ACID-NEUTRALIZATION REACTIONS IN INACTIVE MINE TAILINGS IMPOUNDMENTS AND THEIR EFFECT ON THE TRANSPORT OF DISSOLVED METALS[1]

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Abstract: The H^+ generated by sulfide oxidation in inactive mine wastes is consumed by a series of mineral-dissolution reactions. Previous studies conducted at the Nordic uranium tailings impoundment near Elliot Lake, ON, indicated these H^+-consuming reactions occur in a stepwise sequence: calcite dissolution, siderite dissolution, $Al(OH)_3$ dissolution and $Fe(OH)_3$ dissolution. Similar series of acid neutralization reactions have been observed at tailings impoundments at the Heath Steele Zn-Pb mine, NB, the Waite Amulet Cu-Zn mine, Noranda, QC, the Delnite Au mine, Timmins, ON, and the Copper Cliff and Nickel Rim Ni-mines, Sudbury, ON. At all of these locations, near-equilibrium conditions, as indicated by geochemical modelling conducted using MINTEQA2 or PHRQPITZ, are attained with respect to similar sequences of pH-buffering phases. These acid-neutralization reactions have a strong effect on the transport of dissolved metals. Calculations conducted using the 2-D coupled solute-transport/geochemical mass-transfer model MINTRAN predict pH buffering sequences and metal attenuation mechanisms that are similar to those observed at the field sites. Model simulations suggest that some pH-sensitive metals should be retained within the tailings or underlying aquifers.

Additional Key Words: acid-neutralization, tailings, metals, solute-transport modelling, groundwater, aquifers

Introduction

Acid-neutralization processes are fundamental in controlling the environmental effects of wastes from base- and precious-metal mines. In mine tailings impoundments, the balance between H^+-generating sulfide-oxidation reactions and H^+-consuming mineral-dissolution reactions controls the pH at the location of sulfide oxidation and at locations downgradient along the groundwater flow path. Buffering reactions that control the pH near the location of sulfide oxidation affect the rate of sulfide oxidation and the release of dissolved Fe^{2+} and other metals to the tailings pore water. Buffering reactions that occur along the groundwater flow path affect the mobility of dissolved metals as they are transported through the mine wastes and through underlying geologic materials. Tailings impoundments are characterized by a large number of dissolved constituents and a large quantity of tailings solids, all contributing to the chemical evolution of the tailings pore water. The extent of pH buffering depends on the mass of acid-consuming minerals and on the mineralogy of these materials.

The importance of acid-neutralization reactions in mine wastes has been recognized for several decades, as has the benefit of attaining a balance between acid generation and acid neutralization. In response to the need to predict the potential for release of acid drainage from mine wastes, a series of predictive tests and acid-base accounting procedures has been developed (e.g. Ferguson and Erickson 1987). The objective of acid-base accounting procedures and acid-generation predictive tests is to assess the potential for a given mine waste to generate acid.

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The results of field studies indicate that acid neutralization in mine wastes and underlying aquifers occurs through a complex series of reactions (Smyth 1981; Morin 1983; Dubrovsky 1986; Morin et al. 1988). It is necessary to investigate the benefits and effects of these pH-buffering reactions because of the variety of metal-bearing sulfide minerals present in base- and precious-metal tailings impoundments and because the mobilities of many of the metals derived from oxidation of these sulfide minerals varies with changing pH.

Acid Generation in Mine Wastes

The principal reactions that generate H^+ are the oxidation of sulfide minerals, the oxidation of Fe^{2+} and hydrolysis of Fe^{3+}, and the precipitation of metal hydroxide and hydroxysulfate phases. The most common sulfide minerals in base- and precious-metal mine tailings are pyrite (FeS_2) and pyrrhotite ($Fe_{(1-x)}S$). The oxidation of pyrite produces two moles of H^+ for each mole of pyrite oxidized through the reaction:

$$FeS_2 + \frac{7}{2}O_2 + H_2O \rightarrow Fe^{2+} + 2SO_4^{2-} + 2H^+ \tag{1}$$

Subsequent oxidation of Fe^{2+} and hydrolysis and precipitation of $Fe(OH)_3$ produces an additional two moles of H^+ resulting in the overall reaction:

$$FeS_2 + \frac{15}{4}O_2 + \frac{7}{2}H_2O \rightarrow Fe(OH)_3 + 2SO_4^{2-} + 4H^+ \tag{2}$$

The precipitation of other Fe(III)-bearing phases, such as goethite (α-FeOOH) or schwertmannite ($Fe_8O_8(OH)_6SO_4$) (Bigham et al. 1990), may occur, releasing differing amounts of H^+. Alternatively, Fe^{3+} may be consumed through further oxidation of sulfide minerals, e.g.

$$FeS_2 + 14Fe^{3+} + 8H_2O \rightarrow 15Fe^{2+} + 2SO_4^{2-} + 16H^+ \tag{3}$$

In many tailings impoundments, particularly those resulting from the concentration and recovery of nickel, pyrrhotite is the most abundant sulfide mineral. Oxidation of pyrrhotite may proceed to completion through the reaction:

$$Fe_{(1-x)}S + (2-\frac{x}{2})O_2 + xH_2O \rightarrow (1-x)Fe^{2+} + SO_4^{2-} + 2xH^+ \tag{4}$$

or may proceed to partial completion, generating Fe^{2+} and elemental S^0 through the reaction:

$$Fe_{(1-x)}S + (2-2x)Fe^{3+} \rightarrow (3-3x)Fe^{2+} + S^0 \tag{5}$$

This elemental S^0 may be subsequently oxidized to SO_4^{2-}. Partial oxidation of pyrrhotite, resulting in accumulation of S^0, has been observed at two Ni mine-tailings impoundments (Coggans et al. 1994; Jambor and Owens 1993), and at an inactive Cu-Zn mine-tailings impoundment (Jambor 1987; Blowes and Jambor 1990).

The zone of active sulfide oxidation in most tailings impoundments is limited to the vadose zone, near the tailings surface, where oxygen transport by gas-phase diffusion is rapid. In this zone, the weathering of sulfide minerals may be complete (reaction 2), resulting in the formation of stable secondary precipitates such as goethite, or incomplete, with some of the Fe derived from the initial sulfide remaining in solution as dissolved Fe^{2+} (reaction

1). Ferrous iron-bearing sulfate and hydroxide minerals are relatively soluble. As a result, dissolved Fe^{2+} concentrations contained in tailings pore waters may be high (up to 70 g/L; Blowes et al. 1991). This dissolved Fe^{2+} is displaced along groundwater flow paths and ultimately discharged to the surface water flow system where oxidation to Fe^{3+} and precipitation of ferric oxyhydroxide phases result in the generation of H^+ in the surface-water flow system (Dubrovsky et al. 1984a; Coggans et al. 1994).

Table 1. Principal pH-buffering phases in mine tailings impoundments.

Mineral	Formula
Calcite	$CaCO_3$
Dolomite	$CaMg(CO_3)_2$
Siderite	$FeCO_3$
Mixed carbonates	$(Ca,Mg,Fe,Mn)CO_3$
Gibbsite[1]	$Al(OH)_3$
Ferrihydrite[1]	$Fe(OH)_3$
Goethite	α-FeOOH
K-Jarosite	$KFe_3(SO_4)_2(OH)_6$
Aluminosilicates	

[1] or equivalent amorphous phase

Acid Neutralization in Mine Wastes

The principal acid-neutralization mechanisms in inactive mine tailings impoundments are carbonate mineral dissolution, hydroxide mineral dissolution, and aluminosilicate mineral dissolution (table 1). Unlike sulfide-oxidation reactions, many acid-neutralization reactions are independent of gas-phase O_2 concentration. Acid-neutralization reactions can occur along the groundwater flow path throughout the tailings impoundment and in underlying aquifers, limited only by the availability of acid-consuming mineral phases. These H^+-consuming reactions result in the general increase in pH with increasing depth that is observed at many tailings impoundments (fig. 1; Dubrovsky et al. 1984a; Morin et al. 1988; Blowes and Jambor 1990; Blowes et al. 1991; Coggans et al. 1994). Consequently, the water present near the base of tailings impoundments, or present in aquifers underlying the tailings impoundment, is usually near-neutral in pH, but may contain high concentrations of acid-generating Fe^{2+}.

Acid-consuming reactions affect both the rate of sulfide oxidation and the distribution of sulfide-oxidation products between the tailings pore water and solid-phase precipitates. Numerous studies have concluded that the rate of sulfide oxidation in mine waste piles and heap-leach systems is controlled by the supply of oxidant to the mineral surface (Cathles 1979; Davis and Ritchie 1986; Nicholson et al. 1990). Processes limiting the supply of oxidant are transport through the mine waste pile to the depth of active oxidation and diffusion of oxidants (O_2 or Fe^{3+}) through oxide coatings that surround unaltered sulfide cores (Davis and Ritchie 1986). Nicholson et al. (1990) concluded that transport of oxygen through alteration rims surrounding pyrite grains controlled the rate of pyrite oxidation in an experimental study conducted under near-neutral pH conditions. In the field, Blowes and Jambor (1990) and Coggans et al. (1994) observed well-developed alteration rims, composed of goethite and jarosite, surrounding sulfide grains under low pH (3.5<pH<4.5) conditions. No alteration rims were observed by Blowes et al. (1994) on sulfide grains present under very low pH conditions (pH<1.5). This pattern suggests that the formation of secondary alteration rims may be a primary control on the rate of sulfide oxidation in all but the most extreme low pH settings.

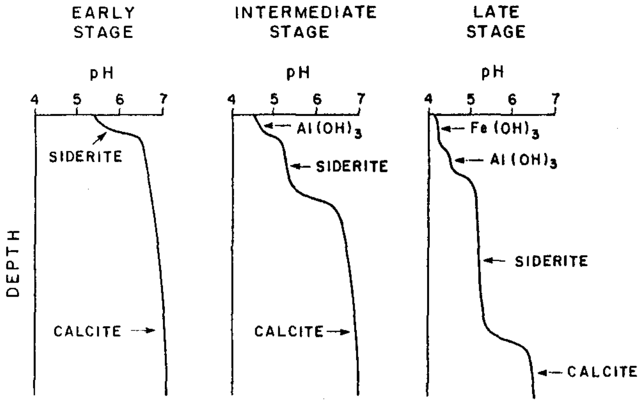

Figure 1. Development of pH-buffering zones during early, intermediate and late stages of sulfide oxidation.

Field observations made at tailings impoundments with similar sulfide contents but varying carbonate mineral contents suggest that the location of H^+ consumption is important in determining the potential for future acid production. The Nordic U tailings impoundment near Elliot Lake, Ontario, and the Delnite Au tailings impoundment at Timmins, Ontario both contain approximately 5 wt% sulfide minerals, primarily as pyrite. The carbonate content of the Nordic tailings is generally <0.1 wt% as $CaCO_3$, whereas the carbonate content of the Delnite tailings is >20 wt%. At the Nordic site, the pH in the vadose zone is low (1<pH<4) and much of the Fe^{2+} released through pyrite oxidation remains in solution, resulting in pore-water Fe concentrations that exceed 20,000 mg/L at some locations (Dubrovsky et al. 1984b). This water is being displaced down through the tailings and laterally through an underlying aquifer where it is neutralized along the groundwater flow path. Concentrations of dissolved Fe^{2+}, however, remain high along the groundwater flow path. Acidic conditions are generated as this ferrous-rich water discharges to the surface-water flow system downgradient from the tailings impoundment. At the Delnite site, the pH in the vadose zone has remained high (6<pH<8), favoring precipitation of Fe oxyhydroxide minerals, dominantly goethite (Jambor and Blowes 1991). As a result of goethite precipitation, Fe concentrations are generally < 50 mg/L (Blowes 1990). These Fe concentrations represent an acid-generating potential that is less than the acid-consuming potential of the pore-water alkalinity, therefore no acidity is generated as this water is discharged to the surface-water flow system. The importance of maintaining high carbonate contents in the vadose zone of the tailings impoundments is thus illustrated. At both sites, acid-neutralization reactions have produced pore waters near the base of the impoundments that are near neutral in pH, but the high concentrations of dissolved Fe^{2+} in the Nordic pore water represent a high acid-generating potential in contrast to the low acid-generating potential at Delnite.

Sequential pH Buffering Reactions

Field studies conducted in a number of tailings impoundments indicate series of equilibrium pH-buffering reactions controlling the pH of the tailings pore water (Smyth 1981; Dubrovsky et al. 1984a; Blowes and Jambor 1990; Johnson 1993). These series are similar to those which Morin et al. (1988) observed in a plume of tailings-derived water moving outward from beneath the Nordic Main uranium tailings impoundment near Elliot Lake, Ontario, where the sequence follows the order: calcite dissolution, siderite dissolution, $Al(OH)_3$ dissolution and $Fe(OH)_3$ dissolution.

The following example is based on results of a field study conducted at the old and new Heath Steele Zn-Pb tailings impoundments, New Brunswick (Blowes 1990; Blowes et al. 1991). As H^+ is released by sulfide oxidation reactions, the tailings pore water becomes undersaturated with respect to calcite and dolomite, leading to calcite and dolomite dissolution buffering the pore-water pH between 6.5 and 7.5.

$$CaCO_3 + H^+ \rightleftharpoons Ca^{2+} + HCO_3^- \tag{6}$$

$$CaMg(CO_3)_2 + 2H^+ \rightleftharpoons Ca^{2+} + Mg^{2+} + 2HCO_3^- \tag{7}$$

Geochemical equilibrium calculations suggest that the precipitation of siderite, $Al(OH)_3$ and $Fe(OH)_3$ is favored as calcite and dolomite dissolve. Ferrous iron generated from sulfide oxidation can react with calcite to form siderite through:

$$CaCO_3 + Fe^{2+} \rightleftharpoons FeCO_3 + Ca^{2+} \tag{8}$$

or can react directly with HCO_3^- released from dissolution of calcite or dolomite to form secondary siderite (Morin and Cherry 1986, Ptacek and Blowes 1993). When calcite is completely consumed, the pore water pH abruptly declines, favoring dissolution of primary siderite originally present in gangue materials or secondary siderite. In the siderite-buffered zone the precipitation of $Al(OH)_3$ and $Fe(OH)_3$ is favored. As siderite dissolves, the pH is buffered to between pH 4.8 and pH 6.3. After siderite is completely consumed, the pH again declines abruptly until the dissolution of the next pH buffer in the series, $Al(OH)_3$, is favored. Dissolution of $Al(OH)_3$ buffers the pH to values between approximately 4.0 to 4.3. When $Al(OH)_3$ is completely consumed, the pH declines, favoring the dissolution of $FeOH_3$ and resulting in pH values that fall below pH 3.5.

Sequences of similar pH-buffering zones have been observed at a number of tailings impoundments, including the Nordic Main U tailings impoundment (Dubrovsky et al. 1984a), the Waite Amulet Cu-Zn tailings impoundment (Blowes and Jambor 1990), the Copper Cliff Ni tailings impoundment (Coggans et al. 1994) and the Nickel Rim Ni tailings impoundment (Johnson 1993). At these sites, the amount of time required to deplete a given mineral phase and the relative thicknesses of the zones developed varied, depending on the original mass of solid-phase buffers contained in the gangue materials and on the initial mass of sulfide minerals. At other sites, where the H^+ generated by sulfide oxidation is less than the acid consumed by carbonate-mineral dissolution, such as at the Delnite site (Blowes 1990; Jambor and Blowes, 1991), the pore-water pH is controlled completely by carbonate-mineral dissolution, limiting the development of the remaining pH buffering zones.

The Effect of pH-Buffering Reactions on Trace Metal Mobility

Complete development of the pH-buffering sequence and its effect on the mobility of dissolved metals are illustrated by observations made at the Nickel Rim Ni tailings impoundment near Sudbury, Ontario (Johnson 1993). At the Nickel Rim site, sulfide oxidation in the vadose zone has released H^+ to the tailings water, resulting in low-pH ($2.1<pH<3.5$) conditions near the tailings surface. Interaction with H^+-consuming minerals has resulted in the progressive increase in pH with increasing depth observed in figure 2. Geochemical calculations conducted using MINTEQA2 (Allison et al. 1990) indicate that the pore waters approach or attain equilibrium with respect to $Fe(OH)_3$, $Al(OH)_3$, siderite and calcite, forming a series of pH-buffering zones moving downward from the tailings surface. The pH-buffering zones in figure 2 are labelled according to the dominant pH buffering process. Irregularities in the shapes of these pH-buffer zones are likely due to variations in the pore-water velocity, composition of the tailings solids and rates of acid generation.

At the Nickel Rim site, the pH of the tailings pore water has a strong effect on the movement of dissolved Al and Ni in the impoundment (fig. 2). At the interface between the Al-mineral pH-buffering zone and the siderite pH-buffering zone the concentration of dissolved Al decreases sharply. For example, at location NR2, located in the central portion of the tailings impoundment, the dissolved Al concentration decreases from > 1,150 mg/L to < 10 mg/L within a one meter interval (fig. 2). In this interval the pH increases from ~4 to ~5.8. Geochemical calculations conducted using MINTEQA2 indicate that the water samples from this location are saturated or slightly undersaturated with respect to amorphous $Al(OH)_3$ and supersaturated with respect to the crystalline aluminum hydroxide mineral gibbsite. These observations suggest that Al is removed from solution through the precipitation of an aluminum hydroxide or hydroxysulfate phase.

The concentration of dissolved Ni is also strongly dependent on the pore-water pH, showing a sharp decrease from > 250 mg/L to < 10 mg/L as the pH increases to above 5.8 within the siderite pH-buffering zone. Geochemical calculations conducted using MINTEQA2 suggest that the water at this depth is consistently undersaturated with respect to all of the secondary nickel-bearing hydroxide and hydroxysulfate phases included in the MINTEQA2 database. It is inferred therefore that Ni is removed from solution through adsorption or coprecipitation with the secondary ferric oxyhydroxide precipitates. Mineralogical study of the tailings solids indicated that the goethite contained near the surface of the tailings impoundment is Ni bearing (Jambor and Owens 1993), further suggesting that Ni is removed from the tailings pore water through adsorption or coprecipitation reactions.

Modelling Acid Neutralization and Trace-Metal Attenuation

The complexity of the interactions between the large number of dissolved constituents in the tailings pore waters and the large quantity of tailings solids make it difficult to determine *a priori* the changes in pH and metal ion mobility that are likely to occur as sulfide oxidation proceeds. Geochemical modelling of tailings pore water and mineralogical analysis of tailings solids can be used to delineate the principal reactions contributing to pH buffering and metal attenuation (Dubrovsky 1986; Blowes 1990). Incorporation of the observed pH-buffering and metal-controlling reactions into a solute transport model allows calculations to be made to estimate the rate of dissolved metal migration. Using the multicomponent reactive solute transport model MINTRAN (Walter et al. 1994; Frind et al. 1994), simulations were conducted to estimate the migration of low-pH conditions through an underlying aquifer affected by acidic tailings water and their effect on the transport of dissolved metals. MINTRAN couples the geochemical speciation mass transfer model MINTEQA2 (Allison et al. 1990) with an efficient two-dimensional solute transport model (Frind et al. 1990). The model, as applied in this study, includes only equilibrium reactions and is limited, therefore, to conditions where the local equilibrium conditions apply.

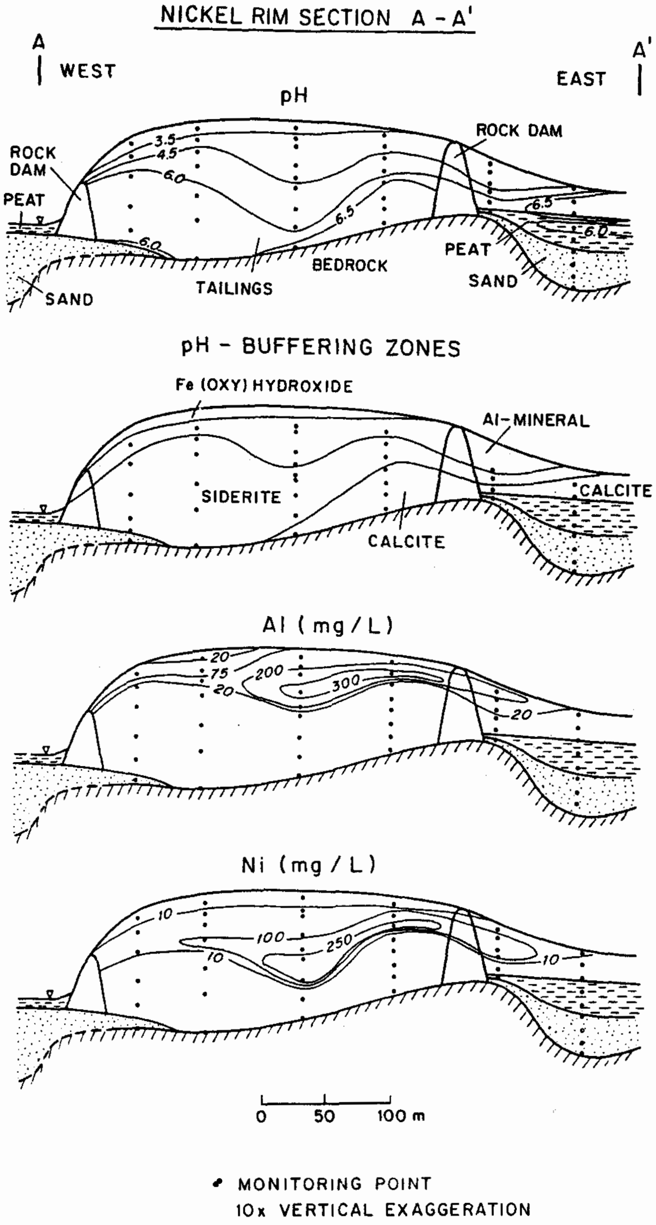

Figure 2. Values of pH, pH-buffering zones, and pore-water concentrations of Al and Ni in the Nickel Rim tailings impoundment near Sudbury, ON.

The simulations were based loosely on the description of the plume of tailings-derived water moving through an unconfined aquifer underlying the Nordic Main uranium tailings impoundment (Morin et al. 1988). The aquifer characteristics and tailings water compositions and background water compositions were selected to be similar to those at the field site. The aqueous components and initial mineral reserves included in the simulation are similar to those observed by Morin and Cherry (1988). The dissolved metals observed at the Nordic site include Al, Fe(II) and Fe(III). Important mineral phases include calcite, siderite, gibbsite, gypsum, amorphous silica and ferrihydrite. To enhance the generality of our simulation, we added higher concentrations of Pb, Cr and Mn and the associated mineral phases anglesite, cerussite, amorphous chromium hydroxide and rhodochrosite. This selection of phases was based on observations we have made at other field sites. In the model simulation, low-pH tailings water infiltrated into the source area of the aquifer for 24 years. The source area covers 10 m < x < 30 m at the watertable. The watertable boundary receives a flux of 0.142 m/yr., the downgradient boundary has a constant head of 13.6 m, and the left and lower boundaries are impermeable. The hydraulic conductivity is 2.5 x 10^{-6} m/s, the porosity is 0.3, the longitudinal dispersivity is 3.00 m, and the transverse dispersivity is 0.03 m.

The results of the simulations (figs. 3 and 4) show the development of a low-pH plume moving outward from the source area. Figure 3 shows contours and vertical profiles at x = 25 m for pH and dissolved Al, Cr and Pb and figure 4 shows solid-phase distributions for the associated mineral phases. The pH plume contains a series of pH plateaus similar to those observed in field studies (fig. 3). Far from the source, the pH ranges from 7.0 to 6.5 and is near equilibrium with respect to calcite, siderite and gibbsite. In this region of the aquifer, calcite is dissolving and siderite and gibbsite are accumulating. Closer to the source, the pH decreases sharply as the calcite initially

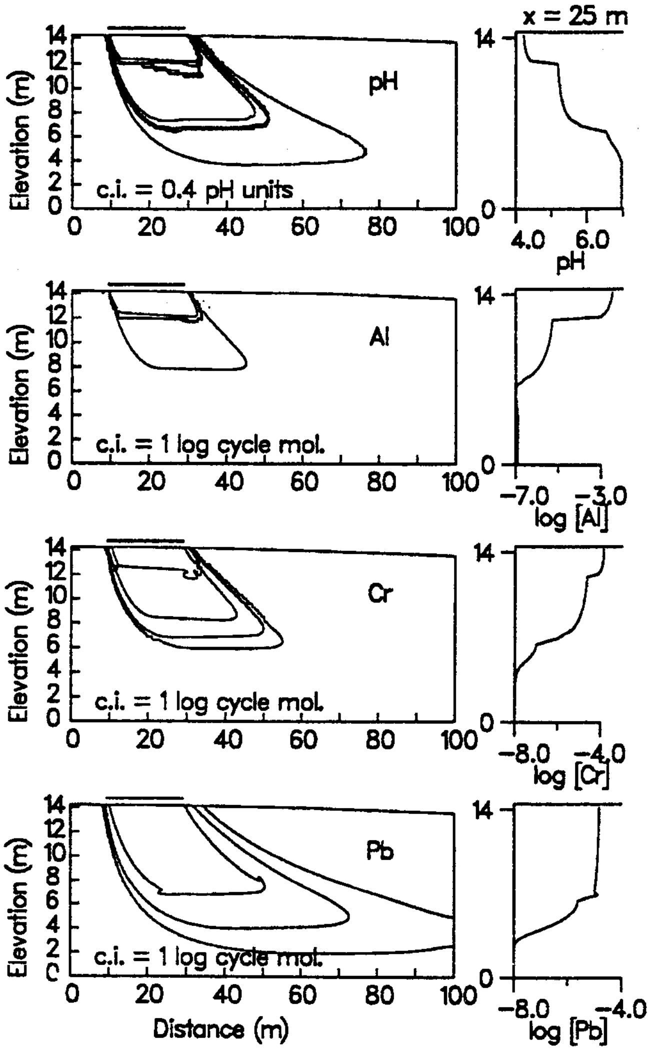

Figure 3. Simulated distributions of pH and concentrations of dissolved Al, Cr and Pb in aquifer receiving acidic drainage from a tailings impoundment at 24 years.

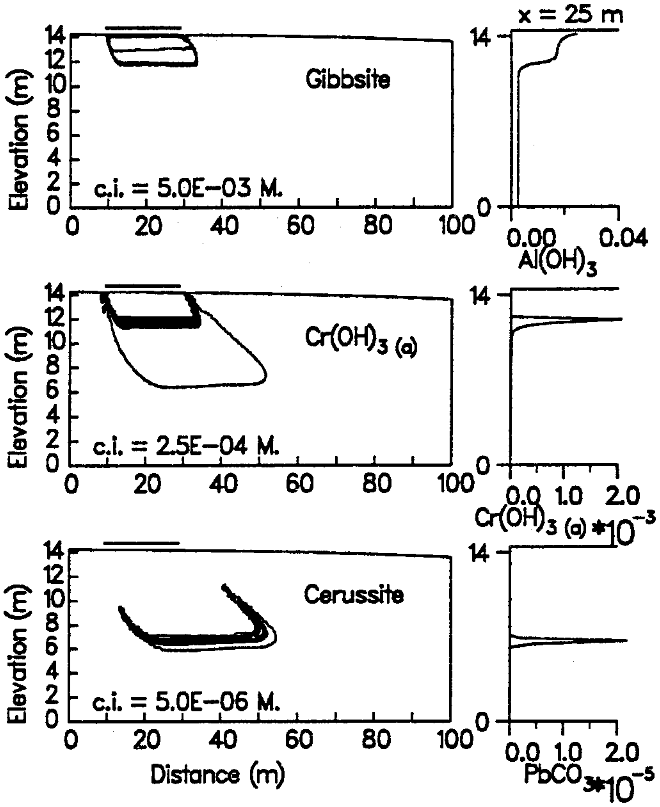

Figure 4. Simulated distributions of solid masses of gibbsite, amorphous $Cr(OH)_3$, and cerussite in aquifer receiving acidic drainage at 24 years.

present in the aquifer is depleted. At this front, the water becomes undersaturated with respect to calcite and the pH decreases from 6.5 to 5.5 (fig. 3) until equilibrium with respect to the second pH-buffering mineral siderite is attained. As siderite dissolves the mass of gibbsite continues to increase. Near the source, the pH decreases again as the primary and secondary siderite present in the aquifer is depleted. At this front, the pH decreases from 5.2 to 4.3 (fig. 3) and equilibrium with respect to gibbsite controls the pH from this point to the source. In this final region, the pH varies from 4.3 to 4.1.

The pH of the plume water affects the concentrations of the dissolved metals Al, Cr, and Pb assumed to be present in the plume. The secondary phases allowed to precipitate and dissolve include gibbsite, amorphous chromium hydroxide ($Cr(OH)_3$), and cerussite ($PbCO_3$). The mobilities of Al and Cr are dependent directly on the pH of the plume water, whereas the mobility of Pb is dependent on the activity of CO_3^{2-}, and therefore, indirectly dependent on the plume-water pH. The dissolved concentrations of Al and Cr show sharp decreases (fig. 3) that correspond with the changes in pH accompanying the siderite and calcite dissolution fronts. These decreases are sufficient to lower the concentration of Cr to below the recommended concentration for drinking water (< 0.05 mg/L; OMOE 1983) and the Al concentration to below the recommended maximum concentration for the protection of freshwater aquatic life. The decreases in dissolved Cr and Al are accompanied by increases in the masses of gibbsite and $Cr(OH)_3$ retained in the aquifer (figs. 3 and 4). The total mass of $Cr(OH)_3$ is much less than the mass of gibbsite; the greatest accumulation of $Cr(OH)_3$ occurs at the siderite dissolution front. Lead is removed from the plume water through the precipitation of cerussite at the calcite dissolution front. Lead concentrations, however, remain above the recommended drinking water limit (< 0.01 mg/L, OMOE 1983) downstream of the cerussite precipitation front. Frind et al. (1994) conducted simulations describing the conditions in the aquifer following the period of sulfide oxidation, representing conditions where sulfide minerals have been depleted or an effective sulfide oxidation control put in place. These simulations show that the low solubilities of the metal-bearing precipitates are sufficient to maintain low concentrations of dissolved metals. The aqueous concentrations of all the metals considered, except Pb, decrease to below current regulatory guidelines shortly after the source of acidic drainage is terminated. These results suggest that metals precipitated in the aquifer are effectively immobilized under these conditions.

Summary and Conclusions

A series of acid-neutralization reactions controls the pore-water pH at four inactive mine tailings impoundments: the Waite Amulet Cu-Zn tailings impoundment (Blowes and Jambor 1990), the Heath Steele Pb-Zn tailings impoundment (Blowes et al. 1991), the Copper Cliff Ni tailings impoundment (Coggans et al. 1994) and the Nickel Rim Ni tailings impoundment (Johnson 1993). The sequence, calcite/dolomite dissolution, siderite dissolution, $Al(OH)_3$ dissolution, and $Fe(OH)_3$ dissolution, is the same as the pH-buffering sequence observed in the Nordic U tailings impoundment near Elliot Lake, Ontario (Dubrovsky 1986) and in a plume of tailings derived water adjacent to the Nordic tailings impoundment (Morin et al. 1988). The sequence of pH-buffering reactions observed at these inactive mine tailings impoundments results in the development of a series of pH regions that controls the mobility of several dissolved trace metals, including Al, Cr, Ni and Pb. The zones of pH buffering and metal attenuation are described well by a model which couples solute transport mechanisms with equilibrium geochemical reactions. Simulations conducted using this model suggest that in tailings impoundments with large masses of acid-consuming materials some of these metals may be retained in the tailings or underlying aquifers throughout the duration of acid release.

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