

Developing a Site-Specific Effective Neutralization Potential to Acid Potential Consumption Ratio: Establishing the Boundary in Sulphidic Tailings

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ABSTRACT

Acid rock drainage (ARD) is a risk to water quality at mines that are associated with waste materials containing sulphide, such as tailings. The classification of mine materials as potentially acid generating (PAG) or non-potentially acid generating (non-PAG) is typically based on the ratio of effective neutralization potential to acid potential (NP/AP) in a sample as well as the rate at which the neutralization potential (NP) is consumed during neutralization of the acid produced by the oxidation of the sulphide minerals, representing the acid potential (AP). While a conservative NP/AP ratio (NPR) of 2 or 3 is typically recommended to account for some uncertainty in the neutralizing reactions (Price 2009), site-specific values can be developed and justified based on appropriate evidence. The NPR for a mine material is not only important in the classification as PAG or non-PAG, the value can also represent the ratio of the NP consumed relative to the AP reacted during sulphide mineral oxidation and accompanying neutralization reactions. The NP/AP consumption ratio is required to model NP consumption rates and to forecast the time to the onset of acid generation in PAG materials and as such, this ratio is important for mine material management. The methodology in this study implicitly determines the effectiveness of the NP at a case study mine site and applies this information to establish the non-PAG boundary in a sulphidic tailings and the approach can be applied at other sites. Lab-scale and field-scale static and kinetic tests results were assessed to evaluate a site-specific NP/AP consumption ratio. The data were screened to ensure that the interpretation of the carbonate molar ratio (CMR) was represented in tests with the appropriate geochemical conditions. Data from the tests indicated that the average CMR from the applicable tests for sulphide tailings was 1.07, and a conservative upper-bound value of 1.2, representing the highest average value identified in one of the kinetic tests, was selected to conservatively represent the NP/AP consumption ratio for site tailings. This representative value was proposed and accepted as a permit criterion to define non-PAG and the value was then applied in long-term oxidation modelling to forecast tailings oxidation risks of acidification and water quality effects through operations and post-closure.

Key Words: ARD, carbonate molar ratio, modelling, forecasting, acidity, NP/AP consumption ratio

INTRODUCTION

Acid rock drainage is a risk to water quality at mines that are associated with waste materials (e.g., waste rock or tailings) containing sulphide. Assessments for ARD include acid base accounting that considers the acid potential (AP) represented by oxidation of all iron sulphide minerals such as pyrite and the neutralization potential (NP) represented by minerals that can neutralise acid to circum-neutral pH values. The neutralization potential ratio (NPR) is defined as the ratio of NP/AP available in the solid mine materials and is often applied as a classification criterion to define the boundary between PAG and non-PAG tailings. In theory, if the rate of AP consumed during acid production is directly proportional to the rate of effective-NP consumption, then a NP/AP ratio of 1 will ensure that drainage from the material will remain neutral and the material can be classified as non-potentially acid generating (non-PAG). Some neutralizing reactions consume effective-NP at two times the rate as the AP consumed and if only those reactions occurred, an NP/AP ratio of 2 would be required to ensure that the materials will remain neutral in perpetuity and to be classified as non-PAG. In reality, the effective-NP/AP criterion for a non-PAG classification of mine materials will always be between 1 and 2.

NPR criterion values, sulphur content inventories, and net neutralization potential inventories have been historically used as a generic comparative values based on static testing to define a classification boundary for non-PAG materials, in the absence of other information, to reduce the risk of PAG material being misclassified as non-PAG. However, a site- or material-specific NPR criterion can be developed with appropriate supporting kinetic test data on the materials of interest.

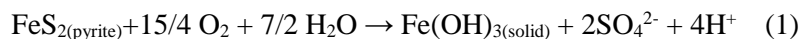
Determining an applicable NPR for a mine materials is not only important in the classification of the material as PAG or non-PAG, but the representative NPR value also represents the ratio of the NP consumed relative to the AP reacted during sulphide mineral oxidation and accompanying neutralization reactions. These terms are critically responsible for long-term forecasting of the site geochemical conditions and resulting net-alkalinity or net-acidity loads from ongoing oxidation. As such, the quantification of a material and site-specific NP/AP consumption ratio represents a critical operational decision point for site managers to appropriately contextualize the risk inherent to site water quality and informs both operational and closure implications to manage and mitigate risk of short- or long-term acid production.

This paper provides the basis for, and a resulting site-specific NP/AP criteria value to define non-PAG tailings at a tailings storage facility with a temperate climate in Canada. This assessment moves beyond using summary static results in comparison to generic criteria to instead use site- and material-specific considerations to characterize the tailings' potential for acid generation and evaluates the observed NP/AP consumption ratios from appropriate site-specific kinetic tests.

SITE TAILINGS CHEMISTRY AND THEORY

To appropriately contextualize the site-specific NP and AP contents the site tailings must be robustly characterized with a thorough understanding of the of the site-specific facility NP values – recognizing that the facility NP values consider the geochemical effectiveness of the NP inventories on-site as opposed to the generic sample bulk-NP values.

Conceptually, acid is produced by iron sulphide mineral oxidation as shown in Reaction 1 when the pH is greater than about 4, where H^+ represents the acidity formed and pyrite represents the primary source of sulphur in the site tailings. In acid base accounting terms 1 molar quantity of sulphur produces 2 molar quantities of H^+ for pyrite oxidation and is equivalent to 1 unit of (AP) represented in units of $kg_{CaCO_3}/tonne$.

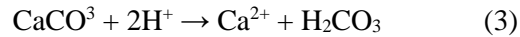


The acid will be consumed and neutralized by calcium and magnesium carbonate minerals, when present and these are the dominant sources of NP in the site tailings. For calcite ($CaCO_3$) as an example, neutralization can occur according to the following reactions (Reaction 2), producing predominately bicarbonate ions (HCO_3^-) when the pH is greater than about 6.4. Bicarbonate represents the main component of alkalinity in natural waters.



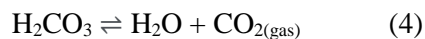
In this reaction, 2 NP units are consumed for each 1 AP unit (or $2H^+$) reacted for an effective NP/AP consumption ratio of 2.

The following reaction can also occur (Reaction 3), consuming $2H^+$ and producing predominantly carbonic acid (H_2CO_3) and no alkalinity when the pH is less than about 6.4. In this neutralization reaction, an equivalent of 1 NP is consumed for each 1 AP (or $2H^+$) reacted resulting in an effective NP/AP consumption ratio of 1.



In waste rock and tailings environments, both neutralization reactions represented by Reaction 2 and Reaction 3 will be occurring, and as such, the actual ratio of NP consumed during neutralization to AP consumed during sulphide oxidation and acid generation is expected to be between 1 and 2 (Price 2009). The precise mix of neutralization reactions is dependent on the existing physico-chemical conditions and therefore there is “uncertainty” regarding the actual potential for acid production when the NP/AP inventory ratios in sulphide waste are between 1 and 2. Complete neutralization is theoretically assured for mine materials with effective NP/AP inventory ratios greater than 2. The potential for acidification exists in the absence of mitigation, for materials with NP/AP inventory ratios of less than 1. This is the basis for the guideline values for NP/AP of 2 or greater to classify sulphide mine materials as non-PAG

One additional reaction that plays a role is the dissociation of carbonic acid (H_2CO_3) to form dissolved carbon dioxide (CO_2) and water as in Reaction 4.



This allows for the degassing of some carbon dioxide into air if a gas phase is present as is the case in well-drained waste rock. Less degassing is expected in moist tailings with limited gas phase and interconnected gas voids than in a well-drained rock pile with gas filled voids. If carbon dioxide degassing occurs, there will be a tendency for the pH of the water to increase and therefore the ratio of bicarbonate (HCO_3^-) to carbonic acid will increase, representing some neutralization with Reaction 2. This suggests that we may expect Reaction 3 to be more dominant than Reaction 2 in moist tailings than in well drained waste rock. In turn this implies that the NP/AP consumption ratio or carbonate molar ratio (CMR) would be expected to be lower in tailings than in waste rock composed of similar geologic materials.

The neutralization reactions discussed above considered calcite (CaCO_3) as the carbonate mineral. However, other carbonate minerals can also represent neutralization potential, including dolomite ($\text{CaMg}(\text{CO}_3)_2$) and siderite (FeCO_3). The effective NP component of carbonate minerals is represented by the calcium (Ca) and magnesium (Mg) but not by the iron (Fe) fractions of the carbonate minerals. The iron carbonate, when dissolved, will store the latent acidity and release that acidity when those elements are oxidized and the oxides precipitate. Therefore, the Fe fractions of carbonate minerals are not considered as effective NP and should not be included in acid base accounting calculations. In the site tailings under consideration in this assessment, the dominant carbonates are calcite and dolomite with trace siderite.

Carbonate Molar Ratio

The actual ratios of NP consumed to AP reacted can be assessed by evaluating the chemistry of the contact waters for materials that are undergoing sulphide oxidation and neutralization reactions. For calcium and magnesium carbonates such as calcite and dolomite, the molar concentrations of calcium (Ca) plus magnesium (Mg) will represent the NP dissolved or consumed and the molar concentrations of sulphate (SO_4) represent the AP reacted. Sulphate concentrations represent the oxidation and AP consumption rate if there are no interferences with either sulphate or calcium concentrations as discussed further below. The $(\text{Ca}+\text{Mg})/\text{SO}_4$ molar ratio, also referred to as the CMR, in the contact water from kinetic tests provides a measure of the effective-NP/AP consumption ratio and represents the actual NP/AP inventory ratio criterion that can be used to classify materials as non-PAG for the site.

Site Tailings Chemistry

Static test characterization for the site has shown that total carbon and total sulphur were excellent surrogates for effective-NP and AP inventory estimation, respectively, with no significant organic carbon

or sulphate content in the tailings – allowing for an expanded dataset for interpretation though this is not a pre-requisite to the application of the interpretive methodology presented herein.

The carbonate neutralization potential (Carb-NP), as measured by pyrolysis or inferred from carbon contents as quantified by LECO combustion and infrared detection, was also found to represent a more conservative estimate and effective source of neutralization potential than the Modified Sobek-NP (Lawrence and Wang 1997) content in site tailings. For reference, from Rietveld XRD it has been identified that the NP source in the tailings is calcite (CaCO_3) and dolomite ($\text{CaMg}(\text{CO}_3)_2$) with trace siderite (FeCO_3). Whereas the sources of sulphide-AP are primarily pyrite with trace chalcopyrite and sphalerite. Average tailings sulphur contents are marginally greater than 1% with a median Carb-NP/AP ratio of approximately 1.3.

The neutralizing reactions that consume the carbonate minerals produce calcium and magnesium with the potential to produce minor quantities of iron if siderite is present. Although there may be some silicate minerals that can consume acid and represent some portion of the NP, these sources are commonly slower to dissolve and have been identified to represent only approximately 10% of the tailings neutralization in kinetic tests. In this assessment, it was conservatively assumed that only calcium and magnesium carbonates represented effective-NP.

This case study assessment moves beyond using summary static geochemical results in comparison to generic criteria to instead use site- and material-specific considerations to characterize the tailings' potential for acid generation and evaluates the observed NP/AP consumption ratios from appropriate site-specific kinetic tests based on site mineralogy and water chemistry. This overall methodology can be applied at other sites to identify a site-specific NP to AP consumption ratio that can be used to establish the material and site-specific non-PAG criterion and to support oxidation modelling.

Literature Review - CMR Interpretation

A brief literature review discussing CMR results from kinetic tests at various third-party sites and various test-scales as well as their implications on mine material CMR quantification and non-PAG classification is presented here. The theory briefly presented above and the results from waste rock-focused assessments are expected to be consistent for CMR evaluation in both waste rock and tailings and would apply at the case study site discussed herein:

- Mattson (2005)
 - Presents and discusses CMR results from humidity cell tests on low-sulphide rock samples. Identified that lower sulphate concentrations in the humidity cell leachate was attributed to the equilibrium dissolution of carbonate minerals at the comparatively high water to solids rinse ratios (W:S) of a humidity cell test. The measured calcium and magnesium in the leachate concentrations were influenced by their solubility rather than their release from reaction with the acidity formed by sulphide oxidation. As such, it was concluded that the CMR values from humidity cell tests may not be representative of the material CMR at low sulphate concentrations.
- SRK (2011)
 - Summarized the theoretical basis for a Carb-NP/AP criterion of 1.5 to be used for PAG tailings classification at a third-party site considering the following lines of evidence: 1) the theoretical NP/AP for tailings is known to be less than 2, 2) the NP and AP mineral components are expected to be highly liberated and reactive in tailings, 3) NP inventories will be determined conservatively using Carb-NP, 4) an Carb-NP/AP value nearer to 1 was appropriate based on kinetic test results, and 5) the reviewed literature data indicate that historical tailings from other sites with NP/AP inventory ratios of greater than 1 have not generated acid.

- SRK (2013)
 - Evaluated the CMR from humidity cells containing waste rock. The CMR was found to vary with the sulphate production or loading rate and high CMR values from lower oxidation rates were attributed to the preferential dissolution of carbonate minerals being an artefact of the humidity cell test process. The authors concluded that the CMR, and therefore, the site specific NPR for the waste rock was close to 1 and a value of 1.5 was selected to be conservative.
- URS (2009)
 - Evaluated the CMR for waste rock and tailings in humidity cell tests. A site-specific NP/AP value for waste rock of 1.7 was selected from the available data and the CMR for tailings was found to be approximately 1.
- Lorax (2018)
 - Evaluated the CMR for humidity cell tests containing PAG, non-PAG and uncertain waste rock. The results showed that the CMR values were biased high when sulphate loading rates were less than 5 mg/kg/week as a result of the comparatively high calcium and magnesium concentrations that were affected by test-specific dissolution effects and would not be representative of field conditions. Conversely, the CMR values were between 1 and 2 for sulphate loading rates of greater than 10 mg/kg/wk and several humidity cell tests had CMR values close to 1.
- Nicholson et al. (2018)
 - Evaluated the CMR values in porewater extracts from waste rock samples collected from piles at a mine site 25 years after closure. The geometric mean Carb-NPR values from all mine rock was 9.5 and the various pile generally had a similar range of Carb-NPR values. The porewater extraction results showed that the CMR varied with sulphate concentrations, exhibiting values much greater than 2 for samples with sulphate concentrations of less than about 100 mg/L, averaging about 2 in samples with sulphate concentrations between 100 and 1,000 mg/L, and that the CMR was consistently near a value of 1 in samples with sulphate concentrations greater than about 1,500 mg/L and is likely controlled by gypsum precipitation.
- Krungalz (2018)
 - This study determined the solubility of alkaline (Na, K) and alkaline earth (Ca, Mg, Sr, Ba) sulphate solids. The sulphate concentrations associated with equilibrium of potassium, sodium, and magnesium sulphate solids are in the range of 10,000 to 100,000 mg/L, values that are uncommon in neutral drainage from sulphide mine materials and it is therefore concluded that the sulphate concentrations are not controlled by those solids. In contrast, the solubility of the calcium sulphate mineral, gypsum ($\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$), will limit sulphate concentrations to values of about 1500 mg/L in neutral pH water. Because calcium is similarly available from other sources such as calcium carbonate dissolution during acid neutralization, the formation of gypsum can limit calcium concentrations to about 500 to 600 mg/L.

The literature review indicates that, there are two key conditions that must be met for the kinetic test to provide useful information to allow for the observed CMR to be applied and interpreted to represent the actual NP/AP inventory ratio for non-PAG classification:

1. Sulphate concentrations should be greater than approximately 20 mg/L to limit interferences due to low rates of oxidation or high rates of water addition where the rate of ongoing sulphide oxidation does not control the rate of NP dissolution.
2. Sulphate and calcium concentrations should be below levels influenced by gypsum solubility to avoid potential interference in estimating the sulphide oxidation-influenced CMR.

METHODS

An extensive review of results from various lab-scale and field scale static and kinetic tests on a range of site tailings performed over many years was completed to evaluate the site-specific NP/AP ratio. The CMR represented by the $(Ca+Mg)/SO_4$ molar ratios in the kinetic test leachate samples represent the potential NP/AP ratios for appropriate conditions.

It is known that the CMR values are not representative of the neutralization of acid by sulphide oxidation in tests with low sulphate concentrations, resulting either from very low oxidation rates or high ratios of water to solids (W:S) in the tests.

Under conditions of low sulphate concentrations, the CMR and therefore the NP/AP ratios typically exceed a value of 2 because the high water flushing rates and solubility controlled dissolution and not the acid production, control the dissolution rates of the carbonate minerals. It is also known that when sulphate concentrations are high enough so that gypsum ($CaSO_4 \cdot 2H_2O$) precipitation can control the concentrations of calcium and sulphate, the CMR, and therefore the NP/AP ratios, will be biased toward a value of 1. All kinetic test data were screened for the appropriate intermediate ranges of sulphate concentrations, generally ranging between 20 and 1000 mg/L were used, to ensure that the CMR or actual NP/AP consumption ratio of a sulphide mine material was evaluated.

A brief summary of the kinetic tests discussed herein is given in Table 1 and it is noted that all kinetic tests evaluated maintained neutral pH conditions over the duration of the testing programs and both the humidity cell and column tests were suitable to assess sulphide oxidation and carbonate dissolution.

Static testing of contact waters in the exposed surficial tailings was also completed but the results suggested that gypsum was controlling the sulphate concentrations, as a result of the low W:S ratio in surficial tailings, where nearly all samples had sulphate porewater concentrations greater than 1000 mg/L and as such, the results were not usable for this analysis.

Table 1. Summary of the kinetic tests conducted on tailings and key test information assessed in this study

Test Year	Test Type	Test duration (weeks)	Sample mass (kg)	Water rinse rate (L/kg/week)	pH at end of testing period	Average sulphate (mg/L)	Average $(Ca+Mg)/SO_4$ molar ratio
2015	Humidity Cell Test	50	1	1	7	48.7	1.02
2015	Humidity Cell Test	50	1	1	7.2	26.4	1.21
2009-2010	Column Test	83.6	17.2	0.027	7.9	729.6	1.00
2009-2010	Column Test	83.6	17.2	0.027	7.8	632.8	0.97
2009-2010	Column Test	83.6	17.2	0.027	7.8	573.3	1.13
2016-2022	Field Test Cell	350	18216	0.0125	8.0	1944	0.84

Note: Average sulphate and carbonate molar ratio values do not include values from the early initial flush stages of each respective kinetic test.

The sulphate concentrations in the water from the tailings kinetic tests were all greater than 20 mg/L as shown in Table 1 and therefore the CMR will be related to the sulphide oxidation and acid generation. All tests, except for the Field Test cell, had sulphate concentrations less than 1000 mg/L and therefore, the CMR will not be influenced by the formation or dissolution of gypsum. The kinetic tests, with the exclusion of the Field Test cell were therefore considered to be appropriate for the interpretation of the CMR as a measure of the NP/AP consumption ratio.

The W:S ratio and the corresponding CMR value for all tailings tests, including the illustrative Field Test data, are summarized in Figure 1. The CMR values ranged between 0.84 and 1.2 and varied only slightly with different W:S ratios, suggesting that the water addition rates were not high enough to affect the CMR

values. A horizontal line is shown at a CMR of 1.2, corresponding the highest CMR value quantified during these tests.

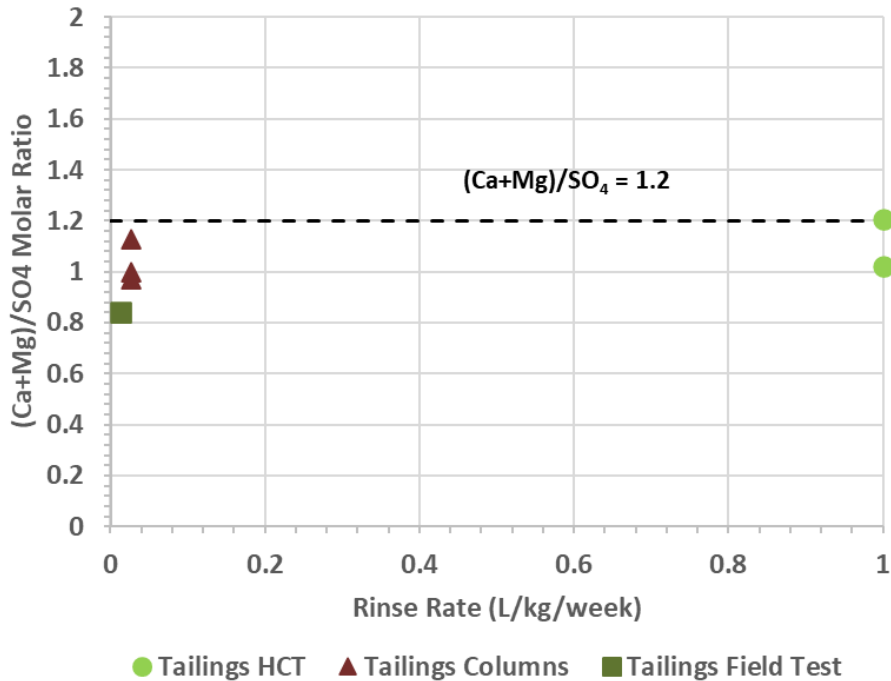


Figure 1. The $(Ca+Mg)/SO_4$ molar ratios plotted against the average water rinse rates for the different scales of tailings tests. Test details are summarized in Table 1.

RESULTS

In consideration of the observed sulphate concentrations the results of the humidity cell and column tests are the most appropriate to interpret the CMR. As shown in Figure 2, the sulphate and calcium concentrations in the humidity cell tests (HCTs) were well below 1,000 and 500 mg/L, respectively, and therefore there is a low probability of gypsum solubility influences on the measured ratios. The pH values remained neutral over the duration of both kinetic tests ranging between 6.6 and 9.0 through the tests and concluding the at values of 7.0 and 7.2.

The column test results shown in Figure 3 exhibit some sulphate and calcium concentrations near 1,000 and 500 mg/L, respectively. The average ratios from those tests were therefore calculated for the period after the sulphate concentrations decreased below 1,000 mg/L to ensure that gypsum solubility did not influence the ratios. The pH values remained neutral over the duration of all three column tests and ranged between 7.2 and 8.4 through the tests and concluding at values of 7.8, 7.8, and 7.9.

The average CMR ratios for the HCTs and column tests ranged from 0.97 to 1.2. These values are relatively close to 1.0 and are consistent with the concept that tailings have more restricted CO₂ gas exchange with the contact water in the pore spaces than can occur in well drained waste rock and therefore have less potential for degassing of carbon dioxide and tend to have higher proportions of carbonic acid in solution and lower CMR values. It is also evident that the CMR ratios from the column tests of 0.97, 1.0, and 1.1, were, on average, closer to 1 than those from the HCTs with values of 1.0 and 1.2. This may reflect even less potential for gas exchange for tailings porewater in the columns than in the smaller HCTs where some gas exchange can occur from the open surface of the tailings in the cell. The tailings columns will be similar to tailings in the tailings storage facility, with more tailings present below the surface of the material where gas exchange can readily occur.

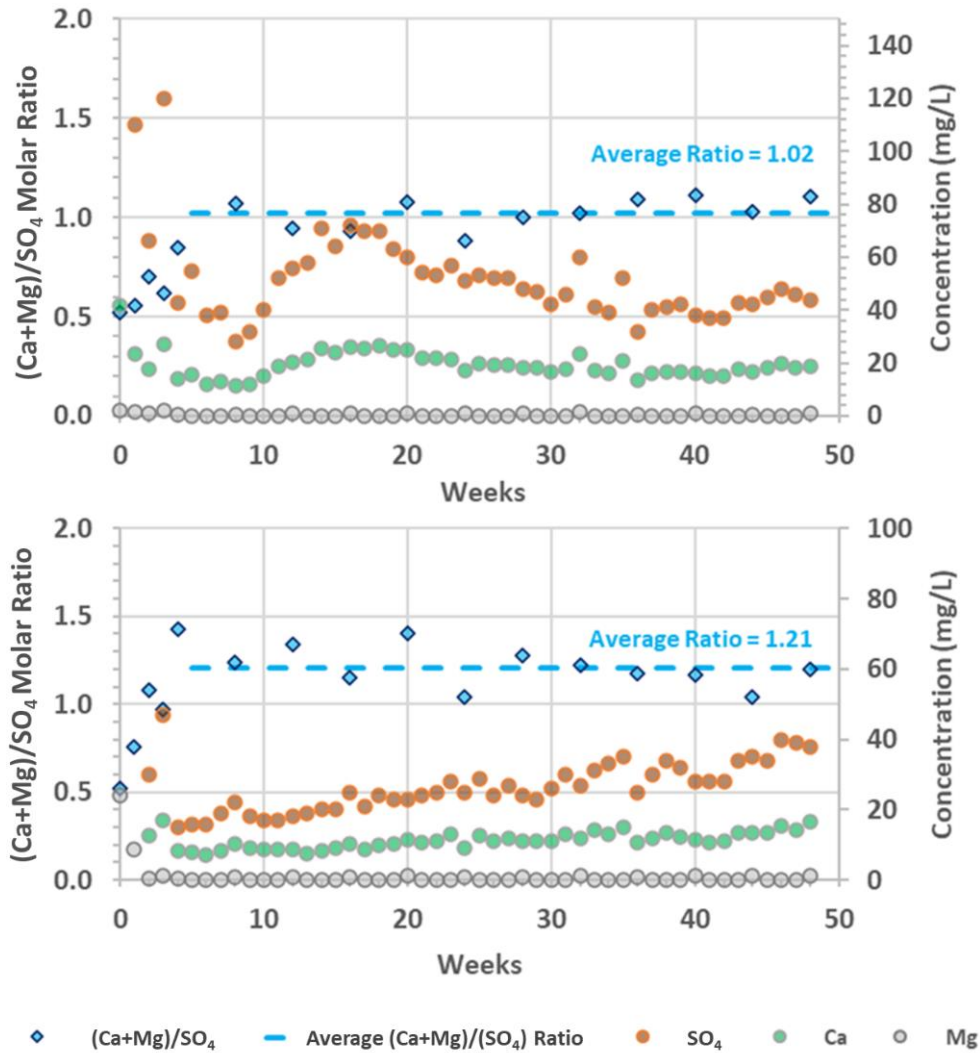


Figure 2. (Ca+Mg)/SO₄ molar ratios, sulphate, calcium, and magnesium concentrations for two tailings humidity cells. Test details are summarized in Table 1.

Overall, the results indicate that the CMR for the tailings in the site tailings storage facility would be expected to be close to 1 and under the conditions observed Reaction 4 and Reaction 3 are dominant in this tailings system at neutral pH values. The upper value of 1.2, as quantified from tailings humidity cell, would be considered conservative for the purposes of classifying non-PAG tailings in the tailings storage facility.

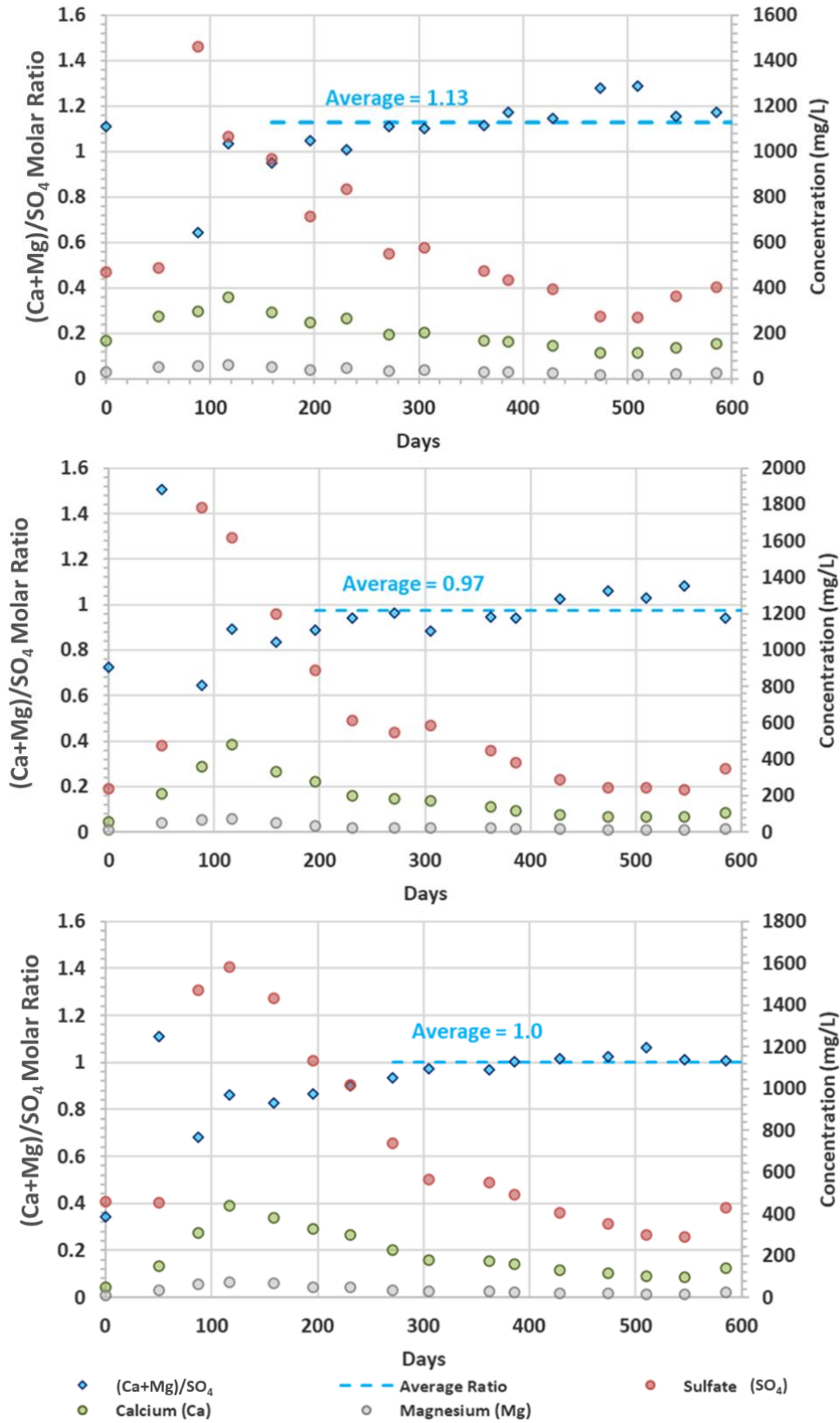


Figure 3. The (Ca+Mg)/SO₄ molar ratios, sulphate, and calcium concentrations for three tailings columns. Average ratios calculated after sulphate concentrations decreased below 1000 mg/L. Test details summarized in Table 1.

CONCLUSIONS

The neutralization potential ratio (NPR=NP/AP) criteria for classifying mine materials as non potentially acid generating (non-PAG) has historically been set at values of 2 or 3 to account for some uncertainty in the neutralizing reactions (Price 2009), a value of 2 is often applied with the understanding that that this is a conservative upper bound limit when NP sources are known to be “effective” (e.g., calcite) (MELCC 2020; Price, 2009). The actual NPR of mine materials using effective inventories is expected to be between 1 and 2 and a site-specific value can be determined and justified with appropriate evidence.

Kinetic testing results can be used to assess the actual NPR representing the NP consumed to AP consumed. The NP is represented by the dissolved calcium plus magnesium released from carbonate minerals and the AP consumed is represented by the sulphate released from sulphide oxidation to solution. The NPR is then represented by the carbonate molar ratio (CMR) expressed as the ratio of (Ca+Mg)/SO₄ concentrations in the water from kinetic tests.

The HCT and column tests on tailings showed that the CMR was consistently close to 1, with an average CMR from the five applicable tests of 1.07. This average 1.07 would be appropriate to use for the calculation of carbonate mineral depletion times and for modelling of alkalinity or acidity production in the tailings storage facility assuming that tailings representative of the ultimate surface layer have similar compositions as analyzed in this test program. This assumption is considered likely for the site storage facility presented herein in consideration of the historical consistency of the site ore-body characterization and operational plans. Further, during ongoing operations the site tailings quality will continue to be periodically monitored during deposition to maintain a thorough understanding of the geochemical properties and characteristics of the ultimate surface tailings. However, for additional operational certainty, the highest single-test average CMR value of 1.2 is considered to represent a conservative NPR value to define non-PAG tailings for management purposes. This representative value was proposed and accepted for permitting of non-PAG tailings determination and the value was then applied in long-term oxidation modelling to forecast tailings oxidation risks through operations and closure.

The quantification of a tailings-specific NP/AP consumption ratio represents a critical operational decision point for site managers to appropriately contextualize the risks to tailings storage as this ratio can affect both operational and closure implications to manage and mitigate risks to water quality, net-acidity onset delays, the potential magnitudes of net-acid production, and the duration of net-acid production periods. For example, the identification of a lower NP/AP consumption ratio for a mine site can affect decision-making as a shift from a generic conservative NPR target to a site-specific value can have important cost implications by limiting the need for tailings desulphurization at facilities with borderline NPR values and can provide more cost-certainty for short- and long-term forecasts. However, validation of the appropriate site-specific values requires a great deal of characterization to ensure appropriate interpretation of representative materials.

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