit and the drainage leaving the site. The main generators of acidic drainage are the East Dump and the highly fractured bedrock in the shallow open pit which still contains abundant sulfides including chalcopyrite. The drainage flows to Pyrrhotite Creek, one of the headwater streams of the Tsolum River. The site has a steep slope; the vertical distance between the southern boundary of the pit and the toe of the East Dump near Weir #1 is 60 m. Snow melt is the main source of surface runoff, with peak discharge occurring usually in late May. Groundwater flow at the site is fracture-controlled with the hydraulic gradient largely dictated by the local topography.

Based on petrographic evidence, Kwong and Ferguson (10) suggested that acid generation in the North Pit was initiated with the stepwise oxidation of pyrrhotite. The relative reactivity of the prevalent sulfide minerals is in the following order:

marcasite > pyrrhotite > chalcopyrite > pyrite = arsenopyrite

Marcasite and elemental sulfur are intermediate alteration products of pyrrhotite, and are in turn oxidized to sulfate and hydrogen ions in solution and ferric hydroxide precipitates. A detailed discussion of the acid generation reactions operative at the site has been presented by Kwong (11). Chalcopyrite is dissolved by the acid generated during oxidation of the iron sulfides. During spring freshet and heavy fall rainstorms, flushing of concentrated acid solutions adhered to mineral or rock surfaces and dissolution of alteration minerals including soluble sulfate and carbonate salts (10) produce high acidity and metal loadings in drainage leaving the mine site. From June to September, the flux of surface drainage from the open pit decreases to a negligible value due to summer drought. In contrast, seepage from the East Dump directed through Weir #1 (Figure 1b) persists, although the flow rate may drop by more than 50%.

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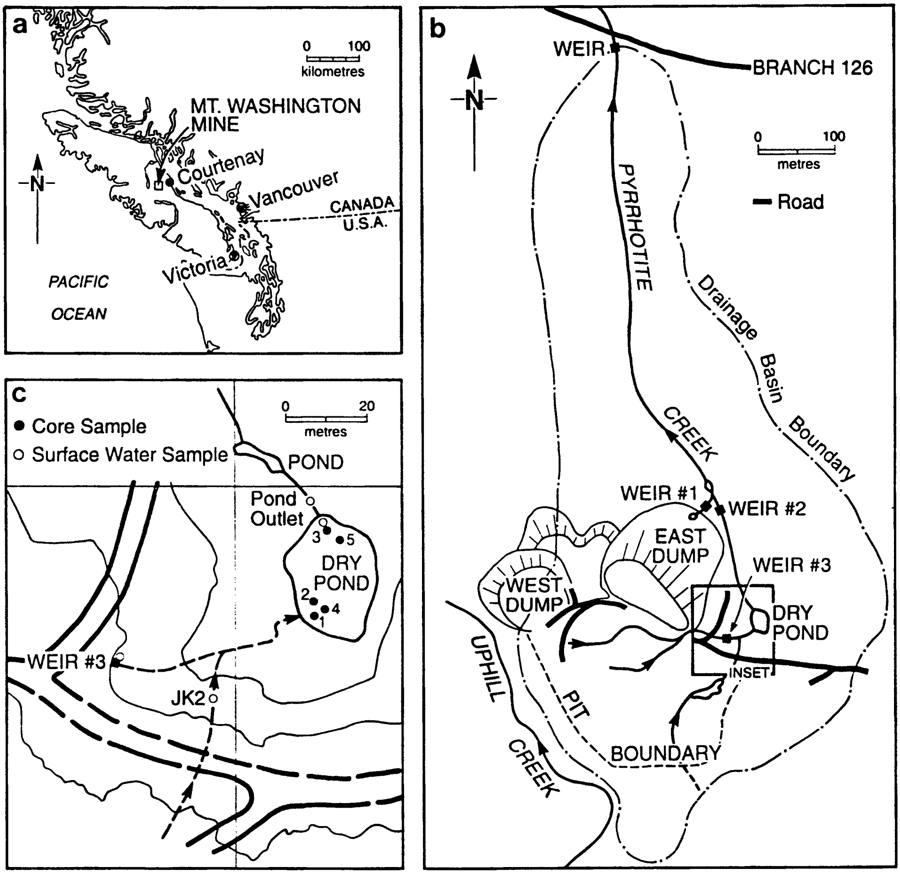

Figure 1. Maps showing (a) location of the Mt. Washington mine, (b) drainage leaving the North Pit (adapted in part from ref. 13) and (c) sample locations in the vicinity of Dry Pond.

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The Dry Pond - A Natural Wetland System

General Setting. The Dry Pond, located 50 m downstream from the open pit, is a natural pond with an area of about 700 m^2 and a maximum depth of approximately 1 m. It drains into the perennial Pyrrhotite Creek system. Surface runoff from the North Pit accounts for the bulk of drainage water volume in the pond during the wet seasons (early spring and late fall), although ground-water seepage prevents it from drying out during summer drought. The pond inlet is characterized by a deltaic environment in which a mixture of yellowish-brown ferric hydroxide sludge and other fine-grained sediments are deposited. Unless stated otherwise, in this paper ferric hydroxide refers to undifferentiated ochreous precipitates of Fe-hydroxide, Fe-oxyhydroxides (ferrihydrite and goethite) and, possibly, minor Fe-oxyhydroxysulfate (12). Near its outlet, the pond has a steep slope and the shore is covered by forest vegetation. Often, an odor of hydrogen sulfide can be detected near the outlet. Between the pond outlet and Weir #2 near the toe of the East Dump (Figure 1b), Pyrrhotite Creek flows through a few short intervals of swampy areas where a soil profile is covered by peat, sphagnum moss or sedges. Elsewhere, the creek bed is rocky with prominent coatings of yellowish-brown ferric hydroxide.

Despite its small size, the Dry Pond system between Weirs # 3 and 2 apparently has a high capacity for attenuation of acid mine drainage products. Through most of the year, the water draining through Weir #2 has a higher pH and lower concentrations of dissolved Cu, Zn, and sulfate relative to Weir #3 (13). To investigate the fate and transport of the acid mine drainage products in the system, a series of surface-water samples and five sediment cores (Figure 1c) were collected in late September (a typical low-flow period), 1990, for various analyses. To shed light on possible seasonal variations in water chemistry, analyses of surface waters collected in early July, 1990 were compared to the September data.

Sampling and Analytical Methods. Surface-water samples were pumped through a 0.45 μm cellulose nitrate membrane filter into a flow cell equipped with a temperature probe, pH and Eh electrodes and a conductivity cell. After the system was thoroughly flushed, all measurements were recorded and two 125 ml (for metal and sulfate analyses) and one 250 ml (for isotope analysis) samples were collected. The samples for metal analysis were preserved with 1 ml of ultra-pure nitric acid.

Three sediment cores were sampled near the inlet and two near the outlet of the pond (Figure 1c) using 6 cm (diameter) PVC tubes. While still under water, the bottom of each core tube was sealed with a rubber stopper. Excess tube was cut off and the core was capped with another rubber stopper and tightly wrapped with duct tape for transportation.

In the laboratory, the cores were sectioned in an argon atmosphere to minimize oxidation (e.g. precipitation of ferric hydroxide). The pore water from each segment was squeezed out using a stainless steel pneumatic pressure cell. Metal concentrations in subsamples of each squeezed core segment were determined by induction coupled plasma analysis. Selected core segments were also examined using a JEOL JSM-840A scanning electron microscope equipped with a KEVEX energy dispersive X-ray analyzer. Mineralogical analyses were conducted using a SCINTAG XDS 2000 X-ray diffractometer with a copper anode. The dissolved Fe(total), Cu and Zn contents of the pore waters and the surface-water samples were determined by atomic absorption spectroscopy and dissolved sulfate by ion chromatography. For sufficiently large water samples, the dissolved sulfate was precipitated as barium sulfate and the sulfur isotope composition determined by mass spectrometry (14).

Observations. The analyses of the September 1990 suite of surface-water samples (Table I) show a pH increase of nearly 0.7 units, and reduction of dissolved

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concentrations of Cu, Zn and sulfate to values lower by factors of 7, 15 and 3, respectively, in the outflow (Weir #2) relative to the inflow (Weir #3). The total dissolved Fe concentrations also tend to decrease downstream. The anomalously low Fe value for the September Weir #3 sample may have resulted from either precipitation of ferric hydroxide or an analytical error. Along the flow path, dissolved sulfate is also progressively enriched in ^{34}S relative to ^{32}S suggesting that sulfate reduction has occurred.

Table I. Aqueous and Sulfur Isotope (dissolved sulfate) Geochemistry of Surface Water in the Dry Pond System, Mt. Washington Mine

Sample	*Sampling Date (1990)*	*pH*	*Cu*	*Zn*	*Fe(total)*	*SO_4*	*$\delta^{34}S$ (SO_4)*
			mg/L				*‰ CDT*
JK2	July 5	6.20	0.33	0.14	<0.03	126	-0.1
Weir 1	Sept. 11	3.61	12.8	0.65	2.17	794	+0.7
Weir 2	Sept. 11	4.49	3.26	0.11	0.31	172	+1.4
Weir 3	July 5	3.50	20.7	0.76	4.78	422	-0.1
	Sept. 11	3.83	20.7	1.67	0.84	581	+0.1
Dry Pond	July 5	3.70	14.9	0.51	1.41	274	+0.2
	Sept. 11	3.98	12.0	0.65	1.67	323	+1.0
Pond Outlet	Sept. 11	4.02	6.32	0.32	0.19	249	+1.1

A comparison of the July and September 1990 Dry Pond samples suggests that the extent of sulfate reduction and metal attenuation is inversely proportional to the inflow volume. September was drier than July and thus the downstream attenuation effect was more prominent in September. In addition, the July data indicate that drainage from south of the Dry Pond (JK2) was near neutral in pH and low in metals. The surface drainage from JK2 has little influence on the overall chemistry of the Dry Pond system because its discharge is generally a small fraction of that from Weir #3. However, the composition of the JK2 drainage can be used as a proxy for uncontaminated ground water in the vicinity of the Dry Pond.

Table II shows the pore-water geochemistry of the Dry Pond sediments. With depth, pH values tend to increase and dissolved concentrations of Cu, Zn, Fe and sulfate tend to decrease . Also, the pore-water sulfate becomes more ^{34}S-enriched with depth. Moreover, just beneath the water-sediment interface, pore-water sulfate near the pond outlet is more enriched in the heavier sulfur isotope than that from near the pond inlet.

The three cores sampled at the pond inlet are dominantly yellowish-brown reflecting the abundance of ferric hydroxide; streaks of black organic matter become more prominent with depth. When examined under a scanning electron microscope equipped with an energy-dispersive X-ray analyzer, elemental copper was observed in close association with ferric hydroxide (Figure 2) in the 10-20 cm section of Core #1 and about 10 cm below the water-sediment interface in Core #2. The presence of native copper was later confirmed by X-ray diffraction analysis. The ferric hydroxide, however, is apparently transparent to X-rays and the energy dispersive spectrum

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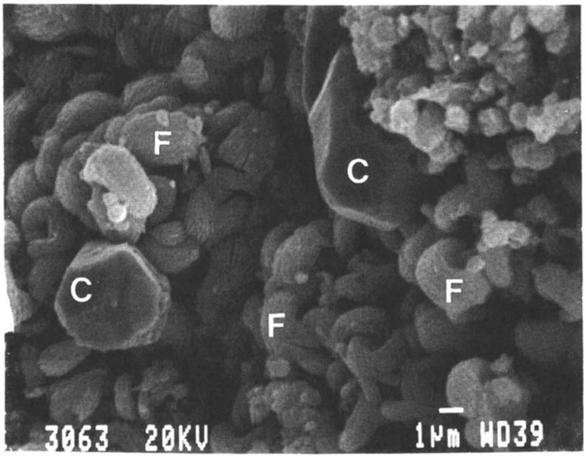

Figure 2. A scanning electron photomicrograph of the ferric hydroxide (F) - elemental copper (C) assemblage observed in shallow sediments near the pond inlet.

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suggests the absence of sulfate. Overall, the total copper contents of these two core segments (after squeezing) are 1.2% and 0.93%, respectively.

Table II. Pore-water Geochemistry of the Dry Pond Sediments, Mt. Washington Mine (Date of sampling: September 11, 1990)

Core # (water depth cm)	*Core Depth (cm)*	*pH*	*Cu*	*Zn*	*Fe(total)*	*SO_4*	*$\delta^{34}S$ (SO_4)*
			mg/L				‰ CDT
1 (0)	0-4	4.87	0.74	0.34	96.6	413	ND*
	4-10	4.80	3.90	0.48	68.4	399	ND*
	10-20	5.05	1.06	0.14	50.1	301	ND*
2 (13)	0-10	3.31	1.40	0.18	40.4	246	+2.1
	10-18	6.30	0.16	0.03	21.4	118	+7.0
	18-28	6.65	0.02	<0.01	4.95	51.8	+10.7
4 (10)	0-32	6.23	0.04	0.09	14.9	132	+1.7
3 (15)	0-8	3.45	1.25	0.17	12.4	312	ND*
5 (20)	0-6	6.08	0.06	0.08	3.10	168	+8.0

*ND = not determined

The two cores sampled near the pond outlet are greyish-black in color, reflecting the abundance of organic debris and reducing conditions. Both cores contain framboidal pyrite in association with iron monosulfide (Figure 3). Whereas the presence of pyrite has been confirmed by X-ray diffraction analysis, the mineralogy of the iron monosulfide has not been determined.

Mechanisms of Metal Attenuation

Based on the hydrologic setting of the Dry Pond, the surface- and pore-water chemistries and the mineral phases observed in the sediments, three processes are apparently responsible for metal attenuation in the Dry Pond system: i) mixing of surface drainage with near-neutral pH ground water that seeps into the pond, ii) precipitation of metals as discrete mineral phases, including copper in the shallow sediment near the pond inlet, and iii) sorption onto surfaces of minerals including ferric hydroxide in the pond inlet area and iron sulfides in the pond outlet area. In addition, as suggested by Machemer and Wildeman (5), sorption of metals onto organic compounds may be an important attenuation mechanism.

Sorption of trace elements onto precipitating iron oxyhydroxides and pyrite is a well-documented phenomenon (e.g. 15, 16). The large surface areas observed in the ferric hydroxide and framboidal pyrite grains (Figure 3) identified from the Dry Pond sediments indicate that abundant active sites are available for sorption of trace metals. However, based on the present data, we could not determine the relative importance of sorption of metals in the Dry Pond system.

Over the long term, precipitation is likely the dominant process for metal removal from acid mine drainage (5). Thus, it is worthwhile to consider possible geochemical reactions occurring in the Dry Pond system that have resulted in the formation of authigenic minerals. Generally speaking, the pond inlet area is an oxidizing

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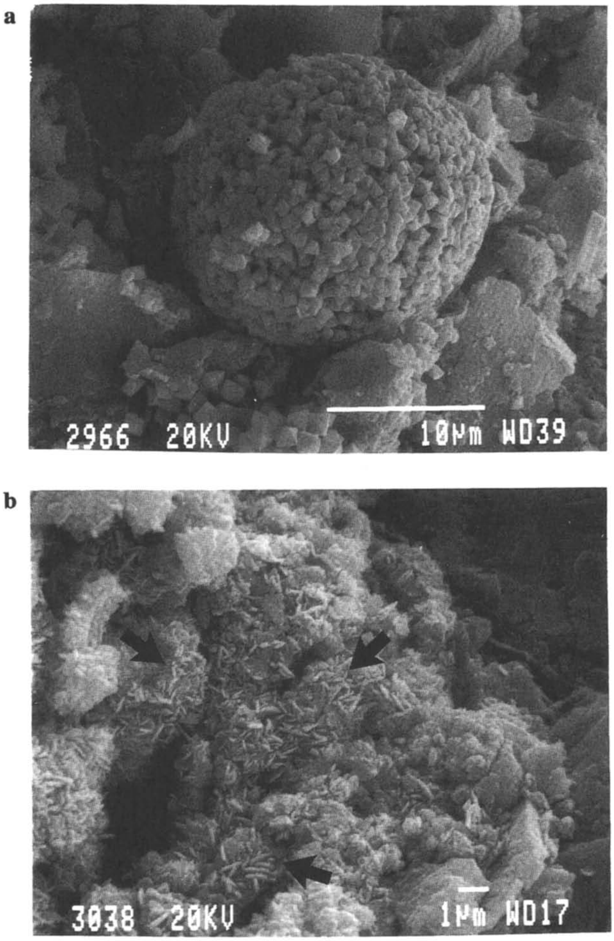

Figure 3. Scanning electron photomicrographs of (a) framboidal pyrite and (b) iron monosulfide (arrows) observed in shallow sediments near the pond outlet.

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environment marked by relatively rapid sedimentation under shallow water conditions. Geochemical reactions that can account for the formation of the observed minerals include the following:

$$2Fe^{2+} + Cu^{2+} \Leftrightarrow 2Fe^{3+} + Cu^0_{(s)} \quad (1)$$

$$Fe^{3+} + 3H_2O \Leftrightarrow Fe(OH)_{3(s)} + 3H^+ \quad (2)$$

Combining Equations 1 and 2, the overall reaction is:

$$2Fe^{2+} + Cu^{2+} + 6H_2O \Leftrightarrow 2Fe(OH)_{3(s)} + Cu^0_{(s)} + 6H^+ \quad (3)$$

In contrast, the pond outlet has a relatively reducing environment as evidenced by the dark color of the sediment and hydrogen sulfide odor. Using CH_2O as a proxy for all organic matter, the following geochemical reactions (cf. 17) can explain the formation of the observed iron sulfides:

$$2H^+ + SO_4^{2-} + 2CH_2O \Rightarrow 2CO_2 + 2H_2O + H_2S \quad (4)$$

$$4Fe(OH)_{3(s)} + CH_2O + 8H^+ \Rightarrow 4Fe^{2+} + CO_2 + 11H_2O \quad (5)$$

$$4H_2S + 4Fe^{2+} \Rightarrow 4FeS_{(s)} + 8H^+ \quad (6)$$

$$CO_2 + 4FeS_{(s)} + 4H_2S \Rightarrow 4FeS_{2(s)} + CH_4 + 2H_2O \quad (7)$$

In all cores analyzed (Table II), sulfate at depth is ^{34}S-enriched relative to Dry Pond surface water (Table I). These isotope data provide further evidence that sulfate reduction is taking place in the natural wetland system, including locally at depth in the pond inlet area.

The formation of pyrite and iron monosulfide is common in many modern reducing sedimentary environments including constructed wetlands used to treat mine drainage (5, 18). In contrast, although native copper is a common supergene alteration mineral (e.g. 19), its formation in modern sediments has not been reported. Therefore, it is useful to consider whether reactions 1, 2 and 3 could have occurred in the Dry Pond environment. In particular, one should examine the change in Gibbs free energy of the overall reaction (Equation 3) accounting for the coexistence of ferric hydroxide and native copper.

Under standard conditions, based on free energy of formation data tabulated by Wagman et al. (20), the reaction as described by Equation 3 should proceed to the left. However, the direction of the reaction is influenced by the relative activities of H^+, Fe^{2+} and Cu^{2+} as evident in the following expression of free energy change for the reaction:

$$\Delta G_r = \Delta G_r^0 + RT\ln\{[H^+]^6/[Fe^{2+}]^2[Cu^{2+}]\} \quad (8)$$

where ΔG_r and ΔG_r^0 are the change in Gibbs free energy under ambient and standard conditions, respectively; R is the gas constant, T the absolute temperature in Kelvins, and, [ ] denotes aqueous activity.

Using the measured pore-water chemistry corresponding to the depth of copper occurrence in Core #1 (i.e. pH = 5.0; dissolved Cu = 1.0 mg/L, assumed all to be in

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cupric state; and, dissolved Fe = 50 mg/L, assumed all to be in ferrous state) and assuming aqueous activity coefficients = 1, at 25°C and atmospheric pressure, the Gibbs free energy change of reaction 3 is still positive indicating that the reaction will again proceed to the left. However, if we assume that the pore water of Core #1 has been affected by mixing and/or diffusion during sample transport and storage and that the hydrogen ion activity observed in Core #2 is more representative of the field conditions, an entirely different conclusion is reached. For pH = 5.5, dissolved Cu^{2+} = 1.0 mg/L and dissolved Fe^{2+} = 30 mg/L (all values interpolated from the top of the sediment to the depth of native copper occurrence in Core #2), reaction 3 has a negative Gibbs free energy change, making the forward reaction possible.

Similarly, Equation 1 represents a reaction that is dependent on the relative ion activities, Fe^{3+}, Fe^{2+}, and Cu^{2+}. The mixing of surface and ground water in the pond inlet area may have produced the right conditions for the reaction to proceed to the right. For Eh = 0.2v and T = 25°C, pH = 5.5, dissolved Cu^{2+} = 1.0 mg/L and dissolved Fe^{2+} = 30 mg/L as above, reaction 1 has a negative change in Gibbs free energy favoring the precipitation of native copper.

Thus we conclude that the proposed reactions 1, 2 and 3 are thermodynamically feasible, given the right conditions. In absence of sufficient data, notably *in situ* Eh measurements and Fe(II) and Fe(total) analyses, a detailed speciation analysis is not warranted.

As shown in Tables I and II, Zn, relative to Cu, is only a minor component of the surface- and pore-water samples. The attenuation of Zn in the Dry Pond system could be explained by sorption and/or precipitation of Zn sulfide. However, we do not have direct evidence to determine which process is dominant.

The extent of attenuation of metals and acidity by various mechanisms is flow-dependent. In the Dry Pond system, acid attenuation by sulfate reduction and the accompanied immobilization of dissolved metals are minimal during high surface flow periods because the rate of flow of H^+ and metals through the system is much greater than the rate of consumption by attenuation reactions. Larger wetland systems occur downstream from the mine site. These could perhaps be developed to manage peak flow conditions to produce a stepwise reduction in acidity and dissolved metals along the drainage flow path.

Summary and Conclusion

In this paper, we present evidence for attenuation of acid mine drainage products in a natural wetland system at the abandoned Mount Washington Mine. In particular, we document the precipitation of authigenic minerals including elemental copper and iron sulfides. The precipitation of elemental copper may be related to mixing with neutral ground water. Another important metal attenuation process may be sorption of dissolved metals onto surfaces of minerals and organic phases. Given that the acidity and metal contents of drainage from the Mount Washington mine site are orders of magnitude lower than those prevailing at other sites, such as Equity Silver or Gibraltar (21), and that sulfate reduction is actively occurring in the Dry Pond, passive wetland treatment may be a viable option for the reclamation of the site. The major challenge lies in how peak flows can be managed effectively during the wet seasons, to prolong water retention in the wetland system so that sulfate reduction can proceed. Perhaps a series of wetland systems could be developed downstream from the site to enhance sulfate reduction and metal attenuation. Furthermore, we recommend that future investigations should examine in more detail the seasonal dynamics of metal fluxes from surface water to sediments.

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