Ekoloji 28(107): 829-836 (2019)

Environmental Impact Assessment of Leachate from Waste Rock Piles on Groundwater Using Numerical Model, Case Study Anhui, China

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Abstract

This study evaluates the transport of leaching water infiltration from waste rock piles on the surrounding groundwater, using Anhui, China, as an example. Based on a detailed analysis of hydrogeological conditions in the study area, a numerical groundwater model was established to simulate the transport of a specific pollutant (manganese). The distribution and levels of manganese pollution in the groundwater environment caused by the infiltration of leachate were assessed for different operating conditions using MODFLOW and MT3D module. The results show that without anti-seepage measures in place in the waste rock piles, the concentration of manganese in the center point of the rock pile reaches the maximum allowable value of 0.1mg/l by the second year. The maximum horizontal diffusion distance was 188 m. The concentration of manganese in the center point reached 0.45 mg/l by the tenth year, which is 4.5 times more than the allowable value, and the diffusion area was approximately 0.06 km^2. As a result of leachate infiltration, bedrock aquifers in the study area will be polluted. However, if anti-seepage measures are in place with an impermeable layer with a vertical permeability coefficient of 1×10^{-7}cm/s, then the maximum concentration of manganese in the center of the waste rock piles is reduced to 0.08 mg/l by the tenth year. This is within the allowable limits. Therefore, we concluded that leachate from the waste rock piles will lead to high levels of manganese in the underlying aquifers, and that anti-seepage measures must be taken to prevent this.

Keywords: leachate, groundwater, numerical model, waste rock piles, Anhui

Shang M, Liu P, Li F, Chen X (2019) Environmental Impact Assessment of Leachate from Waste Rock Piles on Groundwater Using Numerical Model, Case Study Anhui, China. Ekoloji 28(107): 829-836.

INTRODUCTION

To ensure the safety of mining activities, drainage is required. However, the drainage of mining areas causes deep drawdowns, which are highly likely to negatively impact the hydrogeological conditions in the mining areas. Because of the deep drawdowns and changed conditions, a large cone of depression will have been created. The discharge of large quantities of harmful mine water, chemical wastewater, and leachate directly pollutes local water systems, and causes a deterioration of the surrounding water environment, endangering aquatic life, and negatively affecting the safe use of the water by humans and livestock.

Mining can directly lead to surface deformation as a result of unloading (Cuenca et al. 2013, Li et al. 2017, Tiwari et al. 2017), which then generates a large number of secondary fractures. As a result, wastewater and precipitation infiltrates to aquifers through fractures, thus causing the aquifers to be polluted. Organic matter contained in the wastewater can migrate 800–1000 m in highly permeable deposits and fractures (Yang et al. 2015). Therefore, if wastewater directly enters the aquifers without purification, it will lead to a deterioration in groundwater quality. Untreated wastewater in mines contains heavy metals in various concentrations. If the concentrations of these toxic elements and heavy metal elements exceed the allowable limits of the relevant standards, these elements will pollute groundwater (Abraham and Susan 2017, Kusin et al. 2018). For example, a groundwater assessment of 15 heavy metals, including As, Ti, Co, and Ni, in abandoned metal mines in Morocco found As concentrations greatly exceeded groundwater quality standards, and that the groundwater environment was polluted (Julien et al. 2017). Zhang et al. (2018) studied the characteristics of the migration and distribution of heavy metals in iron tailings using a single-factor index

Received: 7 Apr 2018 / Accepted: 28 Jun 2018

and comprehensive index method, and found that, in the same soil, the five heavy metals showed different migration trends in the vertical direction (Cd> Zn> Cr> Pb> Cu). While these elements all have a strong migration capacity in the horizontal direction, Cr has the strongest capacity to migrate under specific climatic conditions. In addition, acid mine water produced during mining can also affect the groundwater environment (Atulya et al. 2018, Han et al. 2017). The pH value can change the water chemistry and composition of river sediments, and also negatively impact the water quality of hydrological systems such as streams and oceans. Sulfate ions generated under acidic conditions cannot easily adsorb and precipitate, so high levels of pollution can still exist after 20 years (Franco et al. 2017). The rock permeability directly affects the transport distance and range of pollutants. Karel et al. (2015) simulated the migration of contaminants in porous media after underground coal gasification had taken place in shallow coal seams. They input vapor permeability, liquid advection, axial diffusion, and adsorption parameter values, and evaluated the impact of aperture, porosity, tortuosity, and reaction conditions on migration. They concluded that porosity, aperture, and tortuosity are key transport parameters that play a major role in the migration of pollutants.

When pollutants enter the groundwater, they cause the water environment to deteriorate, and also threaten aquatic life and the safety of water use by humans and livestock. The pollutants are likely to pose a long-term, carcinogenic risk (Cooke et al. 2016). When the pollution reaches a certain level, the species richness near mine areas will be reduced (Mario et al. 2014, Steven et al. 2016). Odukoya et al. (2017) did a case study on a mine in southern Nigeria, and made health risk assessments of toxic elements such as Pb, As, Co, Cr, and Co in surrounding waters. They found that the levels of Fe, Ba, Mn, Pb, Cr, and Ni exceeded 62.5%, 18.25%, 37.5%, 12.5%, 6.25%, and 6.25%, respectively, of the human drinking water standards, and that As exceeds the acceptable carcinogenic risk for children and adults.

Waste rock piles in mining areas can also be a source of heavy metal pollution. For example, Gao et al. (2014) studied coal gangue leachate at the Heshan City East Mine, and found that the concentrations of Cd, Ni, Pb, and As far exceeded the concentrations in the water used as a washing solution. Precipitation falling on the waste rock piles will generate large amounts of leachate. It is a common assumption that when leachate seeps into the ground through the unsaturated zone, some of its chemical components will be filtered, adsorbed, and purified, and therefore it will have a minimal impact on groundwater. Thus, the impact of leachate on the groundwater environment may not receive sufficient attention (Qin 2015). However, unconsolidated Quaternary sediments and weathered bedrock in hilly areas can be relatively thin, and thus the purification of groundwater is limited. Omar et al. (2012) compared an experimental column with field observations from disused mines to investigate the leaching behavior of Cu, Pb, and Zn. Their results showed that the leaching of Cu and Zn increased as the length of the flow path through the weathering terrain increased, and that Pb concentrations are limited by insoluble sulfate precipitation. They therefore concluded that the thickness of weathered rock is an important parameter in the infiltration of potentially toxic metals to aquifers. Cheong et al. (2012) showed that a thickness of 20 cm of soil in the mining area was insufficient to prevent rainwater infiltrating into the aquifer. Therefore, if the chemical components of leachate directly enter groundwater through fractures without being adsorbed and purified, they will have a major impact on groundwater quality. The above studies point out that although it is easy for excessive amounts of heavy metals to be produced in waste rock leachate, the migration and transformation of pollutants entering aquifers has not been sufficiently analyzed and simulated in hilly areas, where the soils are generally thin, and bedrock may be exposed.

There are many types of deposits, and different pollutants have different physical and chemical properties, and therefore they have different impacts on the groundwater environment. Most of the progress in the study of pollutants in mining areas is based on acidic mine water. Much less research has been done on the transport of pollutants in leachate in iron ore mines. The present study aims to help address these research imbalances, and quantitatively evaluates the environmental impact of characteristic waste-rock-pile leachate pollutants on groundwater in mining areas in hilly terrain. The waste rock piles at the Longqiao Mine, where iron ore is mined, were used for a case study. This mine is located in a hilly area and the thickness of weathered bedrock is limited. A two-dimensional groundwater flow and solute transport model was established. Based on the waste rock leaching test results, we analyzed and evaluated manganese transport in leachate using the MODFLOW and MT3D models. We believe that the results of this study will provide a scientific basis for improved implementation of anti-

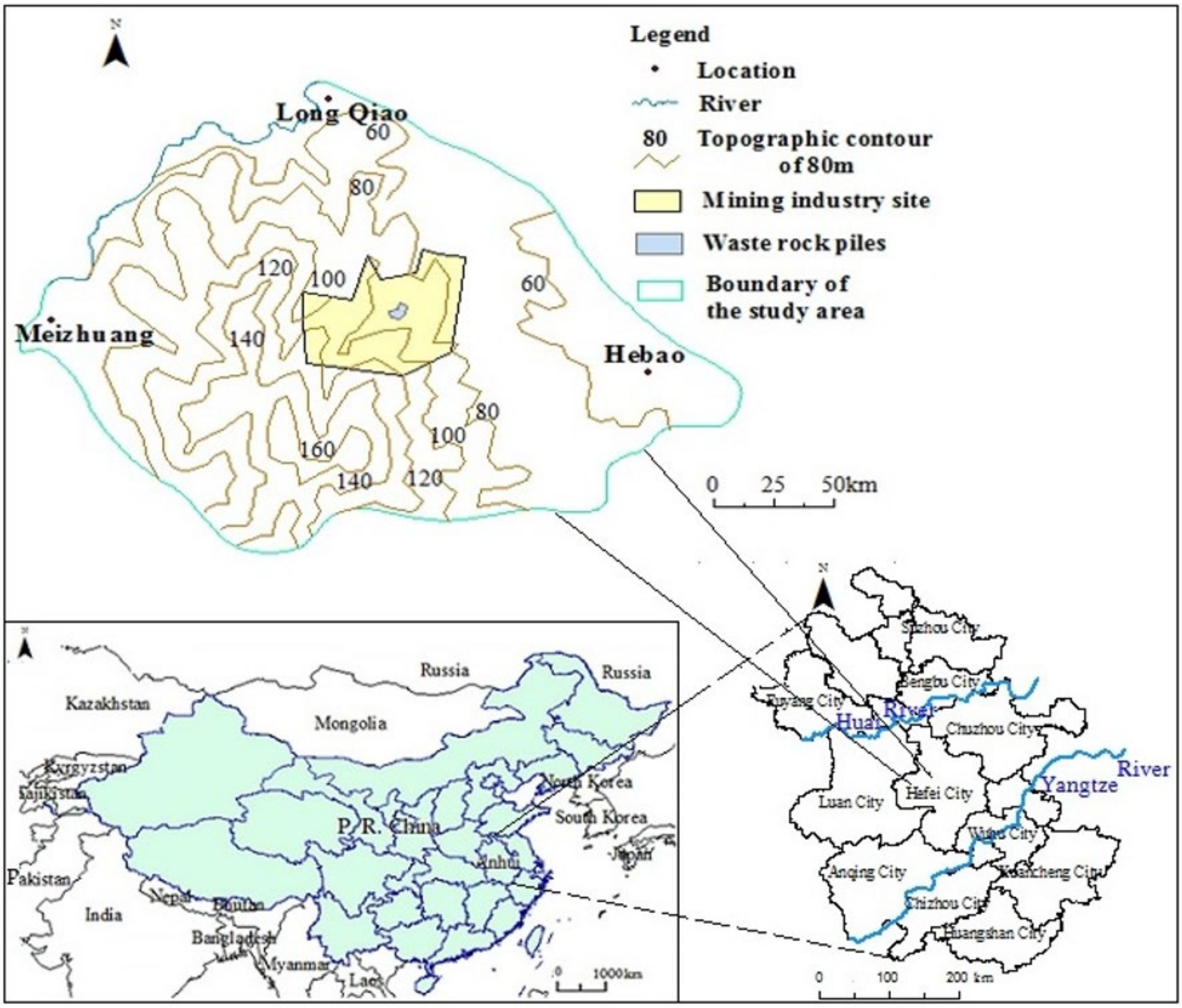


Fig. 1. Map of the study area

seepage measures, and the protection of groundwater quality, in mining areas.

MATERIALS AND METHOD

Study Area

As shown in **Fig. 1**, the study area is located in a low-lying hilly area. The maximum elevation is 206.59 m a.s.l., the minimum elevation is 16.90 m a.s.l., and the relative elevation is 189.69 m. The study area has a humid monsoon climate, and four seasons are discernible. From 1978 to 2016, the average annual rainfall was 1248.2 mm, the average annual evaporation was 1402.3 mm, and the relative humidity was 75%–80%. Most of the rivers in the area are seasonal, and the flood season is controlled by heavy rain in the hills, causing rapid increases and declines in flood flows. When the precipitation in the non-flood season decreases, the upstream water is mainly derived from base flow.

Aquifers in the research area can be divided into four main types: unconsolidated, fractured clastic rock, fractured carbonate rock, and fractured ore bodies. The unconsolidated aquifers consist of alluvial clay and sub-clay from the Quaternary, are commonly covered with gravel, and have a thickness of 0 to 20 m. The fractured clastic rock aquifers mainly consist of Zhuanqiao volcanic rocks beneath a layer of tuff and tuffaceous siltstone, and have a thickness of 60 to 200 m. The fractured carbonate rock aquifers are mainly comprising Luoling limestone breccia, limestone, marlstone, and marble limestone, and have a thickness of 1.25 to 115.65 m. A muddy siltstone usually caps the carbonate rock aquifer. The development and depth of fractures controls aquifer productivity. The fractured ore body aquifers are mainly composed of Zhuanqiao Group trachyandesites, Longmenyuan Group trachyandesites, porphyry, and tuff, and Luoling Group muddy siltstones and magnetite bodies. The thickness of the fractured ore body aquifers is greater than 650 m. While

Table 1. The test results of leachate from waste rock piles

Index	Copper	Zinc	Cadmium	Lead	Chromium	Hexavalent	Mercury	Mercury	Silver
Detected or not	No	No	No	No	No	No	No	No	Yes
Index	Barium	Nickel	Beryllium	Arsenic	Selenium	Manganese	Cyanide	Fluoride	Sulfide
Detected or not	Yes	No	No	Yes	No	Yes	No	Yes	Yes

each aquifer type has distinct hydrogeochemical characteristics, overall their water chemistry is broadly similar: the cations are mainly calcium, magnesium, and sodium, and the water is of a low sodium, bicarbonate type. All the aquifers are connected hydraulically, and there are no obvious impermeable boundaries.

Different kinds of mineral deposits are distributed and concentrated in the study area. The tailings produced by the Longqiao Mine are all used to make bricks, and to fill underground goafs. Mine drainage water is used as recycled water for the mining industry. Domestic sewage and industrial wastewater are also reused, after purification, by the mining treatment system, and therefore have little impact on the groundwater environment. Thus, the main source of water pollution in the study area is leachate generated from waste rock piles. The waste rock piles cover an area of is 0.36 km^2. Their volume is about 450,000 m^3, and the piles contain about 450,000 tons of waste rock and vermiculite.

Evaluating Indices

Sample collection

Eight waste rock leaching tests were conducted to detect the content of various substances in the waste rock. The test results are shown in **Table 1**. **Table 1** reveals that the heavy metals were silver, barium, arsenic, manganese, fluoride and sulfide in the waste rock. And no other heavy metals have been detected.

Single-factor evaluation index method

Single-factor evaluation index method (Li et al. 2016, Yang et al. 2012) was used to evaluate the results of the above eight tests. The evaluation index for each evaluation factor (except pH) is calculated as:

$$S_i = \frac{C_i}{C_{oi}} \tag{1}$$

where S_i is the single-factor standard index for the i-th evaluation factor; C_i is the measured concentration value for the i-th evaluation factor (mg/L); and C_{oi} is the environmental quality allowable value of the i-th evaluation factor (mg/L).

The standard index of pH is calculated by the following formula:

$$S_{pH,j} = \frac{7.0 - pH_j}{7.0 - pH_{sd}} \; (pH_j \le 7.0) \tag{2}$$

$$S_{pH,j} = \frac{pH_j - 7.0}{pH_{su} - 7.0} \; (pH_j > 7.0) \tag{3}$$

where $S_{pH,j}$ is the pH standard index of point j; pH_{sd} is the lower limit of pH in water quality standards and pH_{su} is the upper limit of pH in water quality standards, respectively, pH_j is the measured value of the pH value of point j.

If the standard index of the evaluation factor is less than or equal to 1, then it meets the standard requirements for groundwater quality. While if the standard index of the evaluation factor is more than 1, it is excessive.

NUMERICAL SIMULATION MODEL

Hydrogeological Conceptual Model

Using the hydrogeological data from the study area, the spatial distribution of the different aquifers was combined, and no obvious aquifuges were detected. There was some hydraulic relationship between aquifer petrofabric and groundwater in the same hydrodynamic field. Rainfall infiltration is the only source of recharge. We generalized the aquifers to a unified, integrated aquifer system with four layers. The first layer is an unconsolidated porous aquifer, the second is a fractured clastic rock aquifer, the third is a fractured carbonate rock aquifer, and the fourth is a fractured ore body aquifer. In its natural state, groundwater in the study area flows from south to north. The east and west of the study area are bounded by ridges, and were set as zero flow boundaries. The north of the study area is bounded by the West River, and was designated a constant head boundary. We assumed that the watershed 2.5 km from the southern boundary of the mining area was an impervious boundary because the altitude in the south is higher. The upper boundary of the simulation area was generalized as a rainfall infiltration supply and evaporation boundary. The bottom of the mine was set as the lower boundary because permeability is very low there.

Groundwater Flow Mathematical Model

Based on a hydrogeological conceptual model, we generalized the groundwater flow in the simulation area as two-dimensional, heterogeneous, anisotropic, and unsteady. The corresponding mathematical model is:

$$\begin{cases} \frac{\partial}{\partial_x}(K_x\frac{\partial H}{\partial x}) + \frac{\partial}{\partial_y}(K_y\frac{\partial H}{\partial y}) + w = \mu_s\frac{\partial H}{\partial t} \\ H(x,y,t)|_{\,t=0} = H_0(x,y),(x,y,z) \in D \\ h(x,y,t)\big|_{\,\Gamma_2} = h(x,y,t),(x,y) \in \Gamma_1, t \geq 0 \\ KM\frac{\partial H}{\partial n}\Big|_{\,\Gamma_2} = q(x,y,t),(x,y) \in \Gamma_2, t \geq 0 \end{cases} \tag{4}$$

where K_x and K_y are the components of the hydraulic conductivity in the x and y directions, assuming that the main axis of the permeability coefficient coincides with the direction of the coordinate axis (m/d); $H$is the groundwater head; and $w$is the unit volume flow that represents the amount of water flowing into or out of the sink. In addition, μ_s is the aquifer storage rate (L/m); t is the time (day); H_0 is the initial head and $h$is the boundary head; $M$is the aquifer thickness, $D$is the simulation area, Γ_1 is the first boundary and Γ_2 is the second boundary; and q is the boundary flow.

Groundwater Solute Migration Mathematical Model

We established a mathematical model of the two-dimensional hydrodynamic dispersion equation for groundwater solute migration based on the most adverse principle. The dissolved adsorption and chemical reactions of pollutants have been ignored. The model is:

$$\theta\frac{\partial c}{\partial t} = \frac{\partial}{\partial x}(\theta D_{xx}\frac{\partial c}{\partial x}) + \frac{\partial}{\partial y}(\theta D_{yy}\frac{\partial c}{\partial y}) - \frac{\partial(\theta\mu_x c)}{\partial x} - \frac{\partial(\theta\mu_y c)}{\partial y} - W_c \tag{5}$$

Initial condition: $c(x,y,0) = c_0(x,y),(x,y) \in \Omega, t = 0$

Boundary condition: $(c\vec{v} - Dgradc)\cdot\vec{n}|_{\,\Gamma_1} - \varphi(x,y,t),(x,y) \in \Gamma_2, t \geq 0$

where θ is the porosity of the medium, c is the solute concentration, D_{xx} and $D_{yy}$are the diffusion coefficients for the x and y directions (m^2/d). Respectively, μ_x and μ_y are the actual seepage speeds for the x and y directions (m/d), respectively, c_0 is the initial solute concentration (mg/L), Ω is the solute flow area, $\vec{v}$ is the seepage velocity, D is the hydrodynamic dispersion coefficient, $\vec{n}$ is the outside normal direction of the boundary, and φ is the solute flux of the boundary. In this model, the first equation can be calculated when combined with the initial conditions.

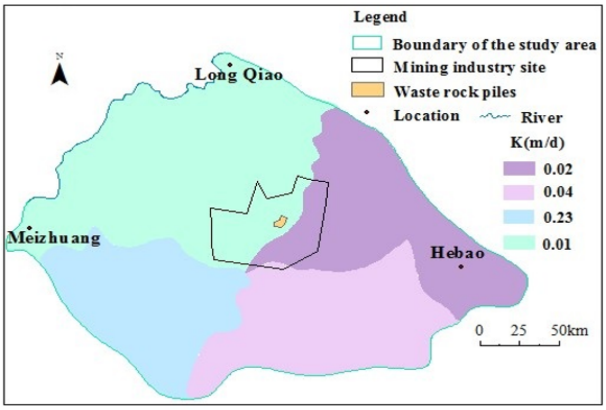


Fig. 2. The model validation results of hydraulic conductivity

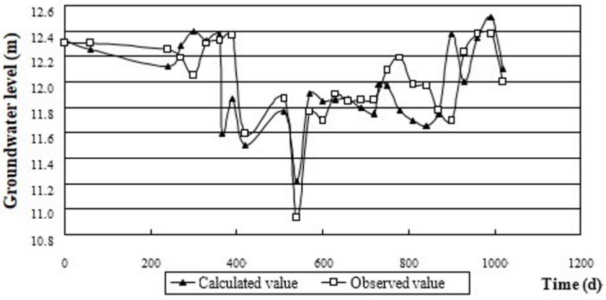


Fig. 3. The model validation results of groundwater levels

Model Validation

After the model was established, it was validated using data collected from the study area. The numerical models results should be compatible with actual hydrogeological conditions. The model validation results obtained by the trial and error method are shown in **Fig. 2** and **3**. **Fig. 2** and **3** shows that the overall trend for changes in groundwater levels calculated by the model were consistent with the actual situation, based on the fitting curves. The absolute error in the water levels at most fitting points was less than 0.5 m, and the simulation of each observation hole was good. The parameters for the different zones can thus reflect the hydrogeological conditions in the simulation area. Therefore, the established numerical model can be used for groundwater prediction analysis in the study area.

RESULTS AND DISCUSSION

Pollutants

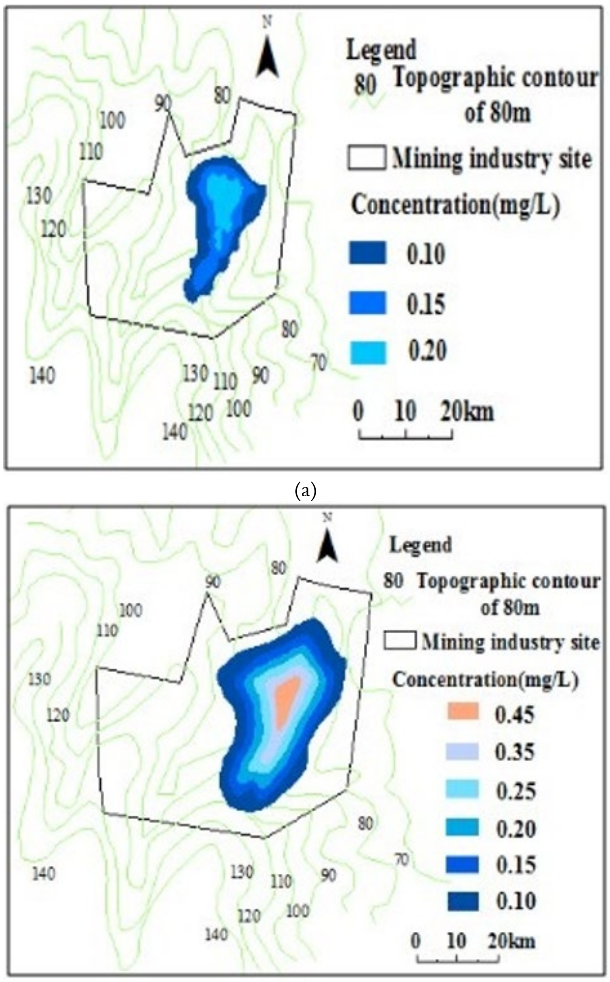
From the results of eight waste rock piles tests, it can be seen that the main factors affecting the groundwater environment are the heavy metal elements in the study area, which mainly included silver, barium, arsenic, manganese, fluoride and sulfide.

Table 2. The results of Single-factor evaluation index method

Index	Barium	Silver	Arsenic	Manganese	Fluoride	Sulfide
C_{oi}	0.70	0.05	0.01	0.1	1.0	0.005
$\overline{C_i}$	0.081	0.0052	0.0068	0.21	0.53	0.004
S_i	0.116	0.124	0.68	2.1	0.53	0.8

Table 3. Transport scope statistics of pollutant manganese

Time/year	Horizontal distance/m	Vertical distance/m	the center point concentration of the rock pile/(mg/L)	Over standard multiples	Diffusion area/km^2
2	111	91	0.10	1	0.01
5	135	92	0.20	2	0.03
10	188	96	0.45	4.5	0.06



(a)

(b)

Fig. 4. (a) the migration and diffusion of manganese pollution at the end of the 5th year without anti seepage measures, (b) the migration and diffusion of manganese pollution at the end of 10th year without anti seepage measures

Using single-factor evaluation index method, the sample analysis results are achieved and they are shown in **Table 2**. $\overline{C_i}$ means the average measured concentration value of 8 test results. The value of Si reveals that all of the standard index of the six evaluation factor are less than one except manganese. And the maximum value of Si is 2.1 of manganese. That is the concentration of the other five evaluation factors is within the allowable value. However, the concentration of manganese is more than the allowable value. The factors of barium, silver, arsenic, manganese, fluoride and sulfide belong to heavy metals. They will have similar contaminant transport characteristics under the same conditions of the study area. Therefore, the factor with the largest Si value is selected to model the transport of leachate from waste rock piles in hilly areas to groundwater.

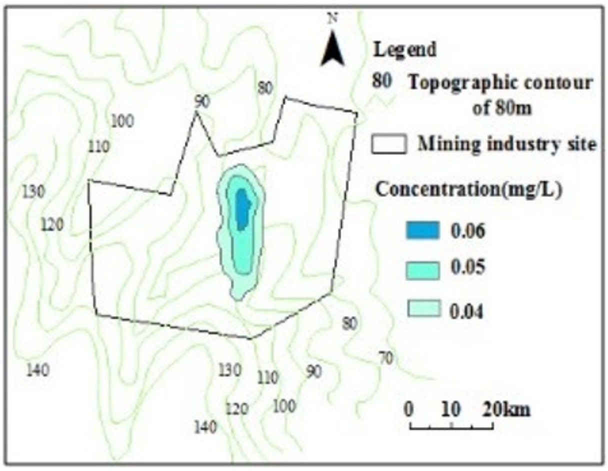
Impact Assessment of Leachate on Groundwater

Without anti-seepage measures

The adsorption of the unsaturated zone was not considered because of the limited thickness of the unsaturated zone. In the simulation of pollutant diffusion, we focused on convection and dispersion.

Water flow and water quality models were combined using Visual MODFLOW software and MT3D modules. The migration of pollutant manganese on a plane is shown in **Fig. 4** and **Table 3**. And **Fig. 4(a)** shows the migration at the end of the 5th year, while **Fig. 4(b)** shows that at the end of the 10th year. The concentration of contaminated plumes on the figure is 0.1 mg/L, as specified by the GB/T14848-2017 III water quality standard.

The analysis of the migration of pollutants for different periods revealed that manganese pollution diffuses mainly downstream during the simulation period because the location of the pollution source is relatively steep. When there were no anti-seepage measures in place on the waste rock piles, the concentration of manganese at the center point reached the maximum allowable value (0.1 mg/l) by the second year. In the tenth year, the maximum diffusion distance in the horizontal direction was 188 m and the center point concentration of manganese was 0.45 mg/l. This concentration is 4.5 times more than the standard, and the range is approximately 0.06 km^2. Under the influence of local hydrodynamic conditions, manganese



(c)

Fig. 4 (continued). (c) the migration and diffusion of manganese pollution at the end of the 10th year with anti seepage measures

will therefore affect groundwater in the study area. Because there is no aquifuge separating the unconsolidated and the fractured aquifers, there is a direct hydraulic connection between the aquifers, and pollutants begin to enter the fractured aquifers at the end of the second year. Without anti-seepage measures, the infiltration of leachate will cause the quality of the water in the unconsolidated and fractured aquifers to deteriorate as a result of the high concentration of manganese in the waste piles. Because the environmental pollution of the groundwater is serious, effective anti-seepage measures are needed.

With anti-seepage measures

To study the effect of leachate infiltration prevention measures on groundwater quality, we modeled anti-seepage measures for waste rock piles. An impervious layer with a vertical permeability coefficient of 1×10^{-7}cm/s was introduced. The resulting range of migration and diffusion of manganese pollution is shown in **Fig. 4(c)**. In this case, although manganese from the waste rock piles was still spreading downstream, the diffusion range was gradually reduced. The center point concentration of manganese was 0.08 mg/L at the end of the 10th year and was within the allowable range. The concentration was reduced by 82% at the end of the 10th year when compared with the no anti-seepage measures. Therefore, it can be concluded that taking anti-seepage measures can effectively control the impact of leachate water on groundwater quality, and ensure protection of the groundwater environment.

CONCLUSIONS

Mining can result in environmental problems for water resources and water quality, cause geological disasters, and threaten the safety of humans and animals. Therefore, the research on the impact of mining pollution on the environment has continued to receive attention and grow. However, the amount of research on environmental problems caused by leachate from waste rock piles is comparatively small compared with the research on acidic water produced in mining areas. The influence of leachate infiltration from iron ore waste rock piles on groundwater cannot be ignored, especially in hilly areas. To increase the knowledge on this topic, we investigated the Longqiao Mine, Anhui Province, China. Our study revealed that:

(1) The heavy metals in the leachate of the waste rock piles are mainly cerium, silver, arsenic, and manganese. Of these, the levels of manganese were the highest.

The thickness of the Quaternary deposits in the hilly areas is very limited, and bedrock is exposed in places. As a result, the rock is highly weathered, and the attenuation effect on pollutants is small. We used modeling to study the characteristics of the manganese pollution. In the absence of anti-seepage measures at the waste rock piles, the center point concentration of manganese had reached 0.45mg/l at the 10th year. This is 4.5 times more than the allowable limits. The area was approximately 0.06 km^2. As a result, the fractured aquifers in the study area would be contaminated. We then modeled anti-seepage measures by introducing an impervious layer with a vertical permeability coefficient of 1×10^{-7}cm/s. The maximum concentration at the center point was then reduced to 0.08 mg/l. This shows that anti-seepage measures can effectively prevent leachate infiltration and protect the groundwater environment.

ACKNOWLEDGEMENTS

This work was supported in part by the National Natural Science Foundation of China Grants 51509064 and 51309071. Data used to produce this paper are available on contact of the corresponding author

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