Arsenic Mobility from Mine Tailings

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Abstract
Arsenic (As) is a common contaminant in some mine tailings, waste rock and effluents resulting from processing of gold and base metal ores. Sources of As in tailings and waste rock are often arsenopyrite and arsenian pyrite present in the ore and wall rocks. During the oxidative dissolution of these sulphide minerals, arsenic is released to the solution and precipitate as arsenates such as scorodite, ferric arsenate and yukonite, or adsorbed onto Fe(III) oxyhydroxides. An understanding of the long-term stabilities of these minerals and compounds, along with arsenopyrite and pyrite, in tailings impoundments and sludge ponds is vital in terms of predicting arsenic releases from the wastes. The paper provides a brief overview of the solubility and stability issues related to common arsenical minerals and compounds and attempts to make an assessment of the controls on arsenic stability and potential releases from mine wastes based on two case studies where arsenic is mobilized at neutral pH and under reducing conditions.

Introduction
Arsenic is a common contaminant in some mine tailings, waste rock and effluents resulting from the processing of gold and base metal ores. Common sources of As include minerals present in the ore and wall rock such as arsenopyrite (FeAsS) and pyrite (FeS_2). Pyrite can contain up to 0.2 mole % As in the structure (Abraitis et al. 2004; Paktunc et al. 2006) especially in refractory gold ores; therefore, it can be considered as another important source of arsenic in mine wastes. Coupled with acid mine drainage, oxidative dissolution of arsenopyrite and pyrite could result in the release of contained As. Common secondary As minerals forming from the oxidation of arsenopyrite and pyrite include arsenates such as scorodite ($FeAsO_4 \bullet 2H_2O$), arseniosiderite ($Ca_2Fe_3(AsO_4)_3O_2 \bullet 3H_2O$), yukonite ($Ca_2Fe_3(AsO_4)_4(OH) \bullet 12H_2O$), and Fe(III) oxyhydroxides such as ferrihydrite and goethite.

Effluents resulting from mineral processing, refining and smelting operations can carry significant concentrations of As and therefore have to be treated before they are discharged to the environment. The Canadian regulatory criterion for As discharges from mine effluents is 0.5 mg/L, defined as the maximum authorized monthly mean concentration (Government of Canada 2002). The regulatory limit of As discharge was reduced from 0.5 to 0.2 mg/L by the new Quebec Directive for new operations (Gouvernement du Québec 2005). Conventional industrial practice for As treatment involves co-precipitation with Fe(III) followed by flocculation and solids removal in a high-density sludge system (Lawrence and Higgs 1999). This process aims to achieve a minimum molar Fe/As ratios of 4/1 to meet the regulatory requirement; however, in practice much higher Fe/As ratios are often needed especially at pH greater than 5.5 (Twidwell et al. 2005). This is the preferred technology because not only it meets the regulatory limit but also it is economical. Widespread availability of ferric iron during processing at many base metal and gold operations provides practical and cost benefits (Riveros et al. 2001). Despite the important practical implications of this technology, the composition of ferric arsenate and the meaning of the Fe/As ratio in terms of controlling As releases have remained uncertain for over three decades. Experimental and advanced characterization studies revealed that ferric arsenate structure is made of short chains of corner-linked FeO_6 polyhedra with bridging arsenate and that the “ferric arsenate” precipitate fixing As at the regulatory level is in fact ferrihydrite (Paktunc et al. 2008). Fe(III) oxyhydroxides including ferrihydrite, goethite and hematite are very effective in

terms of fixing significant As concentrations and that they are generally considered to be stable in oxidized near-surface environments. For instance, goethite and ferrihydrite have been shown to carry As concentrations as high as 20 wt % (Paktunc et al. 2004). Other precipitates that can form during processing and effluent treatment include scorodite, arseniosiderite, ferric sulfoarsenates and jarosite. Jarosite can contain up to about 5.3 wt % As in its structure (Paktunc and Dutrizac 2003). Scorodite is an attractive medium for arsenic control because of its low solubility and its lower ferric iron demand. In addition, some high-temperature ferric sulfoarsenate compounds that can form during pressure oxidation of refractory gold ores can be effective in terms of fixing As (Swash and Monhemius 1994; Dutrizac and Jambor 2007).

Solubilities and long-term stabilities of these As carriers are important in terms of arsenic releases from the wastes. Because the form and occurrence of arsenic in mine wastes are often unknown, predictions as to the long-term stability of As can not be made with a reasonable degree of certainty. In this paper, our current understanding of the solubility and stability of common arsenic minerals will be discussed and an assessment of the controls on arsenic stability and potential releases from mine wastes will be made based on new data from two tailings sites.

Controls on Arsenic Release

Solubility and stability

Scorodite is an effective compound within pH 2 to 5 in relation to the Canadian regulatory criterion for As discharges from mine effluents but its effectiveness becomes limited under the new Quebec Directive (Figure 1). Ferric arsenate is highly soluble with equilibrium As concentrations of 35 to 100 mg/L. Arseniosiderite and yukonite which are common Ca-Fe arsenates forming from Ca-rich effluents (Paktunc et al. 2004) appear to be highly soluble as well based on limited solubility data of Krause and Ettel (1989) and Swash and Monhemius (1994); therefore, these Ca-Fe arsenate compounds would not be suitable for controlling the As concentrations at levels satisfactory for the regulatory criterion.

As shown on Figure 2, scorodite dissolution rates are three to four orders of magnitude slower than the oxidative dissolution of pyrite and five orders of magnitude slower than the oxidative dissolution of arsenopyrite (Paktunc and Bruggeman 2010). In addition to being highly soluble, ferric arsenate dissolution rates are much less than those of scorodite and somewhat comparable to the oxidative dissolution rates of pyrite. In mine tailings, arsenopyrite and arsenian pyrite particles are often enveloped by secondary replacement products of scorodite (Paktunc et al. 2003, 2004; Flemming et al. 2005; Harvey et al. 2006; Moncur et al. 2009) and goethite (Paktunc et al. (2004). In such cases, As releases would be limited to the rate of scorodite dissolution. With substantially slower dissolution rates, scorodite formation around fast-dissolving arsenopyrite and arsenian pyrite particles would be beneficial for long-term As control practices at mine sites with arsenopyrite- and/or arsenian pyrite-rich tailings.

In most Canadian operations, the sludge produced by the ferric arsenate process is stored underwater (Riveros et al. 2001). Provided that the pH of the tailings water is maintained at 4-7 and that the reducing conditions are not established below the surface, the sludge should be considered stable. There are indications that the development of reducing conditions would affect the stability of As compounds. For instance, Martin and Pedersen (2002) reported the release of As up to 8.5 mg/L in the porewaters of Red Lake mine tailings due to the reductive dissolution of As-bearing Fe(III) oxyhydroxides near the sediment-water interface. Furthermore, there are indications of reductive dissolution of jarosite and ferric sulfoarsenates with resultant As releases in the Campbell mine tailings (McCreadie et al. 1998). As a result, it is possible that the current Canadian practice of underwater storage of ferric arsenate sludges may promote the release of As rather than preventing it. Questions remain on the long-term stabilities of arsenates and As-

bearing Fe(III) oxyhydroxides, especially if the favourable conditions change with time including the development of reduced conditions under a water or soil cover.

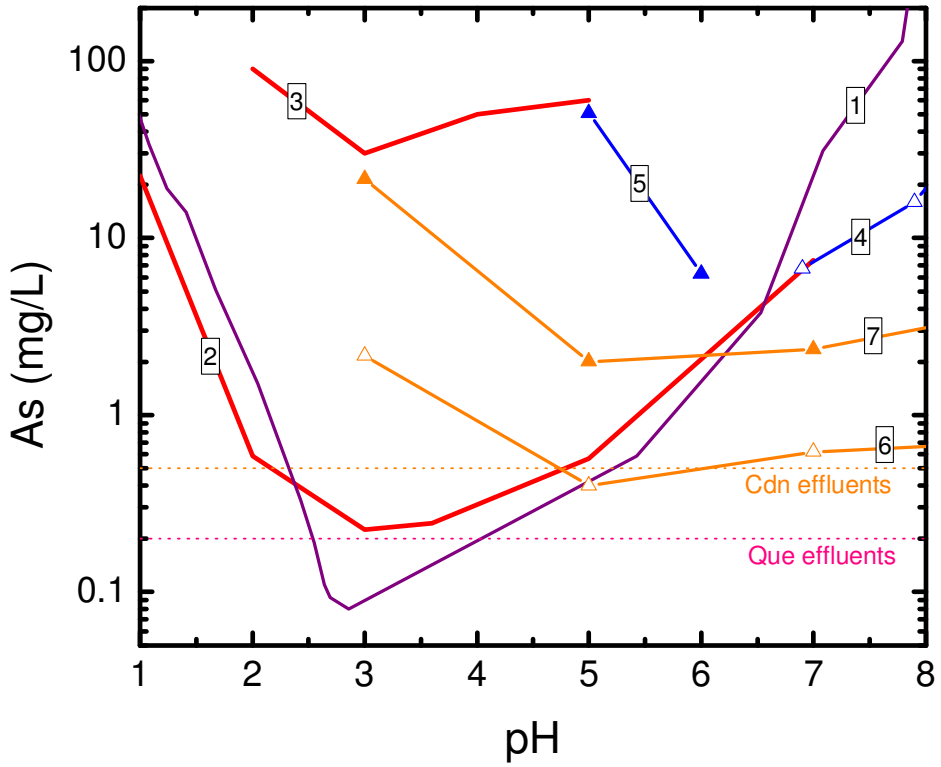

Figure 1. Experimentally-derived solubility curves for common arsenates in terms of As (mg/L) concentrations: 1) scorodite (Krause and Ettel 1989), 2) nanocrystalline scorodite (Paktunc and Bruggeman 2010), 3) amorphous ferric arsenate (Paktunc and Bruggeman 2010), 4) arseniosiderite (Krause and Ettel 1989), 5) yukonite (Krause and Ettel 1989)), 6) arseniosiderite (Swash and Monhemius 1994), 7) yukonite (Swash and Monhemius 1994).

Numerous studies attributed the mobilization of As from natural and anthropogenic sources in subsurface environments to reductive dissolution of Fe(III) oxyhydroxide carriers (e.g. Berg et al. 2001; Harvey et al. 2002; Toevs et al. 2008). Similarly, sub-aqueous placement of mine wastes may result in the mobilization and release of As to the receiving water bodies through reductive dissolution of Fe(III) oxyhydroxides and arsenates. Bacterial reduction of Fe has been shown to be an important biogeochemical process in tailings environments that contain Fe (III) oxyhydroxides (Cummings et al. 1999).

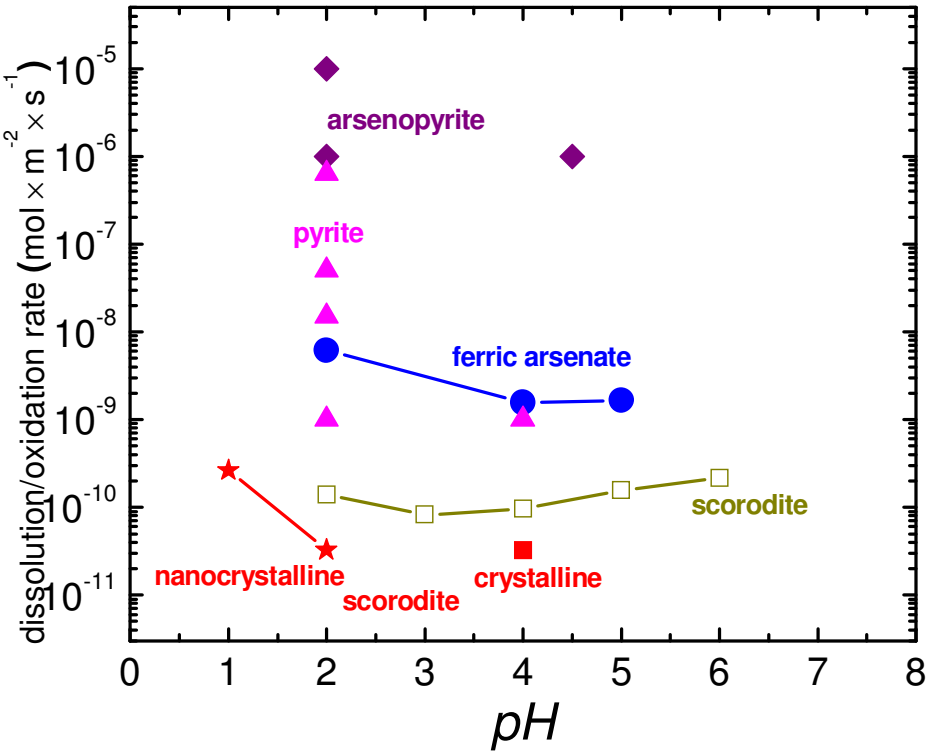

Figure 2. Dissolution rates of crystalline and nanocrystalline scorodite (Harvey et al. 2006; Paktunc and Bruggeman 2010) and ferric arsenate (Paktunc and Bruggeman 2010) as a function of pH and in comparison to oxidative dissolution rates of pyrite (Nicholson 1994; Williamson and Rimstidt 1994; McKibben et al. 2008) and arsenopyrite (McKibben et al. 2008)

Influence of shallow water cover

An assessment of the potential mobilization of arsenic from arsenates and Fe(III) oxyhydroxides influenced by a shallow water cover was made using near surface tailings samples collected from the Ketza River Mine, Yukon. It is a former gold producer operated briefly from 1988 to 1990. The operation was resulted with about 310,000 tonnes of mine tailings containing on average 4 wt % As dominated by arsenates and other secondary arsenic minerals.

In 2006, shallow tailings samples were collected from the exposed and underwater tailings with the intent to examine the mineralogical differences between the exposed and water-covered tailings and the changes in arsenic speciation with depth in the shallow portions of the railings, and study bacterial influences on the mobilization of As from the tailings (Paktunc et al. 2011).

Bulk chemical compositions of the tailings near the surface (0-20 cm) as shown on Table 1 indicate that the tailings are rich in Fe and that the differences between the exposed and water-covered tailings are small. Water-covered tailings appear to have slightly higher As and lower Fe concentrations than those of the exposed tailings.

Table 1. Chemical composition (wt %) of the tailings near the surface

	Exposed	underwater
SiO_2	18.12	20.40
TiO_2	0.03	0.03
Al_2O_3	2.06	2.04
Fe_2O_3(t)	46.95	43.21
MnO	0.20	0.20
MgO	0.78	0.91
CaO	9.02	8.59
Na_2O	0.05	0.05
K_2O	0.41	0.46
S	0.14	0.29
CO_2	7.77	8.38
As_2O_5	4.76	5.82
Total	90.28	90.37

Mineralogical compositions of the tailings are relatively uniform. Dominant minerals include quartz, goethite, calcite, dolomite, muscovite, chlorite and scorodite in both the exposed and underwater tailings. There are no significant changes with depth (0-20 cm) and the mineralogical differences between the exposed and water-covered tailings are negligible. Arsenic carriers include goethite, scorodite, arseniosiderite, yukonite, arsenopyrite and pyrite (Paktunc et al. 2003; 2011). Among these, arsenopyrite and pyrite are minor with combined concentrations of less than about 0.5 wt % in the exposed and about 1 wt % in the underwater tailings. Arsenopyrite and pyrite occur as relict particles often embedded in or rimmed by goethite, scorodite and Ca-Fe arsenates indicating that both are oxidized. Combined calcite and dolomite abundances range from 14 to 22 wt %. With the calculated neutralization potentials of 140 to 239 kg $CaCO_3$ eq/t and the acid generation potential of 4 to 9 kg $CaCO_3$ eq/t, the tailings possess no potential for acid generation.

Characterization of the tailings by X-ray absorption near-edge spectroscopy (XANES) indicated that As is essentially pentavalent in both the exposed and underwater tailings. The dominant As carriers include scorodite, ferric arsenate, Ca-Fe arsenate and goethite as inferred from modeling of the extended X-ray absorption fine structure spectroscopy (EXAFS) spectra (Paktunc et al. 2011).

A modified version of the TCLP tests (USEPA 1992) indicated As releases of about 2 to 6 mg/L gradually increasing with depth in both the exposed and underwater tailings (Figure 3). These changes do not appear to result from the bulk compositions of the tailings since the compositional changes with depth are limited. Corresponding pH values of the TCLP extracts were in the 5-6 range. Leachable As concentrations are comparable to the Ca-Fe arsenate solubility levels and greatly exceed the maximum authorized monthly mean concentration of 0.5 mg/L.

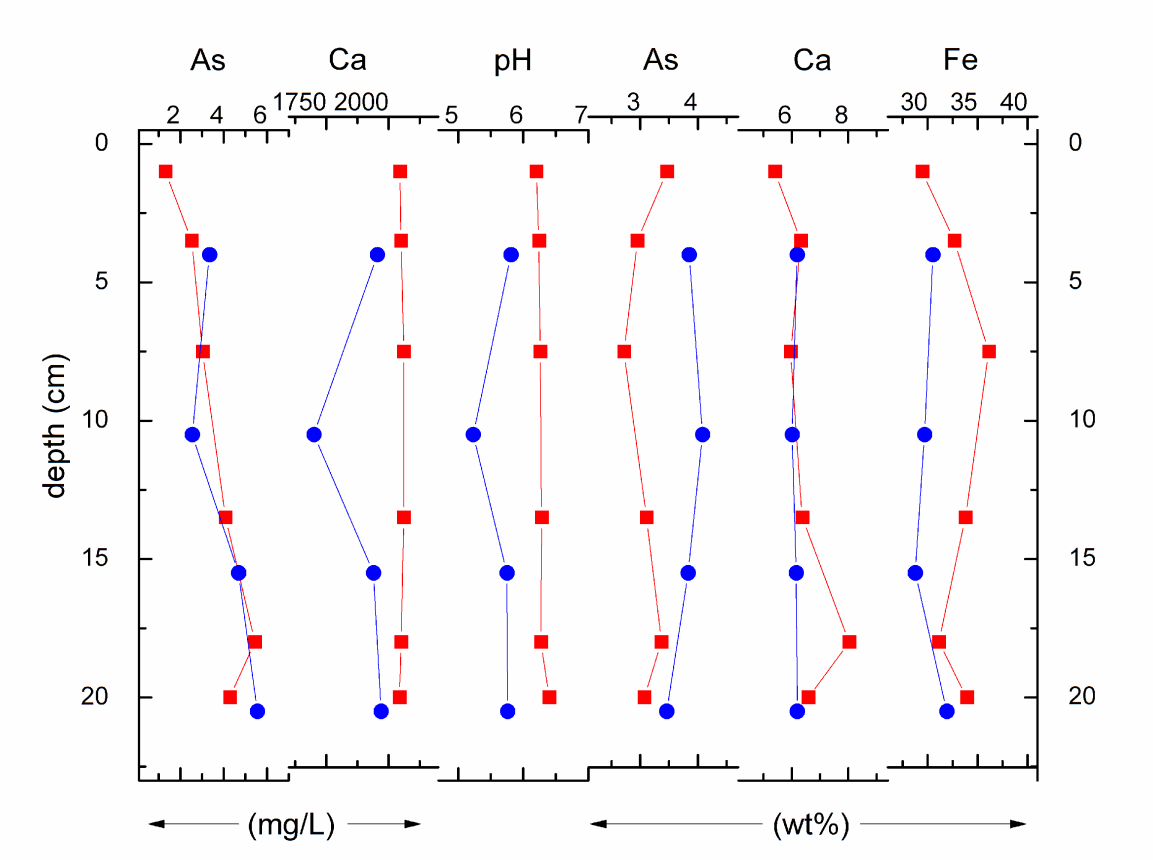

Figure 3. As and Ca concentrations in the leachate (mg/L) along with the corresponding pH value. Also shown are the As, Ca and Fe concentrations (wt %) in the tailings. Square: exposed tailings; Circle: underwater tailings.

Land-based subaerial and subaqueous disposal of tailings containing Ca-Fe arsenates would enhance the mobilization of As which, in the absence of any effluent collection and treatment, has the potential to severely impact the quality of the receiving environment.

Arsenic stability influenced by a biosolids cover

As part of a broader project, Green Mining and Green Energy, several mine tailings sites in northern Ontario are being assessed for their ability to supporting crops for biofuel production without compromising the geochemical stability of the tailings. One of the sites is the Delnite mine tailings near Timmins. The tailings resulted from gold mining and milling operations from 1937 to 1971. There is about 3,000,000 m^3 tailings impounded in an area of about 27 ha with an elevation of 3 to 10 m above the surrounding terrain (Blowes 1990). Following closure, tailings surfaces were amended with limestone and fertilizer, and vegetated.

The tailings contain pyrite and arsenopyrite with potential to generate acidity and release of contained arsenic. In order to assess the potential and integrity of site, several field test plots were established over the mine tailings with biosolids cover of about 1 m thick. After about two years under the biosolids cover, tailings were sampled to about 25 cm depth and characterized with the purpose to determine the changes in the speciation of As and mineralogical composition.

The tailings are mainly composed of the common rock forming minerals including quartz, dolomite, chlorite, albite, muscovite, tourmaline, illite and pyrite. There are no major differences in the mineralogical composition (Figure 4) and bulk chemistry (Table 2) of the biosolids-covered tailings and the control tailings, and no significant variations exist with depth.

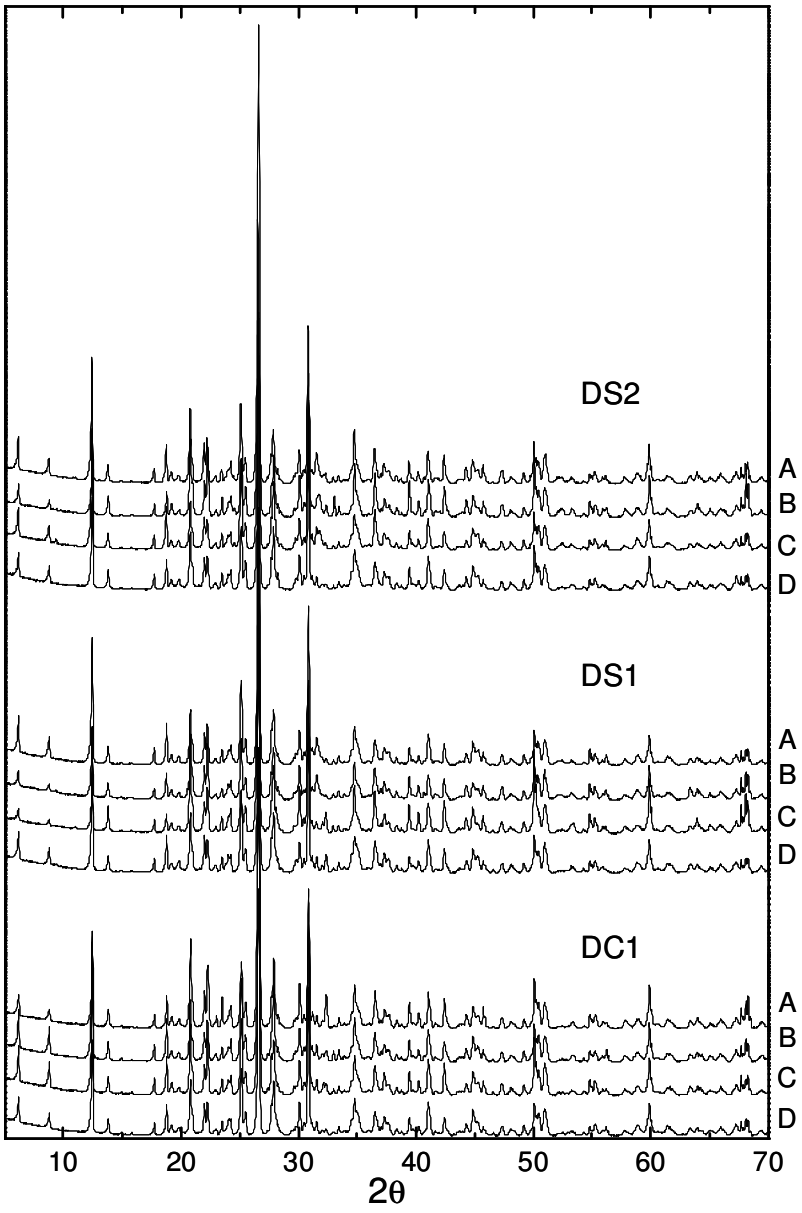

Figure 4. X-ray diffraction patterns of the control (DC1) and biosolids-covered (DS1 and DS2) tailings collected from near the surface (A) to a depth of 20 cm (D).

Table 2. Chemical compositions (wt %) of the near-surface tailings

	DC1	DS1	DS2
SiO_2	42.30	43.17	42.17
Al_2O_3	11.14	10.85	11.23
FeO	10.68	10.78	10.84
MnO	0.16	0.16	0.15
MgO	6.32	6.04	6.12
CaO	5.77	5.50	5.37
Na_2O	1.85	1.85	1.86
K_2O	0.80	0.78	0.81
CO_2	8.85	7.63	7.60
S	0.61	0.70	0.52
SO_4	0.07	0.09	0.10
As	0.24	0.27	0.27
Total	88.79	87.80	87.03

Pyrite quantities are estimated to be 1.2 wt % in the original tailings and 1.2 wt % in the biosolids-covered tailings. Dolomite abundances are 20.6 wt % in the control and 18.6 wt % in the biosolids-covered tailings. With mineralogical neutralization potential values of 173-192 kg $CaCO_3$ eq/t and mineralogical neutralization potential ratios of 10-12, the tailings possess no potential for acid mine drainage.

Arsenic carriers of the original tailings include Fe(III) oxyhydroxide, pyrite and arsenopyrite. Pyrite contains about 0.7 wt % As and Fe(III) oxyhydroxides have highly variable As_2O_5 concentrations in the 1.9 to 22 wt % range (Jambor et al. 1991). Micro-X-ray diffraction studies confirmed that goethite is the dominant Fe(III) oxyhydroxide in the original tailings.

Pyrite is often rimmed by goethite in the control tailings and the deeper portions of the biosolids-covered tailings. In contrast, pyrite particles in the shallower portions of the biosolids-covered tailings are mostly free of secondary oxidation rims, suggesting reductive dissolution of goethite.

Modified TCLP tests indicate that As concentrations in the leachate of the control tailings are below the detection limit of 0.05 mg/L which is an order of magnitude lower than the regulatory criterion for mine effluents (Figure 5). Tailings covered by biosolids on the other hand indicate gradual As increases in the leachate as the biosolids cover is approached. Leachate As concentrations are about 12 mg/L just below the biosolids cover. It appears that the biosolids cover has impacted the geochemical stability of the tailings to depths of about 15 cm. Iron concentrations in the leachate also displayed increases towards the top of the tailings.

As K-edge XANES spectra of the control tailings show that the control tailings are dominated by As^{5+} species and display very little to no change with depth. The biosolids-covered tailings, on the other hand, display changes in As species with depth. The lowermost samples from the biosolids-covered tailings are similar to the control samples but As^{5+} is gradually reduced to As^{3+} as the biosolids cover is approached. This suggests that the reduction front has progressed to depths of about 20 cm within less than 2 years since the establishment of the biosolids cover.

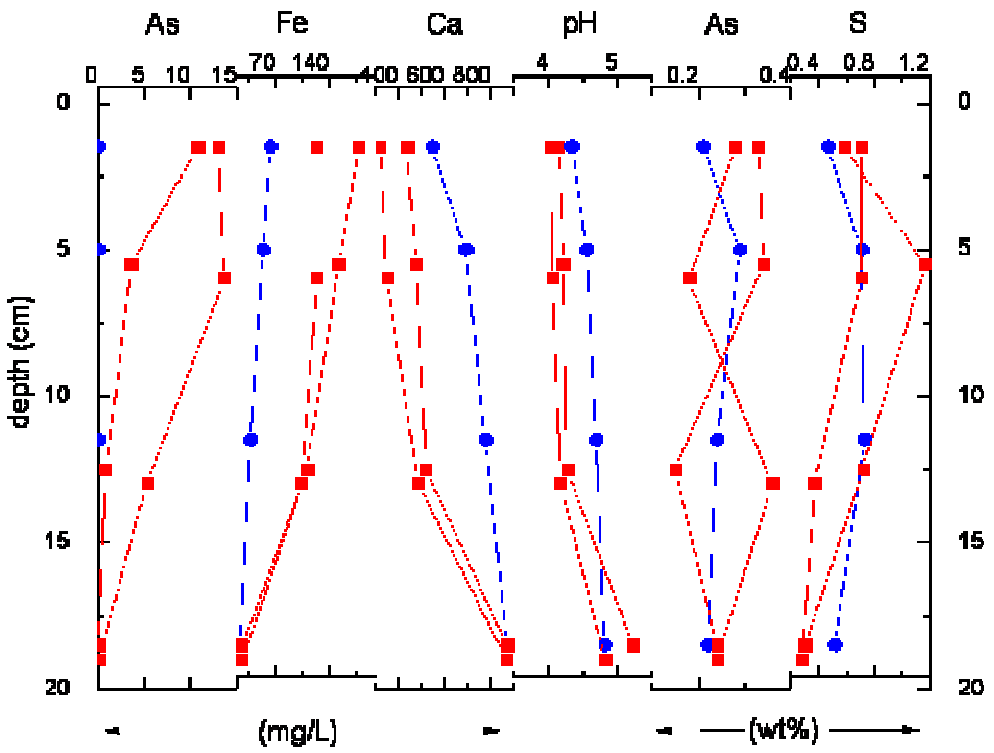

Figure 5. As, Fe and Ca concentrations (mg/L) and pH of leachate resulted from TCLP tests. Also shown are bulk As and S concentrations (wt %) of tailings. Open circles: control tailings; Solid squares: biosolids-covered tailings.

Potential Arsenic Releases from the Ketza and Delnite Mine Tailings

An assessment of the potential As releases from the two tailings sites can be made based on the TCLP test results in comparison to the experimentally-derived solubility data (Figure 6). It should be kept in mind that the TCLP test results do not necessarily represent equilibrium concentrations and as such the assessment should be considered crude. Arsenic and iron concentrations in the TCLP extracts from the Ketza mine tailings seem to indicate controls by ferrihydrite and the pH is buffered by carbonates. Both the exposed and underwater tailings plot along the ferrihydrite solubility in terms of Fe concentrations (Figure 6). Arsenic concentrations straddle the scorodite and perhaps arseniosiderite solubility curves in terms of As concentrations influenced by the incongruent dissolution of scorodite (Langmuir et al. 2006). It appears that regardless of the phases present (i.e. arseniosiderite, yukonite and ferric arsenate), formation of ferrihydrite is effectively controlling the As concentrations. Both the exposed and underwater tailings would produce leachates with As concentrations exceeding the regulatory criterion.

Biosolids cover of the Delnite mine tailings clearly impacted the stability of the main As carrier, goethite in the oxidized tailings. Increased As and Fe concentrations in the leachate, disappearance of goethite rims on pyrite and quantitative changes in the As speciation from pentavalent to trivalent in the tailings immediately below the biosolids cover indicate coupled reduction of Fe(III) and As(V) species. It appears that the biosolids cover influence reached to depths of about 20 cm within 2 years.

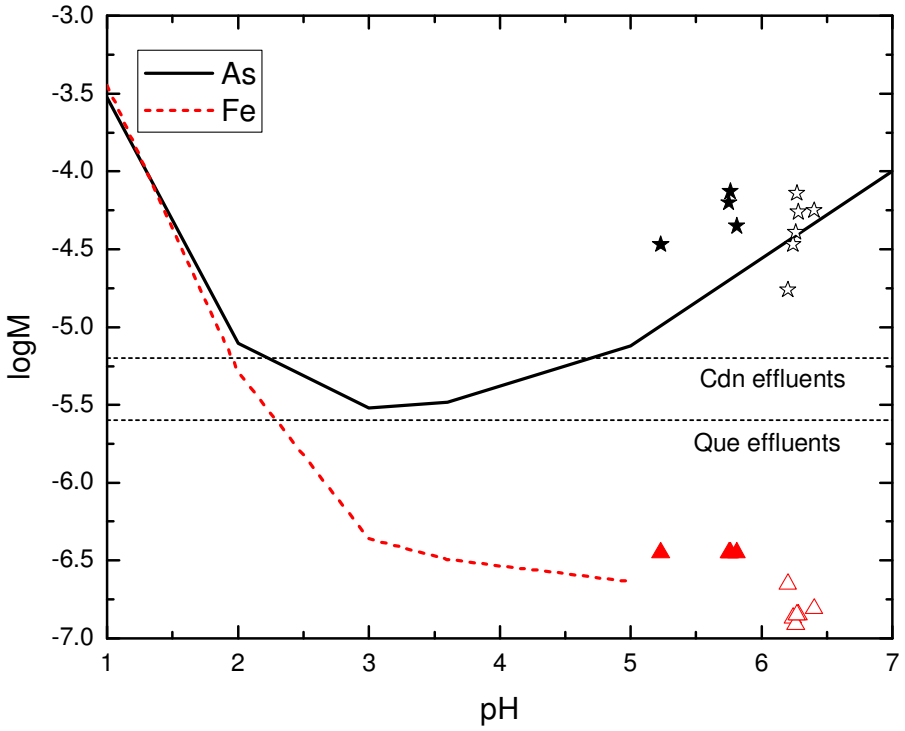

Figure 6. Molar As and Fe concentrations in TCLP extracts from the Ketza River mine tailings shown with the solubility of nanocrystalline scorodite in terms of As (solid) and Fe (dashed) concentrations (Paktunc and Bruggeman 2010). Star: As; Triangle: Fe concentrations; Exposed tailings represented by open symbols and saturated tailings by solid symbols.

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