ABSTRACT: Subsurface contamination of metals and radionuclides are common problems in abandoned mine sites. Metals and radionuclides are likely to occur in the mobile phase or may bind to soil particles and geologic framework. The transport process for reactive contaminants also include chemical reactions in groundwater aquifers, which are complex physical and biogeochemical processes. Accurate tools to reliably predict the movement and changes in concentration of metals and their daughter products (radionuclides) formations in the subsurface environment is important for decision makers to analyze impact, and to evaluate the effectiveness of remediation strategies. An advanced numerical model can provide such useful direction of predictions. A 3-D contaminant transport model for fluid flow, hydrogeologic transport, and biogeochemical kinetic and equilibrium reactions in saturated and unsaturated media was calibrated, validated and implemented to model the hydrogeochemical processes that occur in the subsurface at such a complex contaminated site, consisting of waste rock dumps and flooded mine pits of this abandoned uranium mine site. The developed flow and transport simulation model for no longer in use mine site in Northern Territory of Australia is discussed.

Keywords: Reactive Transport, Flow and Transport Modelling, Groundwater Contamination, Mine site

1. INTRODUCTION

Groundwater plays an important role in human history and groundwater contamination has attracted intensive investigations in the past 30 years. Contaminants in the aqueous environment undergo changes in concentration resulting from physical, chemical, biological processes, and a capability to understand and model these processes is at the core of assessing the efficacy of environmental remediation strategies. The modeling and prediction of the complex multiple species reactive transport process in mine sites is particularly challenging. The development of an accurate model capable of predicting the transport scenario is very challenging, but a necessity for designing future remediation strategies. Therefore, a robust numerical multiple species reactive transport simulation model HYDROGEOCHEM 5.0 [1] has been calibrated and validated for a study area comprising of an underlying aquifer in a no longer operational mine site in the Northern Territory of Australia. Acid rock drainage (ARD) and heavy metal mobilization at the former mine site have led to significant environmental impacts on local groundwater and in the East Branch of the Finniss River [2].

Acid and metalliferous drainage (AMD) also referred to as acid mine drainage or acid rock drainage occurs when sulphidic minerals, such as arsenopyrite, chalcopyrite and pyrite, are exposed to oxygen and water. Although AMD occurs naturally, the majority of AMD is anthropogenic and originates from reactive sulfide minerals stored in waste rock dumps (WRDs), mine pits, and tailings. Notwithstanding the fact that extensive rehabilitation studies at the mine site exist, a well-defined reactive transport model, with a comprehensive suite of reaction types and reaction rates incorporated into a model aimed at the remediation and rehabilitation of the site, has not been implemented yet. These reaction types include but not limited to aqueous complexation, adsorption-desorption, ion exchange, oxidation-reduction, precipitation-dissolution, acid-base reactions, and microbial mediated reactions.

The main sources of pollutants to the environment are the open pit, the waste rock damps and the tailings dam. The waste rock damps and tailing dams most often consist of several minerals and geological matrix, which are exposed to weathering conditions. Pyrite oxidation is the principle reaction in the leaching of metal and radionuclides into the environment. Accurate prediction of the release rate of metal and radionuclides from these sources and their transport into the subsurface environment is a critical factor to the assessment of environmental impacts and to the development of effective remediation strategies. To produce a realistic
representation of the system under study with the existing real complex problems, sophisticated models are required. Reactive transport codes are powerful tools in the evaluation of coupled hydrological chemical processes and in the prediction of the long-term performance of remediation strategies.

A presentation of the application of a reactive-transport model to simulate the transport of reactive multi-species contaminants in heterogeneous, anisotropic, saturated-unsaturated waste rocks damp and mine open pits of the first uranium mine site in Australia is illustrated in this paper. The computer program used to describe the complex hydrogeochemical processes that control the generation of acid drainage and the distribution of solutes between groundwater and waste rock is the hydrogeochemical transport model, HYDROGEOCHEM 5.0. This numerical code has been earlier used for modeling a mine site in Queensland Australia [3]. The computer program is a 3-D numerical model of fluid flow, thermal, hydrologic transport, and biogeochemical kinetic and equilibrium reactions in saturated and unsaturated media.

The purpose of this paper is to focus on the solution results of reactive transport geochemical modeling to predict the important complex processes that control the movement and distribution of contaminants in groundwater. It is also likely that this calibrated and validated transport model will be utilized for a contamination source characterization study for this contaminated aquifer site. Information gained from these investigations should also provide insight into processes that occur at other sites similarly contaminated.

2. NUMERICAL SIMULATION

2.1 Flow Model

The general equations for flow through saturated-unsaturated media are obtained based on following fluid continuity, solid continuity, fluid movement (Darcy’s law), stabilization of media, and water compressibility [1]:

\[ \frac{\rho}{\rho_o} \frac{\partial h}{\partial t} = - \nabla \cdot \left[ K \left( \nabla h + \frac{\rho}{\rho_o} \nabla z \right) + \rho' q \right] \]

(1)

Where F = generalized storage coefficient (1/L) defined as:

\[ F = \alpha_s \frac{\theta}{n_v} + \beta' \theta + n_v \frac{dS}{dh} \]

(2)

\[ K = \frac{\rho s}{\mu} K_s, \quad K_r = \frac{\rho d \rho_o}{\mu \rho_o} K_r, \quad K_w = \frac{(\rho/\rho_o)}{(\mu/\mu_o)} K_w \]

(3)

and \( V = \) Darcy’s velocity (L/T), described as:

\[ V = -K \left( \frac{\rho_o}{\rho} \nabla h + \nabla z \right) \]

(4)

Where: \( \theta \): effective moisture content (L³/L³); \( h \): pressure head (L); \( z \): time (T); \( q \): source or sink of fluid [(L³/L³)/T]; \( \rho_o \): fluid density without biochemical concentration (M/L³); \( \rho^* \): fluid density with dissolved biochemical concentration (M/L³); \( \rho^* \): fluid density of either injection (= \( \rho^* \)) or withdraw (= \( \rho \)) (M/L³); \( \mu_o \): fluid dynamic viscosity at zero biogeochemical concentration [(M/L)/T]; \( \mu \): the fluid dynamic viscosity with dissolved biogeochemical concentrations [(M/L)/T]; \( \beta \): modified compressibility of the liquid (1/L); \( \alpha' \): modified compressibility of the soil matrix (1/L); ne: effective porosity (L³/L³); \( \beta \): modified compressibility of the liquid (1/L); ne: effective porosity (L³/L³); S: degree of effective saturation of water; \( G \): is the gravity (L/T²); \( K \): permeability tensor (L²); \( K_s \): saturated permeability tensor (L²); Kso: referenced saturated hydraulic conductivity tensor (L/T); \( k_r \): relative permeability or relative hydraulic conductivity (dimensionless).

The finite element method was used to solve Eq. (1), (2), (3) and (4), and the constitutive relationships among the pressure head, degree of saturation, and hydraulic conductivity tensor, together with the appropriate initial conditions and boundary conditions. The temporal-spatial distributions of the hydrological variables, including pressure head, total head, effective moisture content, and Darcy’s velocity are simulated.

2.2 Reactive Transport Model

Governing equations for the reactive transport of the reactive biogeochemical system is discussed below. The equations for transport were derived based on the continuity of mass and Fick’s flux laws. The main transport and fate processes are advection, dispersion/diffusion and biogeochemical reactions (including radioactive decay). The general transport equation governing the temporal-spatial distribution of any biogeochemical species in a reactive system is described below [1]:

\[ \frac{\partial C_i}{\partial t} + \nabla \cdot (V C_i) + \sum_{i \neq M} r_i = L(C_i) + \theta_r + M_i \]

(5)

Where L is the transport operator denoting

\[ L(C_i) = -\nabla \cdot (D \nabla C_i) \]

(6)

where \( C_i \) is the concentration of the i-th species in units of chemical mass per water volume [M/L³]; \( r_i \) is the production rate of the i-th species because of biogeochemical reactions in chemical mass per water volume per unit time [M/L³/T].
\{M\} = \{1,2,...,M\} in which \(M\) is the number of biogeochemical species; \(D\) is the dispersion coefficient tensor \([L^2/T]\); and \(M_i\) is the source/sink of the \(i\)-th species in chemical mass per unit volume of media \([M/L^3/T]\).

3. STUDY AREA DESCRIPTION

The former mine site is located approximately 105km by road south of Darwin in the headwaters of the East Branch of the Finniss River, near Batchelor in the Northern Territory, Australia. This former mine was one of Australia’s first major uranium mines and produced approximately 3,500 tonnes of uranium and 20,000 tonnes of copper concentrate between 1954 and 1971 [4]. Uranium was mined from White’s and Dyson’s open pits from 1954 to 1971, while copper was extracted from the Intermediate pit in 1964. The site underwent rehabilitation from 1983 to 1986 at a total cost of $18.6 million. Several rehabilitation works have taken place since 1986 and rehabilitation is still on going. [4].

3.1 Geological and Hydrogeological Characterization

The mine areas consist of east branch of the Finniss River about 8.5 km upstream of its confluence with the West Branch of the Finniss River. Surface water enters the mine site from the east via the upper East Branch of the Finniss River and from the southeast via Fitch Creek. River flows vary in response to intra annual variability in rainfall and changes over the course of a year [5]. The mine area mineral field contains polymetallic ore deposits, such as the Ranger and Woodcutters ore deposits. The mine site includes the Giant’s reef fault. The main lithological units are the Rum Jungle Complex and meta-sedimentary and subordinate meta-volcanic rocks of the Mount Partridge Group. The Rum Jungle Complex consists mainly of granites and the Mount Partridge Group consists of sedimentary units: Geolsec formation, the Whites formation, the Coomalie Dolostone, and the Crater formation [6].

3.2 Conceptualization and Model Discretization

A numerical groundwater flow model is constructed to simulate variations in the groundwater flow system at the mine site from December 2010 to November 2014. This numerical flow model is a mathematical representation of a conceptual model that enables a quantitative representation of real field features. The numerical representation is based on the following assumptions: The aquifer system at the former mine site can be subdivided into hydrostratigraphic units that represent either waste rock damps or the naturally occurring bedrock aquifer, each hydrostratigraphic unit is represented as a single model layer with representative hydraulic properties and recharge is estimated as a proportion of incident rainfall, mine waste is represented by a single model layer of variable thickness of the waste rock damps and Dyson’s (backfilled) open Pit, whereas the geological aquifer units are represented by model layers with constant thicknesses across the model domain, water movement in the hydrostratigraphic units follows Darcy’s law, the flooded open pits is represented by specified head boundary condition that is equivalent to observed water levels in the pits during the simulation period.

3.2.1 Numerical method

The finite element method is used for temporal and spatial discretization of the governing partial differential equations in the flow module. The Galerkin finite element method was used for spatial discretization of the modified Richards equation that governs the distribution of pressure fields. For this model, the critical assumption of this approach is that every element within a particular section of a layer is assigned a set of hydraulic properties to account for heterogeneity. The layers are discretized horizontally into triangular wedge elements but each layer of a finite element mesh is one element thick. The flow of the groundwater is simulated for the observation period 2010 and 2014. Two case scenarios of both steady state and transient flow is simulated for the flow.

3.2.2 Spatial discretization

The numerical model domain was spatially
discretized into a 3-dimensional mesh with a triangular wedge mesh. In planar view, each element is a triangular wedge, whereas the thickness of the elements depends on the number of layers used to vertically discretize the model domain. The thickness of the elements varies depending on lithology. The model is made up of 6 layers and covers a maximum elevation of about 110 m. Surface topography from a recent terrain elevation data was used to define the top of layer 1, including the WRDs and Dyson (backfilled) pit. Figure 2 shows a plan view of finite element model domain. The thickness of the elements depends on the number of layers used to vertically discretize the model domain. The thickness of the elements varies depending on lithology. The model is made up of 6 layers and covers a maximum elevation of about 110 m. Surface topography from a recent terrain elevation data was used to define the top of layer 1, including the WRDs and Dyson (backfilled) pit.

3.2.3 Boundary conditions

Specified heads boundary conditions are assigned to elements from layers 1, 2, 3, and 4 that intersects the perimeters of the flooded pits which includes main, intermediate and brown oxide pits. The elements represent the bedrock aquifer that is in contact with standing water within the pits and was assigned a head that is equal to the measured water level in the pits. Flows within the flooded open pits themselves are not simulated by the model so elements within the head boundary are set to be inactive. Pit water levels and groundwater levels at monitoring locations close are used to represent the open pit as a head boundary. The brown oxide open pit is not completely flooded so heads were only specified for elements in layers 3 and 4. Time varied constant head nodes are set that simulate water level changes in the main, intermediate and brown’s pits and the river. The aquifer parameters imputed to the model area is shown in Table 1

To model and simulate the reactive transport processes, equations of either the conventional finite element methods or the hybrid Lagrangian–Eulerian finite element methods were used for spatial discretization. The chemical equilibrium equations were solved by the Newton–Raphson method or Picard method. This study considers the possible hydrogeochemical transport of 6 components, including $\text{OH}^-$, $\text{Cu}^{2+}$, $\text{Fe}^{2+}$, $\text{Fe}^{3+}$,$\text{Mn}^{2+}$, $\text{UO}_2^{2+}$ and 20 species are considered in this study. These selected sets of components are based on the fact that the primary mining commodities were uranium and copper. Table 2 shows some chemical reactions incorporated in the model.

### Table 1 Aquifer hydrogeologic properties

| Aquifer parameter | Value |
|-------------------|-------|
| Number of nodes   | 6587  |
| Number of elements| 10704 |
| Vertical hydraulic conductivity, $K_v$ | $\text{m/d}$ |
| Layer 1           | 0.01728 - 5.0112 |
| Layer 2           | 0.00864 - 0.7776 |
| Layer 3           | 0.00864 - 0.8467 |
| Layer 4           | 0.00864 - 0.8467 |
| Layer 5           | 0.00864 - 0.0259 |
| Layer 6           | 0.001728 - 0.0648 |
| Vertical hydraulic conductivity, $K_s$ | $\text{m/d}$ |
| Layer 1           | 0.00864 - 0.2419 |
| Layer 2           | 0.00432 - 2.4192 |
| Layer 3           | 0.00864 - 0.8640 |
| Layer 4           | 0.00864 - 1.2960 |
| Layer 5           | 0.00864 - 0.2074 |
| Layer 6           | 0.00864 - 0.0648 |
| Effective porosity, $\theta$ | 0.28 |
| Longitudinal dispersivity, $a_L$ | 10 m/d |
| Transverse dispersivity, $a_T$ | 0.1 m/d |
| Vertical dispersivity ($a_V$) | 0.01 |
| Average rainfall  | 2372 mm/year |

### Table 2 Chemical reactions involved in transport

| Chemical reactions | Constant rate (log K) |
|--------------------|-----------------------|
| $\text{H}_2\text{O}^{(aq)} \rightarrow \text{H}^+ + \text{OH}^-$ | -13.99 |
| $\text{H}^+ + \text{SO}_4^{2-} \rightarrow \text{HSO}_4^{-}$ | 1.99 |
| $\text{Cu}^{2+} + \text{H}_2\text{O} \rightarrow \text{Cu(OH)}^+ + \text{H}^+$ | -9.19 |
| $\text{Cu}^{2+} + \text{SO}_4^{2-} \rightarrow \text{CuSO}_4$ | 2.36 |
| $\text{Cu}^{2+} + 2\text{H}_2\text{O} \rightarrow \text{Cu(OH)}_2 + 2\text{H}^+$ | -16.19 |
| $\text{Cu}^{2+} + 3\text{H}_2\text{O} \rightarrow \text{Cu(OH)}_3 + 3\text{H}^+$ | -26.9 |
| $\text{Fe}^{2+} + \text{H}_2\text{O} \rightarrow \text{H}^+ + \text{FeOH}^+$ | -9.50 |
| $\text{Fe}^{2+} + \text{SO}_4^{2-} \rightarrow \text{FeSO}_4$ | 2.20 |
| $\text{Fe}^{2+} + 2\text{H}_2\text{O} \rightarrow 2\text{H}^++\text{Fe(OH)}_2^{2-}$ (aq) | -20.57 |
| $\text{Fe}^{2+} + 3\text{H}_2\text{O} \rightarrow 3\text{H}^++\text{Fe(OH)}_3^{2-}$ | -31.00 |
| $\text{Fe}^{2+} + 4\text{H}_2\text{O} \rightarrow 4\text{H}^++\text{Fe(OH)}_4^{2-}$ | -46.00 |
| $\text{Mn}^{2+} + \text{SO}_4^{2-} \rightarrow \text{MnSO}_4$ | 2.26 |
| $\text{Mn}^{2+} + \text{H}_2\text{O} \rightarrow \text{MnOH}^+ + \text{H}$ | -10.59 |
running the forward simulation repeatedly and the flow model calibration is carried out by the trial and error procedure of manually adjusting the input parameters selected for calibration, including those used to define initial and boundary conditions, at each time, until a satisfactory match between model results and field data is achieved. The slow reactions were represented by the kinetic expression and associated rate constants to address the dissolution reactions of key minerals present in the waste rock dumps and pits. The reaction network that describes the evolution of the geochemical system is shown in Table 3.

### 3.2.4 Conceptual modeling

The transport model for the mine site was built upon the flow model by including the necessary transport conditions, which includes the transport boundary conditions, the total number of components and the species to be simulated. The assumption is that the transport of the contaminants is based solely on the flow fields simulated whereby there is a possible rock interaction or mineral interactions with the aquifer rock bed and formation of daughter products or additional species.

It is assumed that the oxidative dissolution of pyrite is the driving force in the discharge rate of metal and radionuclides from waste rock dumps. The reactive system is completely defined by geochemical reactions through a network of reactions specifying chemical reactions and the total number of chemical species involved in the reaction. The standard equilibrium expression with a suitable equilibrium constant is used to represent all the fast reactions, such as the aqueous complexation reaction and precipitation of secondary phases. The slow reactions were represented by the kinetic expression and associated rate constants to address the dissolution reactions of key minerals present in the waste rock dumps and pits. The reaction network that describes the evolution of the geochemical system is shown in Table 2.

### 4. RESULTS AND DISCUSSION

The flow model calibration is carried out by running the forward simulation repeatedly and manually adjusting the input parameters selected for calibration, including those used to define initial and boundary conditions, at each time, until a satisfactory match between model results and field data is achieved. The trial and error procedure is used in this study. This is adopted because it is conceptually straightforward and requires nothing more computationally than making multiple forward simulations runs. This procedure is flexible allowing any adjustment in parameter values and parameter structures, including changes in mesh designs and the representation of the geologic framework.

The main aim of calibration was to obtain the estimation of hydraulic conductivity and groundwater levels for the aquifer based on limited field measurement data. Hydraulic head measurement data from 20 monitoring locations distributed across the mining area were used for the simulation model calibration. The hydraulic head data used for model calibration and validation were recorded in 2010 and 2014. A portion of the average rainfall intensity per year was specified as a recharge for calibration of the simulation model also using measured head data from monitoring locations. Calibration targets for the developed model were set to be within 2m intervals of the observed hydraulic head value in monitoring locations with a confidence level of 90%. Exact boundary conditions are difficult to determine with very limited observed measurement data. Hence, realistic boundary conditions need to be specified that matches the site conditions. In the calibration process, one of the most difficult issues is to assign appropriately the correct boundary conditions. Therefore, the boundary conditions need to be determined based on preliminarily calibration results as well. The model’s boundary conditions are adjusted manually to achieve the calibration targets. The measured and simulated heads were compared at selected points.

The developed model was calibrated for flow using observed head data obtained at monitoring points. In the calibration process, estimates of the values of recharge for each recharge zone and the representative hydraulic conductivity of each layer were calibrated. Results of simulated heads calibration are shown in Table 3.

| Location ID | observed head (m) | computed head (m) | Residual (m) |
|-------------|------------------|-------------------|-------------|
| P1a         | 61.44            | 62.48             | -1.04       |
| P1b         | 62.65            | 63.65             | -1          |
| P2          | 63.27            | 63.12             | 0.15        |
| P3          | 60.29            | 64.9              | -0.11       |
| P4          | 60.41            | 60.86             | -0.45       |
| P5          | 57               | 57.14             | -0.14       |
| P6          | 58.33            | 59.09             | -0.76       |
| P7          | 56.39            | 59.05             | -2.66       |
| P8D         | 53.96            | 54.68             | -0.72       |
| P9D         | 56.57            | 56.69             | -0.12       |
| P10         | 57.63            | 58.03             | -0.4        |
| P11         | 57.16            | 57.87             | -0.71       |
| P12         | 56.44            | 56.91             | -0.47       |
| P13         | 56.89            | 56.9              | -0.01       |
| P14         | 57.06            | 57.55             | -0.49       |
| P16         | 56.27            | 57.16             | -0.89       |
| P17         | 56.45            | 57.29             | -0.84       |
| P18         | 57.28            | 58.27             | -0.99       |
The flow model is calibrated first to adjust the model input parameters that control the flow process, such as hydraulic conductivity and recharge. When the flow model is calibrated to a certain degree, calibration of the transport model is initialized to adjust those parameters specific to the transport model, such as porosity, dispersivity and concentration. Recalibration of the flow model is done to further adjust the initially calibrated flow parameters. After readjustment of the flow parameters, a new velocity distribution is calculated and used in the transport model where further adjustments in the transport parameters are made. This iterative procedure is repeated until both flow and transport related field data are matched by the simulation results. The results of the calibrated heads against the observed heads is shown in Table 3. These results show fairly good history matching of the hydraulic heads. The reactive transport model simulation time is 2 years from 2012 to 2014 as of the availability of data. The chemical processes of aqueous complexation, precipitation-dissolution, adsorption-desorption, ion-exchange, redox, acid-base reactions and the changes of porosity, hydraulic conductivity, and hydrodynamic dispersion of aquifer media may be a key hydrogeochemical mechanism in the transport process. Thus, the effect of kinetic and equilibrium reactions on both flow and reactive transport are also considered in the simulations. Copper (Cu\(^{2+}\)), sulphate (SO\(_4^{2-}\)), Manganese (Mn\(^{2+}\)), Uranium (UO\(_2^{2+}\)) and Iron (Fe\(^{2+}\)) are the main species of study. The concentrations of each species at specific monitoring points is utilized as initial concentration which is assume to be involved in chemical reactions listed in Table 2. The assumed initial conditions for contaminant concentrations are specified in the simulation model based on the available data and extrapolation. The sources considered for this study are dyson waste rock damp, dyson open pit, main waste rock damp, intermediate waste rock damp, main pit and intermediate pit. The objective was to demonstrate the flexibility of the model to adapt a known reaction network to an abandon mine site setting and to demonstrate the capability of the code to simulate large complex problems. The results of the simulated concentrations are shown in the Figure 4, Figure 5 and Figure 6.

|     | Observed | Simulated |
|-----|----------|-----------|
| P19 | 57.35    | 58.23     | -0.88     |
| P20 | 51.01    | 51.69     | -0.68     |

5. CONCLUSION

The calibrated flow model, and the developed transport simulation model are capable of simulating the complex flow and multiple species reactive transport processes in the geologically complex multiple layer, heterogeneous aquifer comprising of a no longer in use uranium mine site in NT, Australia. The aquifer processes are considered as 3-dimensional, geologically layered and heterogeneous, with transient flow and transport. It was possible to calibrate the flow model by using sparse spatial-temporal hydraulic head measurements. The contamination simulation scenario resembled the field conditions to a certain extent. The concentrations predicted for the chosen species match with the concentration measurement values available. Therefore, the simulation model, based on HYDRGEOCHEM once calibrated for site conditions, is potentially a good candidate to
model the complex flow and transport processes in a geochemically and geologically complex mine site aquifer.

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