TECHNICAL ARTICLE

Geochemical Modelling of the Effects of a Proposed Uranium Tailings Pond on Groundwater Quality

K. Brindha · L. Elango

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Abstract The impact of a proposed uranium tailings pond on groundwater quality was assessed by geochemical modelling. Groundwater samples were collected from six dug wells in the Nalgonda district, Andhra Pradesh, in southern India, once every 2 months from March 2008 to January 2010, and analysed for calcium, magnesium, sodium, potassium, chloride, sulphate, carbonate, bicarbonate, and uranium. Prediction of groundwater quality was carried out for 100 years using PHREEQC to assess the effects of infiltration of water from the proposed tailings pond. The sensitivity of the model for variations in porosity, hydraulic gradient, hydraulic conductivity, and concentration of uranium in the tailings was evaluated. Geochemical modelling predicts that if the chemical composition of the tailings water is maintained at about the expected mean concentrations, and an appropriate liner is installed with an infiltration rate $<1.0 \times 10^{-9}$ m/s, the concentration of solutes in the groundwater will be increased from present background levels for a down-gradient distance of up to 500 m for the anticipated life of the mine, i.e. 16 years. The concentration of ions in groundwater would exceed background concentrations for up to 100 m at the end of 100 years. This study was used to predict the optimum chemical composition for the tailings and the extent, in terms of time and distance, that the groundwater concentration of various ions would be increased by infiltration of wastes from the tailings pond.

Keywords PHREEQC · Nalgonda · Uranium mining · Dissolution · Precipitation · Geochemical processes

Introduction

Exploitation of uraninite (uranium oxide) is being carried out in several parts of the world. Milling of the exploited uraninite lead to the generation of waste. Deterioration of groundwater quality due to the storage of waste in uranium tailings ponds has been reported (Lottermoser and Ashley 2005; Rodgher et al. 2013; Skipperud et al. 2013; Wang et al. 2012a, b). However, Tripathi et al. (2008) assessed the uranium concentrations around the Jaduguda mines, which are the only currently operating uranium mines in India, and found that its tailings pond had not adversely affected groundwater quality.

The spatio-temporal changes in mine water quality of an abandoned uranium mine in the Czech Republic were reported by Rapantova et al. (2012). Abdelouas et al. (1998) studied soil and groundwater samples from a tailings pond near Tuba City, Arizona, USA with regard to uranium adsorption and precipitation for planning a groundwater remediation strategy. The contamination of groundwater due to uranium tailings in Colorado, USA was reported by Goode and Wilder (1987). Dreesen et al. (1982) used both laboratory and field methods to understand the environmental transport and contamination resulting from the release of trace elements and radionuclides from uranium mill tailings at New Mexico, Colorado, and Utah mines, USA. Davis and Curtis (2007) analysed

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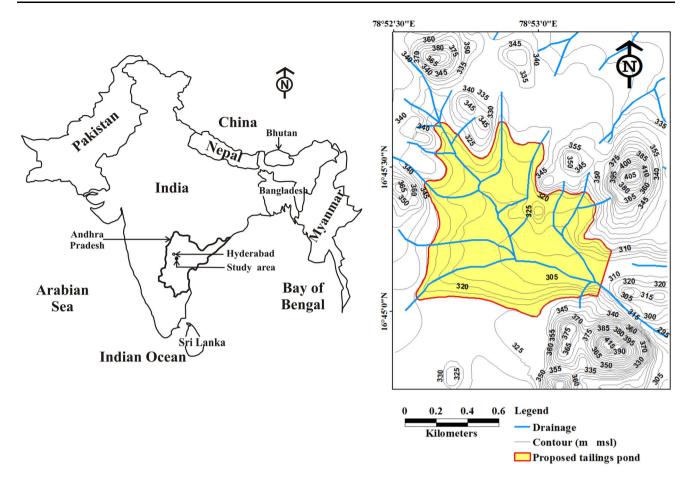


Fig. 1 Location and drainage pattern in the study area

the geochemical issues involved in restoration of ground-water at a uranium in situ leach mining facility to estimate the volume of water necessary to achieve restoration standards, which depends on the geochemical environment and the complexity of reactions that can occur during groundwater restoration. Reactive transport modelling of natural attenuation at a uranium mill tailings impoundment in Wyoming, USA was carried out by Zhu et al. (2001). A study carried out in the restored Los Ratones uranium mine of southwestern Spain (Gómez et al. 2006) brought out a model for uranium mobilisation.

These studies were carried out at existing or abandoned mines and tailings pond sites. Studies of this nature, if performed before mining commences, will allow suitable precautions to be taken to safeguard the groundwater environment. It is important to study the geochemistry of the groundwater before the storage of tailings, so that geochemical modelling based on intensive hydrogeological field investigations can be used to forecast possible changes in groundwater quality. Previous studies of this nature include the assessment of geochemical processes and the impact of uranium mines in Los Ratones Mine, Spain

(Gómez et al. 2003, 2006); Königstein Mine, Germany (Bain et al. 2001; Biehler and Falck 1999); Koongarra, Northern Australia (Payne and Airey 2006); Key Lake Mine, Canada (Schindler et al. 2013). With the availability of advanced geochemical modelling tools, such studies can be conducted to assess future impacts. We report here on a location in the Nalgonda district of Andhra Pradesh, India (Fig. 1), where uraninite mining and milling is planned in the near future. The objective of the study was to understand the potential impact of the tailings pond on groundwater quality and to propose the optimal chemistry of uranium tailings waste that would safeguard groundwater.

Methodology and Site Description

The hydrogeology of the study area was determined based on elaborate field work, well sections, lithologs, groundwater level monitoring, and constant rate pumping and infiltration tests. An intensive field survey was carried out, and nearly 30 wells were considered to choose appropriate sampling wells for continuous monitoring of groundwater

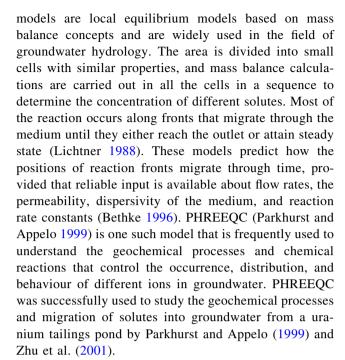


quality. Based on electrical conductivity (EC), six wells located near the proposed tailings pond site were selected for regular collection of groundwater samples once every 2 months, from March 2008 to January 2010. Groundwater level was monitored in these wells during sampling using a water level indicator (Solinst 101). Groundwater samples were collected in 500 mL capacity bottles. Prior to sampling, the bottles were cleaned by soaking them in 1:1 diluted HNO₃ for 24 h and then rinsing them thoroughly with distilled water. These bottles were rinsed again with the well water sample before filling it with the sample, which were collected 30 cm below the groundwater table from the dug wells. These groundwater samples were transported to the laboratory and filtered using 0.45 µm Millipore filter paper for the determination of Ca, Mg, Na, K, Cl, and SO₄ using a Metrohm 861 advanced compact ion chromatograph along with appropriate standards. Carbonate and bicarbonate concentration in groundwater was determined by titrating against H₂SO₄ as per standard methodology (APHA 1995). The uranium concentration in the water samples was determined using a laser fluorimeter, which has a detection limit of 0.1 ppb. Eluents, standards, and blanks were run frequently to check the accuracy of the procedures. For every 10 samples, three samples were run in triplicates to check for concordant readings. The concentration of the standard added to the samples was varied and a calibration curve was obtained to cross check the accuracy of the results. All chemicals used were of analytical grade and were procured from Merck. The complete analysis was checked by calculating an ion balance error, which was consistently <5 %.

The study area forms a part of a watershed of the Pedda Vagu River in Nalgonda district, Andhra Pradesh, India, which is located about 80 km ESE of Hyderabad (Fig. 1). This area has an arid to semi-arid climate. Temperatures range between 30 and 46.5 °C from April to June and between 16 and 29 °C from November to January. Average annual rainfall in this area is about 600 mm; most falls during the southwest monsoon (June-August). The drainage pattern of the area is generally trellis, with a dendritic pattern in certain parts. Granitic gneiss forms the major rock type. The tailings pond location was selected based on the conducive topography, drainage, and geological conditions (Gupta and Sarangi 2005). The location is surrounded by hills with drainage running from west to east, where construction of a bund has been planned to impound the uranium tailings.

Geochemical Modelling

Geochemical modelling is used to understand geochemical reactions that occur during groundwater flow. These



PHREEQC is capable of modelling transport processes including diffusion, advection, and dispersion and can combine these processes with equilibrium and kinetic chemical reactions (Parkhurst and Appelo 1999). This model solves the following advection-reaction-dispersion equation:

$$\frac{\partial C}{\partial t} = -v \frac{\partial C}{\partial x} + D_L \frac{\partial^2 C}{\partial x^2} - \frac{\partial q}{\partial t}$$
 (1)

where C is concentration in water (mol/kgw), t is time (s), v is pore water flow velocity (m/s), x is distance (m), D_L is the hydrodynamic dispersion coefficient [m²/s, $D_L = D_e + \alpha_L v$, with D_e the effective diffusion coefficient and α_L the dispersivity (m)], and q is the concentration in the solid phase (expressed as mol/kgw in the pores) (Parkhurst and Appelo 1999). The advective transport is represented by $-v\frac{\partial C}{\partial x}$, dispersive transport by $D_L\frac{\partial^2 C}{\partial x^2}$, and $\frac{\partial q}{\partial t}$ is the change in concentration in the solid phase due to reactions (q in the same units as C). It is assumed that v and D_L are equal for all solute species, so that C can represent the total dissolved concentration of an element, including all redox species (Parkhurst and Appelo 1999).

Conceptualisation

The region is made up four distinct layers: the soil zone, moderately weathered, highly weathered, and massive rock (Elango et al. 2012). The thickness of the soil zone ranges from 1 to 12 m and the thickness of the highly weathered granite layer ranges from 4 to 15 m. The groundwater level was generally 0.6–12 m



below ground level. The annual groundwater level fluctuation was observed to be around 6 m. The principle source of groundwater recharge is rainfall. The groundwater level rises 1–3 m during July to September, after the monsoon rains. Groundwater generally flows towards the southeast. Though there are many igneous intrusions in this area, they do not act as barriers to groundwater flow due the high intensity of weathering and because groundwater occurs at shallow depths (Rajesh et al. 2012). The dug wells of this area are 2.7–9.2 m deep and have diameters of 2–5 m. The yield of irrigation wells generally ranges from 100 to 150 m³/day, reaching up to 200 m³/day in a few places (CGWB 2007). Most of the wells in this area are used for irrigation.

Uranium occurring as uraninite ore is to be mined in the near future and will be beneficiated by chemical leaching methods. It is anticipated that 1,250 tonnes of ore will be mined a day (Elango et al. 2012). The average grade of uranium in the ore of this area is 0.052 % and a recovery of 90 % of the uranium in the ore is anticipated. A digital elevation model of the study site was made based on interpolating 10 m interval contours of a Survey of India topographic map. Making use of the digital elevation model, the surface area of the proposed tailings pond was determined for different elevations of the bund, running approximately north to south across the drainage between the two hills. A bund height of 16 m will be required to store the tailings that are likely to be produced during the expected operation of the mine, i.e. for 16 years. Figure 2 shows the area and volume of tailings that can be stored at a bund height of 16 m.

The conceptualised one-dimensional transport of ions along with the infiltrating tailings water into the

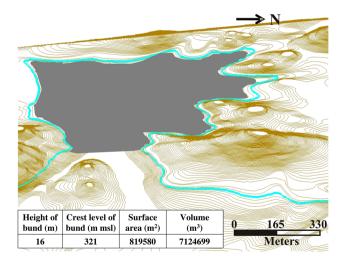


Fig. 2 Digital elevation model of the tailings pond area showing volume of tailings stored

groundwater zone was modelled by considering a column consisting of number of cells (Fig. 3). The number of cells and the length of each cell depend on the pore velocity. The model was formulated with that the first cell of the column at the bottom of the uranium tailings pond; cells continues up to the groundwater table, a distance of 4 m based on the general depth of the water table at this site. Then the column was continued along the groundwater flow direction to consider the transport of ions in the groundwater zone for a distance of 2,000 m. This distance was selected as the trial model runs indicated that geochemical processes associated with the infiltration of tailings water had no potential effect after this distance. Simulations were carried out with an annual time step, which was selected to keep required computational time reasonable, given that the aim of this work was to study the effect over a 100 year period. In many countries, tailings ponds are designed to control radiological hazards for at least 200 years and if possible up to 1,000 years (Abdelouas 2006). An even longer period, 10,000 years, is considered in the regulations of USA, Canada, and Germany.

Hydrogeological Properties

A groundwater table map of the study area was prepared from groundwater levels measured in wells every 2 months, from March 2008 to January 2010, and the hydraulic gradient at the tailings site was calculated based on these 12 values. The average hydraulic gradient was determined to be 0.0128, with a minimum hydraulic gradient of 0.0125 and a maximum of 0.013. The groundwater pore velocity beneath the tailings site was also estimated based on the measured groundwater levels.

The hydraulic conductivity of the formations of this aquifer was determined based on five pumping tests carried out as a part of this study and 12 pumping tests conducted in and around the study site by the Central Ground Water Board/Andhra Pradesh State Ground Water Board. The hydraulic conductivity varied widely, from 0.001 to 19 m/day, so an average value of 0.5 m/day was initially considered. Then, to understand the effect of extreme hydraulic conductivity values, the model was also run with a maximum hydraulic conductivity value of 19 m/day. The porosity of the subsurface formation was considered to range from 0.05 to 0.2, based on analysis of soil samples collected from the area.

A pore velocity, calculated from the hydraulic conductivity, porosity, and hydraulic gradient, was used to derive the number of cells necessary for geochemical modelling. The measured infiltration rate, which decides the movement of solutes from the tailings pond and in groundwater, was $21.47 \text{ cm/h} = 6 \times 10^{-5} \text{ m/s}$, which is very high. This



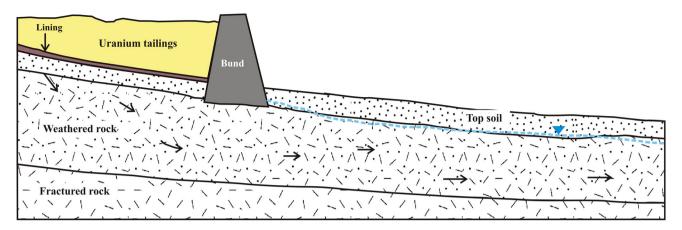


Fig. 3 Conceptual model of the proposed tailings pond

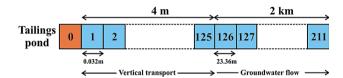


Fig. 4 One dimensional discretisation of the tailings pond site

infiltration rate would create a high risk of groundwater contamination. To avert this, liners should be used to limit infiltration rates. This was assessed by using different values for the infiltration rates during the geochemical modelling, starting at 6×10^{-5} m/s. By this iterative trial and error method, it was determined that an infiltration velocity of 1.0×10^{-9} m/s would be both safe and achievable with liners; this would limit the quantity of recharge of the tailings water from the pond to 69.19 m³/ day. For the cells that start at the bottom of the tailings pond and continue up to the groundwater table, i.e. a vertical flow of up to 4 m, the cell length depends on the calculated infiltration velocity of 1.0×10^{-9} m/s. Thus, for a time step of 1 year, the length of each cell was 0.032 m. For the total length of 4 m, 125 cells, each 0.032 m long, were used (Fig. 4). It was assumed, as an extreme scenario, that the cells beneath the tailings pond were saturated with the tailings water before the simulation commenced.

Based on the estimated groundwater pore velocity, an average hydraulic gradient of 0.0128, and a hydraulic conductivity of 0.5 m/day, the flow path comprises 86 cells, each 23.36 m long (Fig. 4). The diffusion coefficient of all the aqueous species considered for this study was taken as 1×10^{-10} m²/s, which is considered to be a reasonable value for the geological materials of the study

site. The dispersivity was similarly estimated at 0.5 m based on the geological materials. The boundary conditions were assumed to be constant at the top and in flux at the end.

Groundwater Chemical Composition

Groundwater samples were analysed from March 2008 to January 2010 by systematic collection of samples every 2 months. The aim of the study was to understand the potential effect of tailings water on groundwater. Hence, the sample that had the highest concentration of all ions was taken as the chemistry of groundwater in this area. This admittedly conservative approach was taken to enable us to determine the maximum impact that the tailings pond might have on groundwater quality.

The geochemical processes that are responsible for the chemical composition of groundwater of this area were also studied (Rajesh et al. 2012). Weathering and dissolution of silicate minerals control the concentration of major ions such as calcium, magnesium, sodium, and potassium, but ion exchange process was also responsible for variations in the calcium and sodium concentrations. The identified processes were also verified by carrying out inverse geochemical modelling. The chemical composition of groundwater in two pair of wells along the general groundwater flow direction of the tailings pond was used for this purpose. Inverse modelling indicated that calcite precipitation was responsible for the change in the chemical composition between these wells. Hence, ion exchange and mineral phases such as calcite and uraninite were considered while carrying out the forward geochemical modelling of the effects of infiltration of uranium tailings into groundwater.



Table 1 Chemical composition of groundwater and tailings pond water

Parameter	Groundwater	Tailings pond			References	
		Min	Mean	Max		
рН	8.4	7.5	8	8.6	Annual Health Physics Report for the year 2004 (2005)	
Ca (mg/L)	22.06	_	310	-	Zhu (2003)	
Mg (mg/L)	62.42	_	1,000	-	Zhu (2003)	
Na (mg/L)	251.07	_	360	_	Zhu (2003)	
K (mg/L)	2.67	_	60	_	Zhu (2003)	
Cl (mg/L)	147.36	92	139	178	Annual Health Physics Report for the year 2004 (2005)	
SO ₄ (mg/L)	74.27	380	662	1,034	Annual Health Physics Report for the year 2004 (2005)	
HCO ₃ (mg/L)	469.7	_	6.3	_	Annual Health Physics Report for the year 2004 (2005)	
U (μg/L)	118.4	1	3.8	7.1	Annual Health Physics Report for the year 2004 (2005)	

Table 2 Different parameters for model prediction; mean used for scenario 0

Parameter	Minimum	Maximum	Mean	Scenario
Porosity	0.05	0.2	0.1	1
Hydraulic gradient	0.0125	0.013	0.0128	2
Hydraulic cond. (m/day)	0.001	19	0.5	3
Uranium conc. in tailings waste	Mean values of all ions and min. value of U, as per Table 1	Mean values of all ions and max. value of U, as per Table 1	Mean values of all ions, as per Table 1	4

Chemical Characteristics of Tailings Water

As mining and milling has not yet commenced, the chemical composition of uranium tailings were taken from existing literature (Table 1), i.e. Annual Health Physics Report for the Year 2004 (2005); Zhu (2003). The mining is proposed to be carried out for 16 years, so it was assumed that the waste will only be dumped in the tailings pond during that period. Hence, after that, the level of tailings water will decrease due to infiltration. The decreased level of the tailings water is likely to be replaced by rainwater, which will dilute the concentration of ions in the tailings pond. The dilution in the concentration of ions in the water of tailings pond due to the mixing of rainfall was calculated for every 5 years (a 5 year period was used for computational convenience). Based on the annual rainfall (600 mm) and the infiltration rate at the pond, the volume of tailings water that will infiltrate into the formation over 5 years was estimated to be about 3.75 % of the total volume of the tailings pond. The chemical composition of rainwater was obtained from Kulshrestha et al. (2003).

Model Calibration and Validation

In order to validate the model developed for the study site, the change in the chemical characteristics of groundwater due to the storage of uranium tailings needs to be known with respect to time. But such data is unavailable since the uranium tailings pond is not yet constructed. At this stage though, the model results are primarily for understanding the possible implications that infiltration from the tailings pond might have on groundwater chemistry, rather than precisely determining the groundwater composition. Therefore, inverse modelling was carried out to identify the geochemical processes that would change the chemical composition of groundwater during its movement, using literature values for tailings water chemistry. The results concur with the processes reported by Rajesh et al. (2012).

Model Predictions

The PHREEQC code was initially populated with the anticipated chemical composition of the tailings waste and



the site's hydrogeological properties (Table 2). The different parameters considered for model prediction and corresponding model scenario are given in Table 2. The parameters used in the model actually vary by several orders of magnitude so, to study the effects of the more extreme values, the model was run assuming various scenarios (Table 2). The impact of the tailings pond on groundwater quality was assessed based on the distance at which the concentration of ions was found to be greater than background concentrations (Table 1).

Scenario 0: Mean Concentration of Ions in the Tailings Pond

In this scenario, the average values of the hydrogeological properties of the medium were used. For this scenario, based on the hydraulic gradient (0.0128), hydraulic conductivity (0.5 m/day), and porosity (0.1), the cells size worked out to be 23.36 m. The predicted concentration of a few ions in groundwater with respect to distance from the tailings pond is shown in Fig. 5. It can be noted from

Table 1 that the concentration of uranium, bicarbonate, and chloride are higher in the groundwater than in the tailings waste. The concentration of these ions increases along the flow direction and reach the level of the groundwater after some distance. The calcium concentration decreases from 310 mg/L at the tailings pond to 22 mg/L at a distance of 20 m by the end of 1 year, causing the water to become undersaturated with respect to calcite. The calcium concentration falls to groundwater levels at 260 and 500 m at the end of 8 and 16 years, respectively. Simulated trend of magnesium, potassium and sulphate concentrations with respect to distance resemble that of calcium. Sodium concentration is predicted to be higher than the measured groundwater concentration for 50 years; however, it does decrease over time due to dilution by rainwater. In fact, dilution, along with infiltration, causes the concentration of all ions in the tailings pond to decrease after tailings disposal stops. Thus, no adverse effects were predicted due to infiltration of tailings on groundwater quality at a distance of 500 m from the tailings pond.

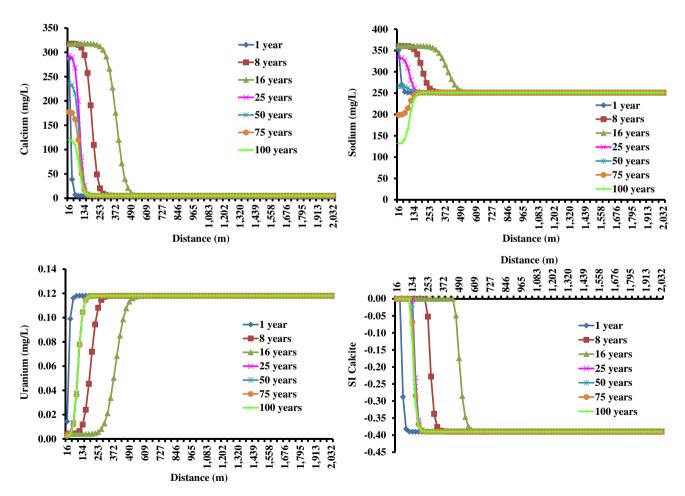


Fig. 5 Simulated concentrations of a few ions and SI of calcite in groundwater, assuming mean concentrations of ions in the tailings pond, based on the cited literature



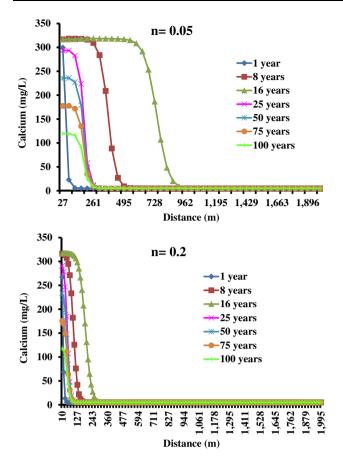


Fig. 6 Simulated concentrations of calcium with minimum and maximum porosity ($n=0.05,\,0.2$)

Scenario 1: Effect of Variation in Porosity

Porosity of the formation controls the velocity of ground-water flow. The soil and weathered rock of this region have a porosity ranging from 0.05 to 0.2 from analysis of soil samples. In this scenario, the model was run while altering the porosity values. The predicted concentration of various ions in groundwater with respect to distance from the tailings pond with a minimum porosity of 0.05 is shown in Fig. 6. With this minimum porosity, the transport of ions reached 1 km at the end of 16 years. At the highest expected porosity, 0.2, the effect of the tailings would not be noticeable at a distance of 250 m at the end of 16 years (Fig. 6).

Scenario 2: Effect of Variations in Hydraulic Gradient

Hydraulic gradient is the driving force of groundwater flow and hence is potentially an important parameter, since groundwater flow velocity depends on the hydraulic gradient. At the minimum hydraulic gradient (i=0.0125) based on the measured groundwater levels, the model predicted that the concentration of ions would reach groundwater concentrations at 470 m by the end of 16 years (Fig. 7). With

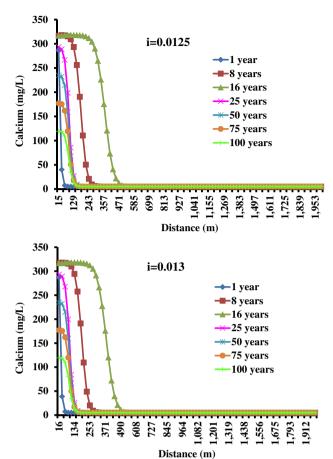


Fig. 7 Simulated concentrations of calcium with minimum and maximum hydraulic gradient (i = 0.0125, 0.013)

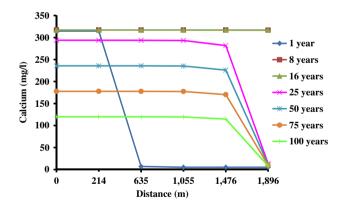


Fig. 8 Simulated concentrations of calcium with a hydraulic conductivity of 19 m/day

dilution due to precipitation, the concentration of ions in groundwater would reach background concentrations at 130 m at 25, 50, 75, and 100 years. At a higher hydraulic gradient (i = 0.013), the ions will be elevated for 490 m down-gradient at the end of 16 years, and for 140 m in the end of 100 years (Fig. 7). Thus, the model predicted that the



effect of the tailings would not be noticeable at a distance of 500 m. When the maximum hydraulic gradient was considered, the model predicts that the ions will be elevated for about 10 m more than with the minimum hydraulic gradient.

Scenario 3: Effect of Variations in Hydraulic Conductivity

Hydraulic conductivity is another important parameter, the values of which are based on limited number of pumping tests. As previously discussed, the maximum measured hydraulic conductivity of 19 m/day was used for this scenario. Figure 8 shows the simulated concentration of ions in the down-gradient direction. At the high assumed hydraulic conductivity, the model predicts that the concentration of ions in groundwater could be elevated for as much as 2,000 m from the tailings pond.

Scenario 4: Variations in the Concentration of Uranium in Tailings Pond

The maximum and minimum concentration of uranium in tailings waste from the Annual Health Physics Report for the Year 2004 (2005) were considered for this scenario. The concentration of uranium in groundwater of this region is naturally elevated due to the uranium levels in the rock strata (Brindha and Elango 2013; Brindha et al. 2011), whereas the concentration of uranium in tailings pond water is less (Table 1). Simulated concentrations of uranium in groundwater with respect to distance from the tailings pond indicate that the effect of the tailings water is not noticeable at a distance of about 500 m after 16 years (Fig. 9); this distance is predicted to shrink to about 150 m after 100 years.

Limitations of this Study

A study of this type always has some limitations. In this case, the limitations include: (1) possible external causes for changes in groundwater chemical composition, such as application of agrochemicals, domestic waste, and other anthropogenic sources; (2) radioactive decay of uranium was not considered; (3) due to computational limitations, only one dimensional transport along the principle direction of groundwater flow was considered, and: (4) thermodynamic equilibrium was assumed and reaction kinetics were not considered. In spite of these limitations, which are very difficult to overcome in a study of this nature, this research has provided insight into some potential problems that could develop due to storage of the mine wastes at this site. For example, the results of the simulation with mean values of porosity, hydraulic conductivity, hydraulic gradient and concentration of ions in the tailings pond is shown in Fig. 10. This indicates that there is no effect of

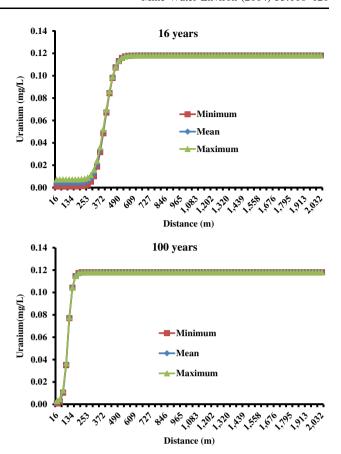


Fig. 9 Simulated concentrations of uranium in groundwater assuming anticipated minimum, maximum, and mean concentrations in tailings pond, based on the cited literature

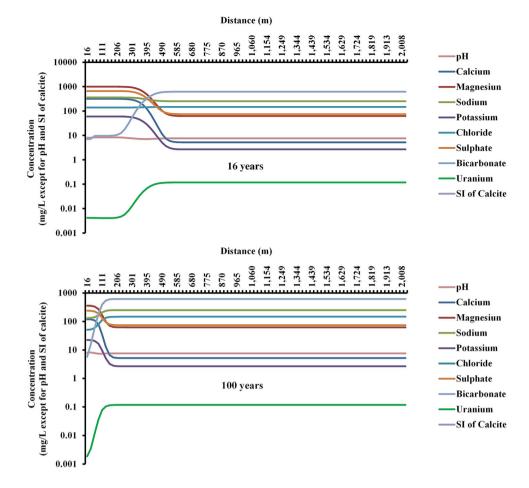
storage of tailings on groundwater quality beyond a distance of about 500 m from the tailings pond.

Conclusion

Geochemical modelling was used to estimate the concentration of ions in groundwater due to the expected seepage from the proposed uranium tailings pond area. The aim of this work was not to predict the exact values of solute concentrations in groundwater but to understand the effects that infiltration of water from the uranium tailings pond might have on groundwater chemistry. Such a study is useful in planning and designing a uranium tailings pond that will have minimum impact on the groundwater environment. As explained earlier, if the chemical composition of the tailings water is about the mean concentration given in scenario 0, there will no threat to groundwater quality beyond 500 m from the tailings pond area at an infiltration rate of 1.0×10^{-9} m/s. Hence, the liners should be designed and placed to achieve an infiltration rate $<1.0 \times 10^{-9}$ m/s. After tailings disposal stops, the groundwater quality should gradually improve and should reach the pre-tailings pond



Fig. 10 Simulated concentrations of ions with optimum parameters at the end of 16 and 100 years



operation levels at about 100 m after a period of 100 years. This study helped to understand the effect of natural geochemical processes and leaching of tailings waste on groundwater quality. Thus geochemical modelling has given an insight in understanding the possible future effects of storage of uranium tailings on groundwater quality.

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