

Numerical modelling of contaminated neutral drainage from a waste rock field test plot



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ABSTRACT

Seven field test plots were installed at a mine site where Ni-containing contaminated neutral drainage (CND) is leaching from waste rock piles. The effluent quality has been monitored continuously since installation in 2006. The numerical code MIN3P for multicomponent reactive transport was applied to simulate the behaviour of the waste rock test plots and to predict long-term hydrogeochemical evolution. The main objective was to simulate CND production and Ni sorption. The 1-D conceptual model includes sulphide oxidation, acid neutralisation by silicates, and adsorption of Ni onto iron oxides as a complexation mechanism.

RÉSUMÉ

Sept cellules de terrain ont été installées sur un site minier où du drainage neutre contaminé (DNC) au nickel provenant des haldes à stériles est parfois observé. La qualité de leur lixiviat fait l'objet d'un suivi depuis leur installation en 2006. Le code numérique MIN3P, simulant le transport réactif à plusieurs composantes, a été utilisé pour simuler le comportement des cellules ainsi que pour prédire l'évolution hydrogéochimique à long terme. L'objectif principal était de simuler la production de DNC et la sorption du Ni. Le modèle conceptuel en 1D comprend l'oxydation des sulfures, la neutralisation de l'acidité par les silicates et l'adsorption du Ni sur les oxydes de fer par un mécanisme de complexation.

1 INTRODUCTION

Surface storage of waste rock is an important environmental concern in the mining industry. Waste rock is produced when barren rock is extracted to expose the ore, and is much more abundant in the case of open-pit mines. Waste rock is generally placed in large piles that can cover from a few to several tens of hectares. Since the piles are exposed to atmospheric conditions, water and air flow through the waste rock may generate contamination, depending on the rock's natural composition and on geochemical processes such as oxidation, dissolution, precipitation, and sorption. When sulphide minerals are present in the waste rock, their oxidation may lead to acid mine drainage (AMD), which involves high sulfate, low pH, and high metal content in the percolating water.

Another type of contaminated drainage related to the leaching of soluble metals in near-neutral conditions is called contaminated neutral drainage (CND). Indeed, CND may arise when sulphide oxidation-generated acidity is sufficiently buffered by neutralization reactions, but where some metal ions remain soluble (e.g. Pettit et al. 1999; Nicholson 2003). The most common metal ions involved with CND are nickel (Ni) and arsenic (As)

because of their low solubility at neutral pH conditions (Pettit et al. 1999; Scharer et al. 2000; Nicholson 2003).

The mine site under study has been in production for over 50 years, and has generated waste rock piles containing mostly plagioclase (labradorite), ilmenite, and some pyrite, micas, spinel, and chlorite. For many years, effluent quality was within the Quebec norms (Directive 019). More recently, however, while pH has remained near neutral, nickel concentrations near (or slightly above) the 0.5 mg/L limit have been observed in the waste effluent, particularly from older sections of the waste rock pile. It has been postulated that Ni is retained within the fresh waste rock as adsorbed species, but as the rock ages, either adsorption sites become saturated or they start to release adsorbed Ni. Laboratory and field work was therefore initiated to investigate the CND contamination and possible adsorption phenomena.

Physico-chemical modelling of waste rock piles has been attempted by several authors. However, the focus was usually placed on pyrite oxidation and AMD generation. Lefebvre et al. (2001b; 2001c; 2001a) modelled several AMD-generating waste rock piles using heat, water, and oxygen flow, and using the shrinking core formulation for pyrite oxidation. Molson et al. (2005) added a reactive transport module to a flow model to

include further geochemical processes including mineral precipitation and dissolution, and were able to model a waste rock pile in 2-D.

Some models, not limited to waste rock piles, included more advanced geochemical processes along with fluid flow (e.g. Bain et al. 2001; Nicholson et al. 2003; Linklater et al. 2005). Schwartz and Kgomanyane (2008) showed the implication of adding sorption processes to the simulation of groundwater heavy-metal contamination from a leaking AMD-generating tailings impoundment. In the present study, we will use the multicomponent model MIN3P (Mayer et al. 2002). To the authors' knowledge, physico-chemical modelling of contaminated neutral drainage along with sorption, especially in the case of waste rock, has not yet been reported in the literature.

2 FIELD WORK

This research project was initiated in collaboration with an industrial partner who has observed Ni-contaminated drainage from its waste rock piles. The field project involved the construction of seven test plots to evaluate the geochemical behaviour of several types of waste rock present at the site, each type in its individual test plot. Two types of test plots were built: six test plots contained waste with the surface exposed to natural conditions, and one test plot was built to simulate underwater disposal or submerged conditions (not discussed in this paper). Two main waste characteristics were tested: 1) concentration of ilmenite in the waste rock, and 2) level of oxidation.

The configuration and dimensions of the test plots are shown in Figure 1. Test Plots 1 to 3 were filled with freshly blasted waste rock with ilmenite content varying from low grade (20%, Test Plot 1), medium grade (40%, Test Plot 2), to high grade (60%, Test Plot 3). Test Plots 4 to 6 have the same increment in ilmenite grade, but were made with older waste rock that was deposited in the main piles 25 years ago.

The test plots were monitored for effluent quality and flow rate by monthly sampling campaigns performed by the mine staff. Measurements of pH, oxidation-reduction potential (ORP), electrical conductivity, water temperature, and flow rate were conducted on site, while samples were shipped to the URSTM-UQAT laboratory for ICP-AES analyses of metal content. A meteorological station was also installed to provide site-specific climate data. The precipitation data that was used for the model were collected between May 1 and November 30, 2008.

Test Plot #3, with a high ilmenite concentration and composed of freshly blasted material, was chosen for the initial modelling. Further details on the other test plots are available in Pepin (2009) and Pepin et al. (2008).

The waste rock used in Test Plot #3 is composed mostly of ilmenite (45.7%), hematite (20.3%), plagioclases (labradorite, 16.1%), and pyroxenes (enstatite, 8.5%), as measured from the normalized XRD analysis presented in Table 1. Sulfide minerals (pyrite and chalcopyrite) are present in low amounts.

Table 1: Mineralogical characterization of the waste rock

Mineral	Weight fraction (%)	Density (g cm ⁻³)	Volume fraction (m ³ m ⁻³)
Ilmenite	45.70	4.72	0.2517
Hematite	20.26	5.30	0.0994
Labradorite	16.11	2.69	0.1557
Enstatite	8.54	3.20	0.0694
Orthoclase	4.54	2.56	0.0461
Pyrite	1.60	5.02	0.0083
Chlorite	0.90	3.00	0.0078
Spinel	0.69	3.64	0.0049
Rutile	0.59	4.25	0.0036
Muscovite	0.47	2.82	0.0044
Biotite	0.42	3.09	0.0036
Chalcopyrite	0.19	4.20	0.0012

The particle size distribution was evaluated by sieving. Relevant geotechnical parameters are provided in Table 2. According to the USCS classification system, the waste rock is classified as coarse, and as a well graded gravel with cobbles and boulders.

Table 2: Selected geotechnical parameters for the waste rock from Test Plot #3

Parameter	Value
Particle size distribution	
• D ₁₀	• 0.78 mm
• D ₅₀	• 13.20 mm
• C _c	• 0.31
• C _u	• 52.88
van Genuchten parameters	
• α _v	• 5.97 m ⁻¹
• n _v	• 3.61
• AEV	• 10 cm
• θ _r	• 0.07
Saturated hydraulic conductivity	5 x 10 ⁻⁴ m s ⁻¹
Porosity	0.35

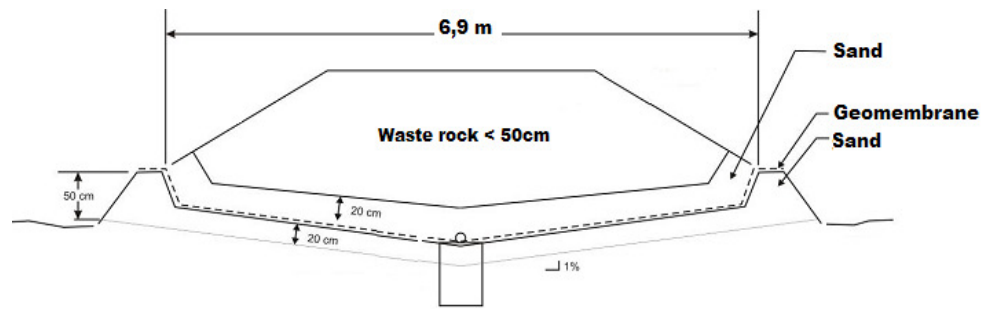


Figure 1: Configuration of the test plots

Water retention curves were evaluated in the laboratory using columns at Ecole Polytechnique and then fitted with the van Genuchten RETC code (Table 2). The saturated hydraulic conductivity was also obtained and predicted using the modified Kovacs model; the value of 5×10^{-4} m/s was selected for the MIN3P model.

3 CONCEPTUAL MODEL

The development of an accurate conceptual model is a critical step in numerical modelling. The model developed for the present study involved two major assumptions that needed to be addressed: the source of nickel, and the sorption mechanism.

3.1 Source of nickel

The waste rock placed in the field test plots was analysed carefully because of their nickel contaminated effluent. However, no major nickel-bearing mineral was identified by XRD (see Table 1). A chemical analysis by ICP-AES of the waste rock revealed that the sample from Test Plot #3 contained 0.043% Ni. Further mineralogical analyses performed by optical and scanning electron microscopy indicated that the nickel-bearing mineral millerite (NiS) was present as inclusions in some pyrite grains, as shown in Figure 2 (Pepin, 2009). The quantity was too low, however, to be detectable by x-ray diffraction.

Millerite was therefore added to the mineralogical composition of the waste rock in the model, at a volumetric fraction calculated from the Ni content obtained from the chemical analysis. A mineral content of 0.001% was estimated, corresponding to a volumetric fraction of 4.78×10^{-6} . To simplify the model, standard kinetically-controlled dissolution behaviour was assumed for millerite using data and parameters obtained from the MIN3P database and from the literature (Amos et al. 2004).

3.2 Sorption mechanism

Laboratory work was performed in parallel with the modelling work in order to investigate the sorption mechanisms that were suspected to interfere with Ni

release into the waste rock effluent. Indeed, in-situ Ni-contaminated effluent occurred mainly in aged waste rock, where it is believed that sorption is reduced. Laboratory work included sequential adsorption tests and analyses in an attempt to identify the adsorption capacity of the waste rock and the species involved. Details on the laboratory work can be found in Plante (2010) and Plante et al. (2008).

Because sorption has an impact on in-situ CND, it was necessary to add this process into the model. MIN3P includes two types of sorption mechanisms: ion-exchange reactions, and surface complexation reactions. The laboratory work showed that surface complexation would be the most likely adsorption mechanism. In the conceptual model, some simplifications were required. It was first assumed that nickel ions are adsorbed onto iron oxides (such as ilmenite). Ilmenite is the highest-fraction component of the waste and has a significant adsorption capacity as evident from the laboratory investigation. Plagioclase minerals are also believed to be involved in adsorption of nickel, however this hypothesis was not included in the model. Analyses by XPS identified nickel hydroxide as a possible sorption complex (Plante et al. 2010), which strengthens the assumption of surface complexation with iron oxides.

Nickel adsorption was modelled similarly to zinc adsorption, since both metals are generally considered to behave similarly. In the model, $\text{FeOH}_{(s)}$ is the surface species that accepts Ni, while FeNi^+ is considered the adsorbed species. Parameters such as surface site mass, area, and density were obtained from both laboratory results and iterative modelling to calibrate the values.

3.3 Geochemical system

The Test Plot #3 was conceptualized as a 1-dimensional column containing 2.4 m of waste rock discretized with 200 control volumes. The geotechnical properties presented in Table 2 were included in the model, along with the mineralogical composition presented in Table 1. The geochemical reactions for each mineral, redox couple, and gas are mainly typical values used by other authors.

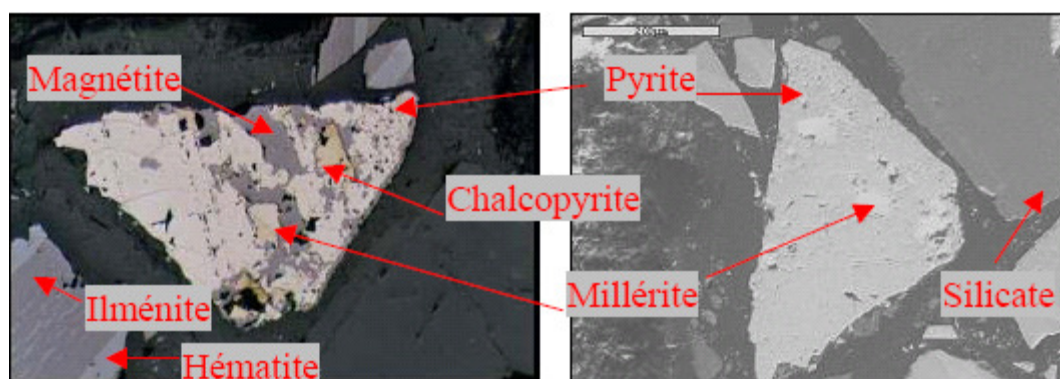


Figure 2: Pyrite grain with inclusions, left: under optical microscopy (polarized, 100x); right: scanning electron microscopy (from Pepin 2009)

3.4 Boundary conditions

The top boundary condition, representing the surface of the waste rock exposed to the atmosphere, is expressed as a transient fluid flux based on seasonal variations. Observed precipitation data were used as recharge water flux, with its chemical composition given in Table 3. The bottom boundary condition represents free outflow of water, thus it was assigned a constant pressure head condition of 0 m. The system was initially saturated with water having the same composition as the recharge water.

Table 3: Recharge water composition applied in the model.

Element	Concentration (mol L ⁻¹)
Ca ²⁺	2.2 x 10 ⁻⁵
K ⁺	2.0 x 10 ⁻⁵
Cl ⁻	1.0 x 10 ⁻⁴
H ₄ SiO ₄	2 x 10 ⁻²⁰
Al ³⁺	1 x 10 ⁻²⁰
Fe ²⁺	1.3 x 10 ⁻¹⁰
Fe ³⁺	2.0 x 10 ⁻⁵
SO ₄ ²⁻	1.0 x 10 ⁻⁵
HS ⁻	2.6 x 10 ⁻¹²
Mg ²⁺	8.23 x 10 ⁻⁷
Na ⁺	2.0 x 10 ⁻⁵
Cu ²⁺	1.0 x 10 ⁻¹⁰
Ni ²⁺	1.0 x 10 ⁻¹⁰
Ti ²⁺	1.0 x 10 ⁻¹⁰

4 NUMERICAL RESULTS

4.1 Model calibration – flow system

Model calibration was first done with respect to the flow system, without including any geochemical reactions. Geotechnical parameters were input into the model along with precipitation data. To calibrate the model, van Genuchten parameters were slightly varied to obtain a better fit between observed flow and predicted flow. Since the waste rock is coarse grained and unsaturated, recharge water travels through the column fairly quickly, reaching the bottom usually within a few hours of each rain event (Pepin 2009). This behaviour was also observed using the model, as shown in Figure 3.

4.2 Model calibration – neutral drainage

Once the flow system was calibrated, the reactive geochemical system was simulated by adjusting the reaction rates and initial mineralogical mass fractions. The first step involved the adjustment of the pH values without including nickel in the model.

At the Tio waste rock site, the conceptual model for contaminated neutral drainage includes simultaneous pyrite oxidation and acid neutralization. Silicates were identified as the primary neutralizing minerals, so the pH was first adjusted by modifying the pH-dependent dissolution rate of anorthite. Anorthite dissolution also regulates the calcium concentrations, so both pH and calcium were calibrated simultaneously. Pyrite oxidation rates were also adjusted until the simulated pH and sulfate concentrations matched those observed typically in the leachate.

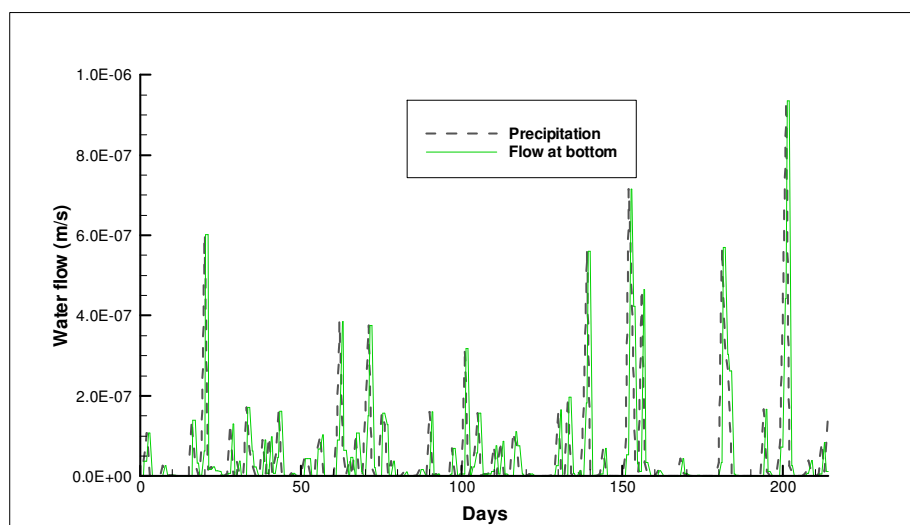


Figure 3: Simulated water flow rate at the bottom of the Test Plot #3 compared to precipitation events

Pyrite oxidation was simulated using the shrinking core model with an equivalent grain radius D_H of 2.35 mm, calculated from the expression (Aubertin et al. 1998):

$$D_H = \left[1 + 1.7 \log \frac{D_{60}}{D_{10}} \right] \times D_{10} \quad [1]$$

With the appropriate reaction rates, pH and sulfate concentrations corresponded satisfactorily with typical measured values for a 13-day simulation. Results are presented in Figure 4. Measured pH values are around 6.2, while the modeled pH values at the outlet were between 5.7 and 6.1. Calcium concentrations measured at 0.002 mol/L were modeled between 0.0015 and 0.003 mol/L, while sulfate was predicted between 0.0025 and 0.004 for a measured value of 0.002.

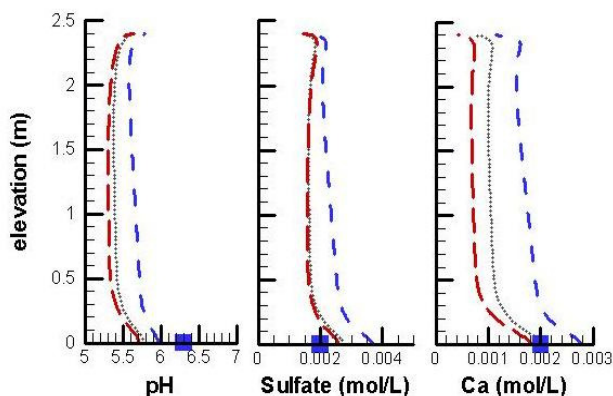


Figure 4: Simulation results for day 1 (dashed line), day 5 (dotted line), and day 10 (long dashed line), compared to typical measured values (blue square).

4.3 Model calibration – nickel adsorption

After calibration of the model for sulfate, calcium, and pH, the primary Ni-bearing sulphide mineral millerite was added to the reactive system. Since millerite was present at such low volume fractions (0.001%), its dissolution should not significantly affect the oxidation – neutralization mechanism established in the previous calibration step.

Initially, millerite was added without any sorption mechanism; however in this scenario, the simulated Ni concentrations in the leachate were significantly higher than the in-situ measurements. It appeared that sorption was indeed necessary to reduce the Ni content to measured values. Adjustment of the sorption parameters was done iteratively. Figure 5 shows that nickel is released within the waste rock but its concentration is

reduced to low values at the outlet, indicating that Ni is retained in the waste rock. The low values predicted at the leachate outlet are consistent with measured values for “fresh” waste rock.

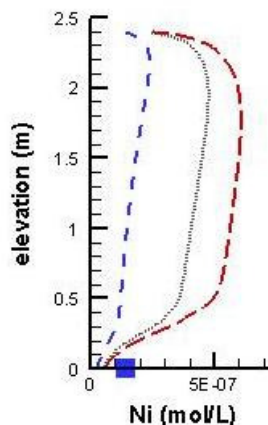


Figure 5: Simulation results for day 1 (dashed line), day 5 (dotted line), and day 10 (long dashed line), compared to typical measured values (blue square).

4.4 Modelling for an entire season (May – November)

The model was then executed for the entire 214 days for which precipitation data were available in 2008. The simulated concentrations vs. time at the base of the column are presented in Figure 6. Measured effluent quality is also presented in Figure 6 for comparison. The simulated pH remained stable through the year, while the measured values oscillated between 5.5 and 7. Simulated and measured sulfate concentrations were in the same range, except for two higher measured values.

Calcium started high, but decreased to very low values after 150 days. This behaviour was unexpected because calcium dissolution should closely follow sulfate release since SO_4 and Ca are reflective of, respectively, oxidation and neutralization of acidity. The measured values, while in the same range of values, do not show this decreasing trend in the 100 days of monitoring for the season. It is possible that some assumptions in the mineralogical content of the waste input in the model restrict the amount of calcium available for neutralization, this aspect will be investigated further.

Dissolved nickel increased steadily from the beginning of the simulation time and seemed to level off near $1.2 \times 10^{-7} \text{ mol L}^{-1}$. The measured values were somewhat higher than those predicted, but still within an order of magnitude. When these concentrations are converted into mg/L, Ni values vary between <0.004 mg/L (detection limit) and 0.046 mg/L. Therefore, during the monitoring season shown, Test Plot #3 did not exceed the provincial limit of 0.05 mg/L Ni.

5 DISCUSSION: NEXT STEPS

The model being partly calibrated, further long-term predictions can be performed. Indeed, the short-term, i.e. one season, prediction capability is deemed appropriate. However, for longer-term modelling, some issues remain to be resolved. These issues will be described briefly in this section.

5.1 Long-term prediction

The MIN3P model does not account for freezing or snow-cover conditions, so for long-term predictions a time gap must be added representing frozen (no-flow) conditions and during which all reaction rates are assumed negligible.

Furthermore, some mineralogical issues may arise when modelling over a longer period of time, as already observed with calcium during the 214-day simulation. When the field test plot will be dismantled, chemical and mineralogical analyses will provide some insight on the actual reactions and possible mineral precipitation that occurred during the test plots' lifespan (approx. 5 years). This information will be helpful in refining the model.

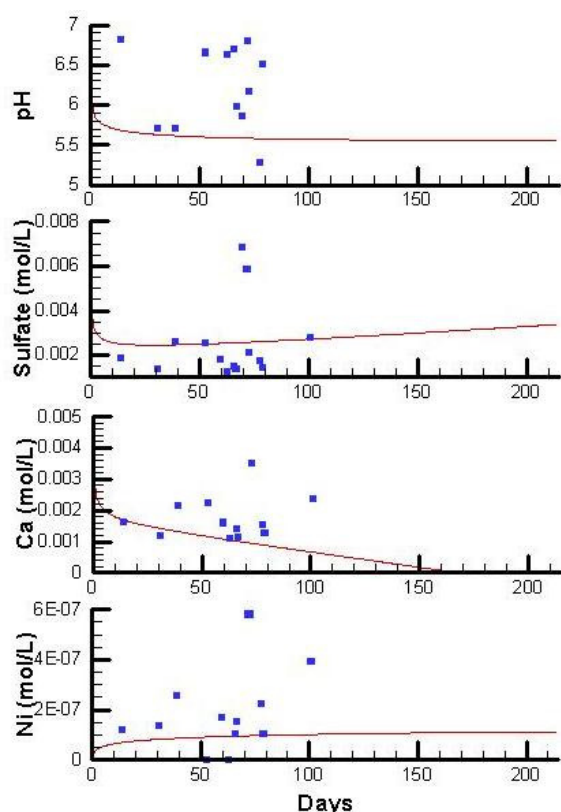


Figure 6: Simulation results for 214 days (line) at the bottom of the model and measured values (squares) during one season at the outlet of Test Plot #3.

5.2 Sorption conceptual model

The field data, from the test plots as well as from historical data of the waste rock piles, indicate that

adsorption varies with time. The field test plots showed that fresh waste rock has more adsorption capacity than aged waste rock, as was seen from the Ni concentration in the leachate. This trend is assumed to reflect the gradual saturation of sorption sites over time. Ideally, the model would adjust the sorption parameters with time to reflect the loss of adsorption with time, however, MIN3P cannot simulate variable sorption kinetics. Once the work with Test Plot #3 is completed, a similar procedure will be used to model Test Plot #6, which has similar waste rock but aged for 25 years. Field results showed that Test Plot #6 releases significant nickel, indicating different adsorption parameters. With the two sets of adsorption parameters, it was proposed that a "step function kinetic rate" could be developed and used to make very long-term predictions of the leachate nickel content, with several "steps" of decreasing adsorption capacity until all sorption sites had been filled.

5.3 From test plot to waste rock pile

Prediction of water flow and effluent quality is not trivial in the case of waste rock piles. A waste rock pile is heterogeneous with a highly variable particle size within the pile, with particle diameters ranging from less than one mm to almost a meter (Fala et al. 2005; Martin et al. 2005). Particle segregation has also occurred in some areas, resulting from the method of waste deposition during pile construction. Water saturation is also spatially and temporally variable, with complex preferential pathways for flow. Thus, the model developed in this work is not suitable for modelling an entire waste rock pile. For such a large mass of material, a one-dimensional model may not be appropriate since flow may not be assumed to be entirely vertical. MIN3P has the capacity to simulate two-dimensional flow, so the model could be extended but the assumed homogeneity of the waste would not be valid. The conceptual model would have to be divided into several sections to reflect the variation in material properties, and it would greatly increase the complexity of the flow and reactive transport calculations.

6 CONCLUSIONS

A 1D numerical model was developed and successfully applied to represent a field test plot at a mine site characterized by contaminated neutral drainage. The model considered sulphide oxidation, acid neutralization, and sorption of nickel. Choices were made about the source of nickel ions, namely the mineral millerite, and the mechanism of nickel adsorption, being surface complexation. Calibration of the model was performed using field data that enabled assigning reaction rates to represent the geochemical system. A full season was modeled which showed increasing nickel concentration at the outlet up to a plateau-like concentration of 1.2×10^{-7} mol/L (0.006 mg/L). Further work is required to adjust the model for better long-term predictions, particularly to

account for seasonal freezing and varying adsorption capacity over time.

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