Geochemical Characterization with High Ionic Strength Methods for Arid-Climate Mines Utilizing Seawater

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Abstract
Mining in extreme arid climates, as found in the Atacama Desert of northern Chile, requires innovative methods for water supply and management. Use of seawater in the process circuit to supplement water supplies may be an option for some sites, particularly for flotation and to transport tailings to storage facilities. The high ionic strength and elevated pH of the resultant process water may result in different geochemical behaviour for materials, in particular with respect to the leaching of metals and other elements due to increased complexation.

Standard environmental geochemical characterization methods (e.g., short-term and long-term leaching tests using relatively dilute lixiviants) may not provide adequate characterization of these situations. This study compares standard testing methods (short-term leaching and humidity cell testing) with modified methods using high ionic strength solutions, and includes a discussion of potential issues. Testing was performed on tailings for proposed copper porphyry mine sites in northern Chile. Use of seawater solutions in testing is common for tailings or mine materials near marine environments; however, in this case the tests must be modified for high ionic strength solutions and evaluated in the context of arid environments with little to no precipitation and high evaporation rates.

Key Words: high ionic strength, metals leaching, modified humidity cell testing, Atacama Desert

Introduction
Several mine sites in the Atacama Desert in Northern Chile use or are evaluating potential use of seawater as process water due to scarcity of water and increasing stress on existing groundwater resources (e.g., Viveros et al. 2009, Philippe et al. 2011). For example, seawater is used for flotation at the Las Luces beneficiation plant in northern Chile (Moreno et al. 2011) and is planned for use at other sites such as the Esperanza site in region II of Chile (e.g., Bernal and Parraguez 2008, Parraguez et al. 2009). Further, saline groundwater is used at several sites in Australia and Chile. At some sites seawater is desalinated by reverse osmosis before use (Petry et al. 2007); however, for cost efficiency, raw seawater is used if resource recovery within the process plant is not materially impacted.

Seawater has an elevated total dissolved solids (TDS) content, with approximately 39,000 mg/L TDS and a resultant ionic strength of approximately 0.7. Use of seawater as a process solution will result in changes, for example through reagent addition, chlorination for biological issues, or pH control in a mill. Furthermore, TDS concentrations in the process water are expected to further concentrate over time as water evaporates and is reclaimed from the tailings management facility (TMF). At the Las Luces mine, seawater is re-circulated from the tailings impoundment and TDS reached 46,400 mg/L after 15 years (Moreno et al. 2011). Higher concentrations are certainly possible depending on the water balance and source waters.

The chemistry and high ionic strength of seawater, and the resultant process water, has the potential to affect geochemical processes. The use of seawater in industrial and mining process circuits is known to increase corrosion, affect scaling properties, result in algae growth, and alter reagent consumption (e.g., Moreno et al. 2011). From a geochemical standpoint, the seawater chemistry and high ionic strength has the potential to affect metals leaching and acid rock drainage (ARD) generation from tailings materials. Studies of the geochemical effects of seawater on acid generation potential and metal leaching potential have been undertaken, though focusing mainly on submarine tailings discharge (e.g., Matthies et al. 2011).

These studies have found that leaching of elements was enhanced by the use of seawater as a flushing medium; however, conditions expected in submarine discharge are very different from those in a TMF where water is re-circulated through time, particularly for oxidation-reduction conditions. In addition, geochemical effects of processing tailings with high ionic strength solutions have been studied from a metallurgical standpoint in Chile and elsewhere in the world (e.g., Liddicoat and Dreisinger 2007, Chadwick 2009, Parraguez et al. 2009).

Studies of geochemical effects and testing of tailings from an environmental standpoint resulting from mineral processing with seawater in extreme arid environments (rather than marine environments) are not common and limited information is available.

In this paper we present results of standard and modified testing for environmental purposes performed in parallel on tailings samples that attempt to account for the expected high ionic strength process solutions. The testing methods are based on modifications to widely used standard methods; modified testing was performed using high ionic strength solutions as lixiviants. This approach was selected over development of new testing methods because a large body of data is available for comparison using the standard methods and because of the wide acceptance of these methods by regulatory agencies. The data is used to identify different geochemical behaviour during leach experiments due to ionic strength effects and various issues related to the approach are discussed, including laboratory analyses.

Methodology

Tailings samples from laboratory scale or pilot scale metallurgical work were tested. The samples represented tailings for two different sites located in regions I and II of Northern Chile. Both sites are located in extreme arid environments with annual precipitation of less than 50 mm per year. Both sites are porphyry copper deposits and represent deposits that will be mined via open pit mining with extraction by flotation, as is common in Chile. The resultant tailings will be transferred to TMFs. Similar to the Las Luces site (Moreno et al. 2011) water in the TMFs will be re-circulated with pH control of the process solution. The tailings samples represent materials that will be sent to the TMF, following all metallurgical processes; as such, this paper only addresses expected behaviour in the TMF, not metallurgical leaching processes.

For both sets of tailings, a complete geochemical testing program was performed following standard guidance (e.g., INAP 2009, MEND 2009). The geochemical testing programs are summarized in Table 1. The focus of this paper is on effects of high ionic strength solutions; as such, details and results of the overall geochemical testing program are only discussed where pertinent for understanding of short-term and long-term leach test results. For Site 1, a sample of whole tailings (i.e., the fraction expected to report to the TMF) for a sulfide ore was tested. For Site 2, samples of whole tailings for a transition ore and a sulfide ore were tested.

Table 1: Summary of Complete Testing Program

Program	Analysis	Site 1	Site 2
Static	Acid Base Accounting (ABA):	X	X
	Neutralization potential	X	X
	Sulphur speciation	X	X
	Paste pH	X	X
	Inorganic carbon	X	X
	X-ray diffraction (XRD) with Rietveld Refinement	X	--
	Particle size analysis	X	--
	Elemental content by acid digestion and ICP-OES	X	X
	Net Acid Generation (NAG)	X	X
	Synthetic Precipitation Leaching Procedure (SPLP)	X	X
	SPLP with synthetic seawater or 3% NaCl solution	X	X
	NAG effluent	X	X
Kinetic	Humidity cell	X	X
	Humidity cell with synthetic seawater or 3% NaCl solution	X	X

For the modified tests, including the short-term and long-term tests, lixiviant solutions applied were either a) a synthetic seawater solution (Site 1) composed of a series of salts (magnesium chloride, potassium chloride, sodium sulphate, sodium bicarbonate, and calcium chloride) or b) a 3 percent (%) sodium chloride (NaCl) solution (Site 2) of approximately 11,000 mg/L sodium and 19,000 mg/L chloride. The approximate composition of the Site 1 synthetic seawater solution is given in Table 2.

Table 2: Synthetic Seawater Content (Site 1)

Constituent	Approximate Concentration (mg/L)
Na	11,500
K	550
Ca	300
Mg	1,300
Cl	19,000
SO_4	2,800
HCO_3	150

Test methods included the synthetic precipitation leaching procedure (SPLP) and the ASTM humidity cell test (HCT) method (EPA 1994; ASTM 2007, respectively). The SPLP test is a screening level test for preliminary evaluation of element mobility from a material. The ASTM HCT, a cyclic kinetic test, was selected primarily given its common use and regulatory acceptability. However, in this case it is assumed to be applicable to site conditions, given that water will be continually re-circulated to the TMF and the potential for drying exists given the extreme arid climate.

Laboratory testing was performed by commercial laboratories in Canada (Site 1) and in Chile (Site 2). Each laboratory has its own quality control and quality assurance program and experience performing both the SPLP and ASTM HCT methods. Laboratory methods for analysis of leachates varied by laboratory and included inductively coupled plasma mass spectrometry (ICP-MS) for Site 1 and both ICP-MS and inductively coupled plasma optical emission spectrometry (ICP-OES) for Site 2. Laboratory methods were adjusted with time for the Site 2 program in order to achieve needed detection limits. As a

consequence, results for the short-term leach testing cannot be compared with Site 1 due to higher detection limits. In addition, detection limits for the long-term leach testing program were improved as the program progressed.

Results

Results of the overall geochemical testing program for tailings materials from both sites indicated that all of the materials have the potential for acid generation based on acid base accounting (ABA) tests (using both net neutralization potential ratio (NPR) and net neutralization potential (NNP) calculations) and the net acid generation (NAG) testing did result in acidic leachate (Figure 1). These results are consistent with expected behaviour for typical copper porphyry deposits (e.g., Plumlee et al. 1999), such as those commonly found in northern Chile. Total sulphur contents range from 1.04 to 1.37 wt% S. The neutralization potential (NP) values range from 4.8 to 9.0 $tCaCO_3/kt$.

Despite the classification of potentially acid generating, none of the tailing samples were initially acid generating, with paste pH values ranging from 6.9 to 8.2. Furthermore, the short-term leach testing and long-term leach testing have not generated acidic conditions to date. At the time of submission, long-term leach testing has been performed for 24 weekly cycles for Site 1 and 34 weekly cycles for Site 2 (all humidity cells are being continued beyond this time). Therefore, while the materials are all classified as potentially acid generating by static testing, a significant lag time to onset of acid generation is expected. As such, the tailings are expected to remain near neutral during operations, particularly given that a portion of tailings will remain saturated during this time and pH control of process solutions are expected. Post-operation, oxidation of some portion of the tailings may occur; however, the lack of precipitation water at these sites will limit transport of ARD from the TMFs. Therefore, the results of the study are applicable to conditions during operations, given the neutral pH conditions of the tests and given that during the operational time period there is a potential for transport of solutions from the site due to the addition of water to the TMF via the tailings slurry.

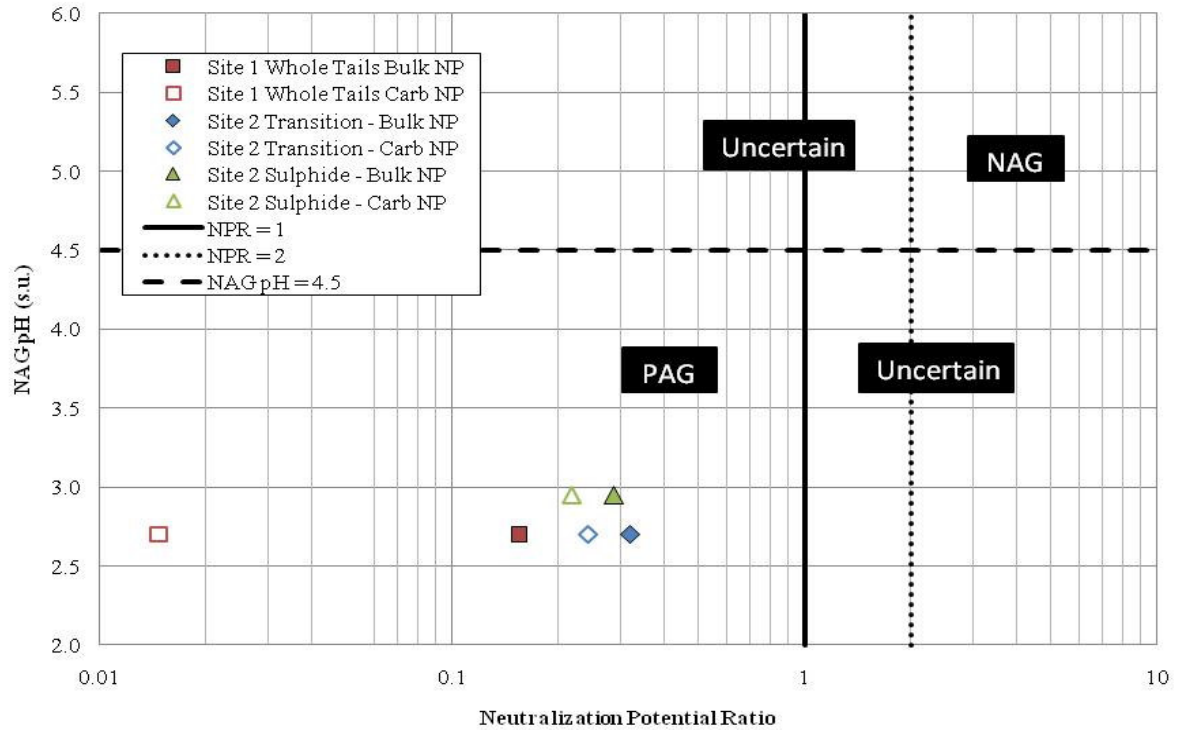

Figure 1. Summary of ABA and NAG Results

Short-term leach testing

For Site 1, results using the standard SPLP and synthetic seawater SPLP are presented in Table 3. Leachate concentrations for short-term leach tests with synthetic seawater are higher for the majority of metals analyzed. With respect to trace metals, antimony, barium, bismuth, chromium, copper, lead, lithium, manganese, nickel, selenium, silver, strontium, tin, and uranium were leached at higher concentrations in the seawater during the short-term leach test (i.e., greater than five times the concentration in the standard test, as some natural heterogeneity and variation is expected). No significant difference or lower concentrations are noted for aluminium, beryllium, cadmium, iron, molybdenum, silicon, and vanadium independent of the expected speciation in solution as cation or anion, respectively.

As noted above, detection limits for the seawater SPLP tests for Site 2 were too high to allow meaningful comparison between the standard and seawater SPLP tests and are being re-analyzed.

Table 3: Results of short-term leach testing for whole tailings sample (Site 1).

Parameter	Units	Standard SPLP	SPLP with synthetic seawater	Seawater leach relative to standard leach
Leachate pH		9.3	7.9	-
Aluminum (Al)	mg/L	0.0534	0.0088	↓
Antimony (Sb)	mg/L	0.0009	0.01	↑
Arsenic (As)	mg/L	0.0009	0.0024	↑
Barium (Ba)	mg/L	0.00621	0.0677	↑
Beryllium (Be)	mg/L	< 0.00002	< 0.00002	--
Bismuth (Bi)	mg/L	< 0.00001	0.0002	↑
Boron (B)	mg/L	0.0503	0.148	↑
Cadmium (Cd)	mg/L	0.000063	< 0.000003	↓
Chromium (Cr)	mg/L	< 0.0005	0.0064	↑
Cobalt (Co)	mg/L	0.00183	0.00518	↑
Copper (Cu)	mg/L	0.0013	0.0191	↑
Iron (Fe)	mg/L	0.003	< 0.002	↓
Lead (Pb)	mg/L	0.00008	0.00053	↑
Lithium (Li)	mg/L	0.003	0.223	↑
Manganese (Mn)	mg/L	0.0053	0.454	↑
Mercury (Hg)	ng/L	< 0.01	< 0.01	--
Molybdenum (Mo)	mg/L	0.0054	0.00539	--
Nickel (Ni)	mg/L	0.0003	0.0308	↑
Selenium (Se)	mg/L	0.00344	0.0197	↑
Silicon (Si)	mg/L	0.89	1.02	↑
Silver (Ag)	mg/L	< 0.00001	0.00004	↑
Strontium (Sr)	mg/L	0.0508	5.43	↑
Thallium (Tl)	mg/L	< 0.0002	< 0.0002	--
Tin (Sn)	mg/L	0.00045	0.0036	↑
Titanium (Ti)	mg/L	0.0004	0.0015	↑
Uranium (U)	mg/L	0.000032	0.00275	↑
Vanadium (V)	mg/L	0.00008	< 0.00003	↓
Zinc (Zn)	mg/L	0.003	0.009	↑

Notes: Elements used in preparation of synthetic seawater are not shown.

Kinetic Testing

Site 1

As noted above, acidic conditions have not been realized in either the standard or synthetic seawater HCTs for Site 1 following 24 weeks of testing. Long lag times are possible for acid generation and, as such, the HCTs are being continued. Leachate pH values for both HCTs fluctuate through time, though remain between 6.9 and 8.3 (Figure 2). Alkalinity concentrations in the standard test have steadily decreased over the 24 weeks, from 160 to 16 mg/L as $CaCO_3$; alkalinity in the synthetic seawater test have remained buffered by the lixiviant near 120 mg/L as $CaCO_3$ (data not shown). A first flush of major cations and anions and trace metals is observed, corresponding with a flush of soluble salts and residual constituents from the seawater used in the pilot plant testing.

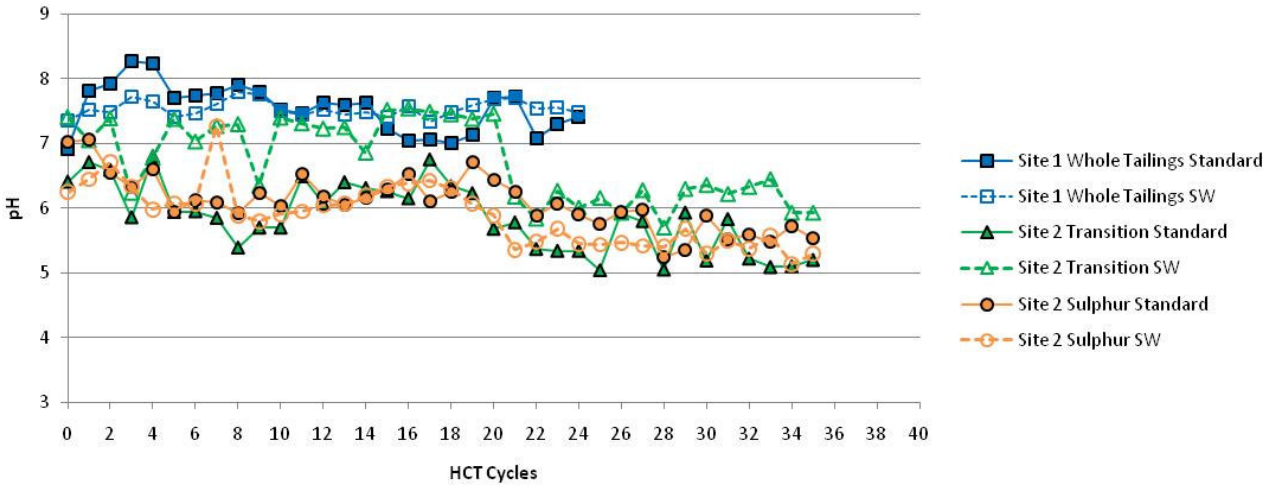

Figure 2. Humidity cell pH values with time

With respect to metals leaching over the long term, metals are generally elevated in the synthetic seawater HCT relative to the standard HCT. Selected metals are shown in Figure 3. Of particular note are metals concentrations that are notably higher in the synthetic seawater HCT relative to the standard test (more than one order of magnitude), including: arsenic, silver, cadmium, chromium, lead, lithium, nickel, strontium, thallium, and uranium.

Site 2

Similar to Site 1, acidic conditions have not been realized in either the standard or HCTs with 3% NaCl solution (NaCl HCT) for Site 2 following 34 weeks of testing. Long lag times are possible for acid generation and the HCTs are being continued. Leachate pH values for all HCTs were initially between pH 6 and 7.5, though beginning in week 20 the pH values have shifted and currently range between 5 and 6.4 for all four tests (Figure 2). Leachate alkalinity is less than 10 mg/L as $CaCO_3$ in all tests.

With respect to metal leaching, several different trends are identified. Following 34 weeks of testing, some metals are elevated in the NaCl HCTs relative to the standard HCT for both the transition and sulphide tailings, including barium and silver (Figure 3). Cadmium and nickel are elevated in the NaCl HCT relative to the standard test for the sulphide tailings only. Cobalt, copper, uranium, and zinc, are also elevated in the NaCl HCT relative to the standard HCT for the sulphide ores, yet show opposite behaviour for the transitional tailings, where concentrations in the NaCl HCT are lower than the standard HCT (Figure 3). Molybdenum is elevated in the transitional NaCl HCT relative to all of the other HCTs (Figure 3).

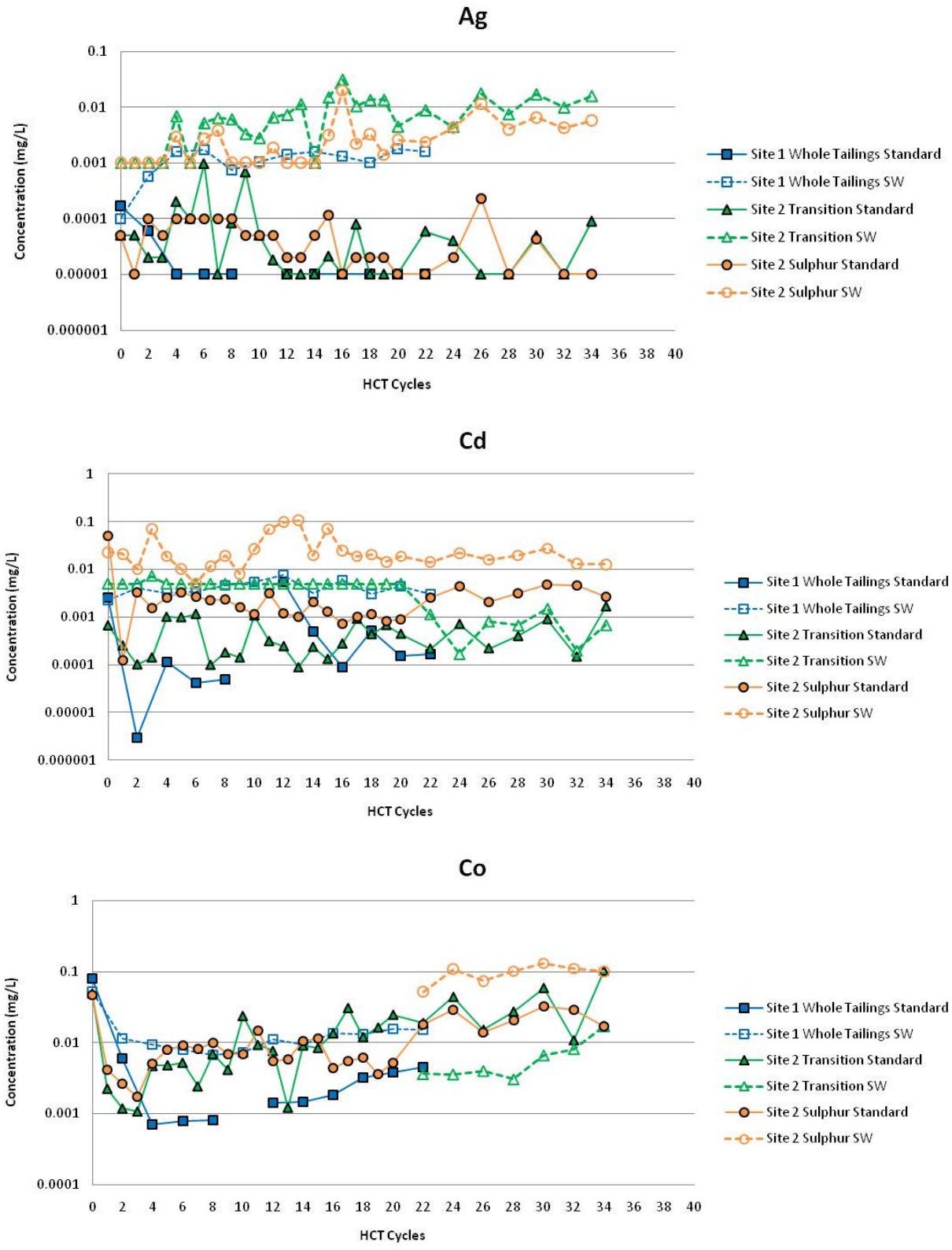

Figure 3. Selected humidity cell results

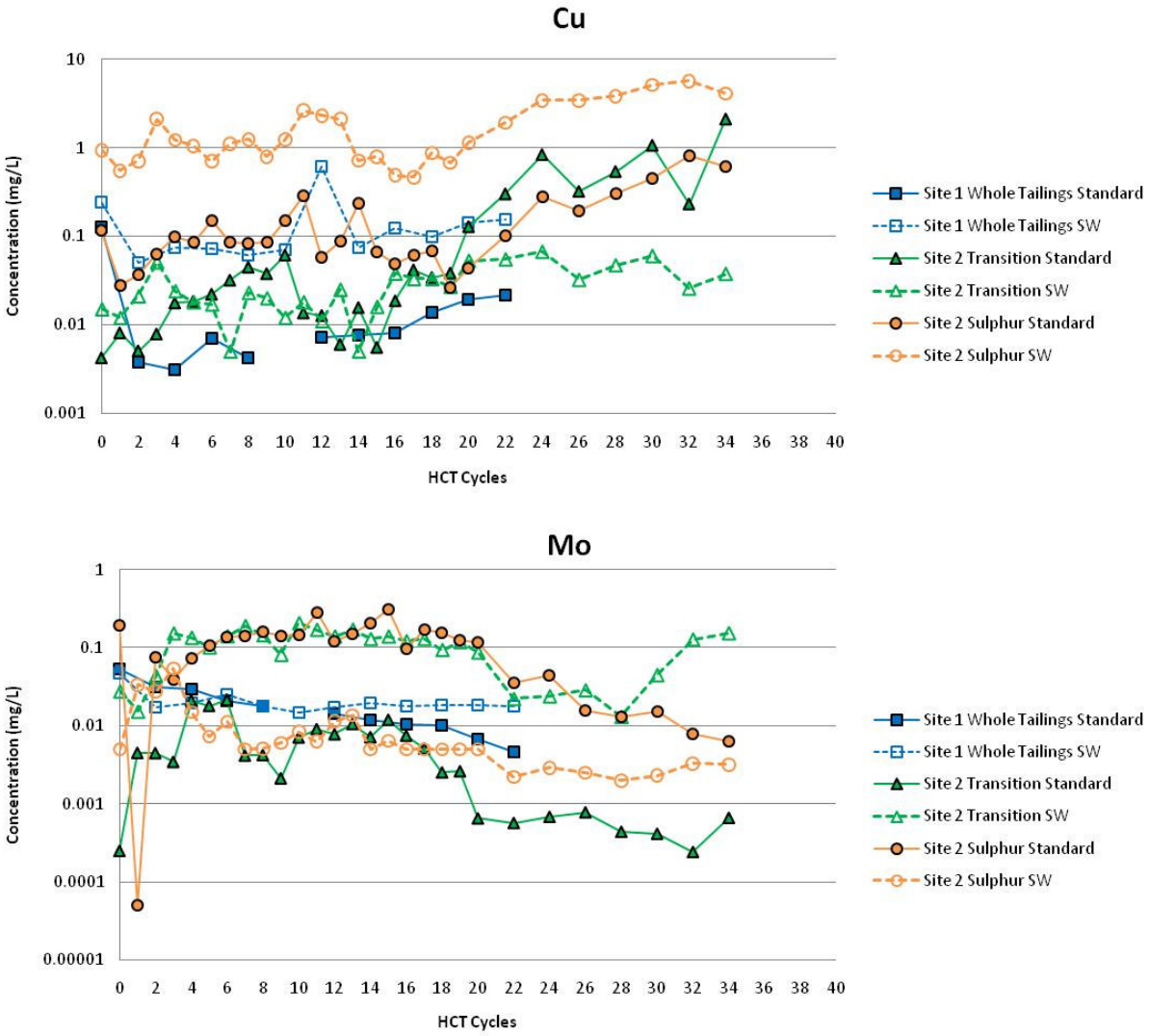

Figure 3 continued. Selected humidity cell results

Discussion and Further Considerations

Effects of Seawater on Metals Leaching

The leaching of several metals and elements increased using high ionic strength lixiviants; however, for some other elements leaching was unaffected or decreased. The increased leaching may be due to a combination of mechanisms. For example, at high concentrations of chloride dissolved metals form complexes in solution, enhancing the amount of metal that can be dissolved at equilibrium. Chloride complexes may remove metals that have adsorbed to mineral surfaces, increasing the metals in solution. Alternatively, chloride may undergo specific adsorption or interact with mineral surfaces and play a mechanistic role in increasing dissolution rates (Awakura et al. 1980). Ligand-promoted dissolution of oxides and sulphides is possible. For oxides, a ligand forms a surface complex by ligand exchange with surface hydroxyl-groups. The ligand-surface complex shifts electron density towards the metal ion, lowering the Lewis acidity of the metal ion. This polarizes the metal-oxygen bond, allowing detachment of the metal from the surface (Furrer and Stumm 1986; Stumm 1992). For sulphides, chloride has been found to increase non-oxidative dissolution rates of monosulphides (Awakura et al. 1980; Lotens and Wesker 1987). Other ions in solution will also play a role through complexation or competition for surface sites or ligands in solution.

Given mechanisms such as chloride complexation or ligand-promoted dissolution described above, enhanced metals leaching would be expected for the soft or B-type metals (Pearson 1963, Stumm and Morgan 1996) by a seawater or NaCl solution. For the three pairs of humidity cells presented, this is the case for soft metals such as lead and silver (Figure 3), but other soft metals have inconsistent behavior, in particular for the transitional tailing sample (Site 2). For example, cadmium, cobalt, copper, and zinc are higher in the seawater and NaCl HCTs relative to the standard test except for the transition sample, where leachate concentrations are lower for these metals in the NaCl HCT. Other inconsistencies with the complexation or ligand-promoted dissolution also exist such as in the case of barium, which is not considered a soft metal, yet has enhanced leaching in the HCTs with high ionic strength solutions.

The discrepancies above indicate that other factors, such as mineralogy, alteration, mineral availability, or other weathering factors play a role in metals leaching. The sulphide and transitional tailings have different mineral assemblages, consistent with their origin, that affect leaching behaviour. In addition, these samples may have been affected differently by metallurgical processes prior to this testing program. A detailed chemical and mineralogical examination, element by element, and resolution of the specific discrepancies is beyond the scope of this paper and beyond the scope of typical geochemical characterization studies; however, these results support the need to perform this type of testing to identify potential metals mobility. Simple assumptions based on chemical principles that metals leaching will be affected cannot predict all of the various mineral and weathering interactions.

Recommendations for Future Testing Programs

The results of parallel testing of tailings with standard and high ionic strength solutions indicate that some metals and other elements may be mobilized to a greater degree and some to a lesser degree due to the use of seawater for processing. As such, sites expecting to use seawater in processing may need to consider additional environmental testing to understand potential impacts. We offer the following recommendations for future testing programs.

- HCT testing with high ionic strength lixiviants provides more conclusive information relative to short-term leach testing. Results of the short-term leach testing do not likely provide sufficient useful information to justify use alone; in this case they accurately indicated issues associated with manganese, but not selenium or other metals. In addition, if acidic conditions occur, the short-term tests did not indicate this potential; however, the tests serve two purposes in this particular case: 1) a screening level test, similar to standard geochemical characterization

programs and 2) a preview to working with the laboratory to identify potential issues, such as detection limits, that may arise in use of high ionic strength solutions.

- Use of simple high ionic strength solutions, such as a NaCl solution, is preferred over synthetic seawater for several reasons:
    - The simple solution is easier for the laboratory to prepare consistently over time; true seawater may be affected by fouling after keeping it for weeks in the laboratory.
    - Interpretation of results is more complicated using synthetic seawater, for example: depletion calculations using sulphate cannot be used, alkalinity from the synthetic solution may buffer acid generation, and use of a range of salts in synthetic seawater may allow for a greater range of impurities to be introduced.
    - Solutions of ionic strength greater than that of seawater may be prepared without complication, which may be more appropriate for arid environments where evaporative concentration is expected.
    - Use of a simple high ionic strength solution may not identify interactions with other components of real seawater, such as sulphate, alkalinity, or biological components. However, this trade-off must be evaluated with the advantages listed above as well as the extent to which the seawater will be modified to serve as a process solution (e.g., pH control or control of biological issues). Preparation of a representative long term process solution is challenging at the laboratory scale.
- Residual seawater or evaporative salts may be present from the pilot plant and should be accounted for in short-term and long-term testing.
- Use of a modified standard method, rather than development of a new method, is appropriate for a large scale mining project where many samples need to be analyzed. Use of a modified standard method facilitates both interactions with the analytical laboratory (which may not be able to accommodate novel methods easily or effectively) and facilitates acceptance by the regulatory agencies.

In addition to the points above, this type of testing program reinforces the need for good communication and teamwork with an analytical laboratory, particularly in locations where experienced laboratories are not available. Considerations for communication with the laboratory include lixiviant composition and preparation, potential for contamination through leaching of glassware by the high ionic strength solutions, and selection of analysis methods and respective detection limits that are appropriate for high ionic strength solutions.

Conclusions

Based on a comparison of accepted testing methods using standard lixiviants and high ionic strength lixiviants, use of seawater as a process solution may affect mechanisms of the release and availability of metals and other elements. The specific mechanisms affecting leaching will vary by element, mineralogy, and other factors. Coinciding with this, the observed magnitude to which leaching was enhanced or inhibited in the high ionic strength lixiviant tests varied by metal/element; for some key metals/elements, increases were significant in that they may affect predictions of water quality impacts. For other metals the resultant increases would have little overall effect on water quality predictions. Furthermore, it is important to recognize that changes in leaching behaviour alone do not indicate that environmental regulatory criteria will be exceeded, as a number of other factors, such as hydrogeology and susceptibility of receptors, need to be considered. In addition, overall increases in salinity and TDS may need to be considered.

These results are based on accepted testing methods with modified lixiviant through the use of a high ionic strength solution. Use of these standard methods in this program was chosen to facilitate regulatory acceptance, laboratory capacity to manage non-standard approaches, and comparison to accepted and available information.

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Acronyms

ASTM	American Society for Testing and Materials
EPA	United States Environmental Protection Agency
HCT	Humidity Cell Test
ICP-MS	Inductively Coupled Plasma Mass Spectrometry
ICP-OES	Inductively Coupled Plasma Optical Emission Spectrometry
INAP	International Network for Acid Prevention
MEND	Mine Environment Neutral Drainage
NAG	Net Acid Generation
NPR	Neutralization Potential Ratio
SPLP	Synthetic Precipitation Leaching Procedure
TDS	Total Dissolved Solids
TMF	Tailings Management Facility
XRD	X-Ray Diffraction

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