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Different Sorption Approaches and Leachate Fluxes Affecting on Mn^{2+} Transport through Lateritic Aquifer

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Abstract: Problem statement: Contamination of the underlying aquifer beneath a mining area is usually of great concern even when a prevention plan has been implemented. **Approach:** To assess the potential risk of heavy metal contamination, the simulation of heavy metal transport was carried out with different leachate fluxes and sorption parameters derived from equilibrium models with linear and Langmuir isotherms and chemical non-equilibrium two-site model. The HYDRUS-2D numerical model was applied to simulate the transport of Mn^{2+} under single- and multi-metal systems with two variable leachate fluxes (0.002 and 0.0026 m day^{-1}) through the lateritic aquifer, approximately 5 km down gradient of the tailing pond. The model assumed that the compacted clay layer of the Tailing Storage Facility (TSF) had been cracked and led to contamination of the shallow ground water. **Results:** The simulation showed that the time required to reach the Thailand drinking water standard at a specific location of Mn^{2+} for multi-metal system were faster than those for single metal systems, although different models were applied. The Mn^{2+} concentration fronts derived from simulation with chemical non-equilibrium two-site model came earlier than those of both equilibrium models under single and multi-metal systems. In addition, with a 30% increase in the leachate flux, from 0.002-0.0026 m/day, the time required to reach the drinking water standard at the nearest well, 1 km down-gradient from the source (well 1) decreased. It took about 57 and 106 years (a 17 and 19% decrease, respectively) for Mn^{2+} under multi- and single-metal systems, respectively. **Conclusion:** In conclusions, sorption parameters and leachate fluxes should be carefully determined and these predictive patterns used as a management tool for planning water well installations under field conditions.

Key words: Mining area, sorption parameters, modeling assessment, groundwater contamination, groundwater system, leachate fluxes, multi-metal systems, single metal systems, hydraulic properties, Tailing Storage Facility (TSF), Two-Site Model (TSM)

INTRODUCTION

Soil and groundwater contamination from heavy metals released from mining activities is a worldwide environmental problem. Heavy metals and other contaminants might release from mining sites, especially from waste dumps or tailing ponds (Alligui and Boutaleb, 2010; Lei *et al*., 2010). So, the monitoring wells should be established around the nearby areas in order to monitor the groundwater pollutions. However, such monitoring is quite expensive and time consuming. Various mathematical simulation models have been developed for the assessment of groundwater vulnerability to groundwater contamination, water resources management and design of monitoring well systems. HYDRUS-2D software codes (Fellner and Brunner, 2010) have been developed to simulate water movement and solute transport. The program can numerically solve the Richard's equation for saturated-unsaturated water flow and the advection-dispersion equation for solute and heat transport. In this software, the solute transport module also considers equilibrium and non-equilibrium advection-dispersion in the liquid

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phase, non-linear equilibrium physical and chemical sorption. Furthermore, the transport of heavy metals leached from mine tailings to groundwater systems needs to be realistically predicted in order to be able to assess the risk (Sabahi *et al*., 2009; Saghravani *et al*., 2010) and consequently, to develop and select the most appropriate strategies in monitoring and remediating the contaminated site. In order to apply numerical models, the hydraulic properties of the soils and solute transport parameters, such as sorption isotherms or retardation factors, must be accurately estimated. Most studies have been applied sorption properties of solute from batch experiments (Alshaebi *et al*., 2009), but there are some limits to describe solute behaviors in real world condition. Moreover, column experiments have been introduced as an alternative to investigate the displacement of dissolved solutes through soil and may provide information that are not available using equilibrium batch experiments such as physical and chemical non-equilibrium sorption (Akyol *et al*., 2011).

However, few simulations have been used chemical non-equilibrium sorption incorporate with unsaturated-saturated hydraulic properties to predict solute transport. Recently, from previous dissertation, the chemical non-equilibrium two-site model was shown to describe heavy metal transport in lateritic soil better than the equilibrium convection-dispersion models. Moreover, one of the most important hydraulic parameters is the leachate flux from a tailing pond where the clay liner has become permeable, such as after cracking. So, the objective of this study was to simulate the movement of heavy metals leached from mine tailings through shallow groundwater systems with variable leachate fluxes and different sorption parameters. To achieve such objective, the parameters, obtained from unsaturated-saturated soil tests and the chemical non-equilibrium two-site model were applied in the field-scale metal transport simulations here. Mn^{2+} was chosen as the representative heavy metal because it can leach from tailings at a higher level or rate than other metals. The HYDRUS-2D numerical model was used for the field-scale simulation in this study because it can evaluate the potential transport and contamination of heavy metals in groundwater.

MATERIAL AND METHODS

Study area: The manganese transport released from Tailing Storage Facility (TSF) in the Akara mining site (Fig. 1) was numerically simulated using HYDRUS-2D. This area located between 16°16'25'' and 16°17'41'' north latitude and 100°38'50'' and 100°40'15'' east longitude and about 280 km north of Bangkok in Phichit Province, central Thailand. It was selected for the study as it is an active gold mine with comparable geology and mining strategies to many other such mines. Thus, as well as being of national importance, any such study here is likely to be of broad applicability to many other mining systems in the tropics.

Methods: A Tailing Storage Facility (TSF) in the mine has been designed to safely store the mine tailings and is located at the southern portion of the mining site, covering an area of approximately 320,000 m^2. Groundwater levels typically conform to the surface topography. A north-south orientated hydraulic groundwater divide is located through Khao Mo and Khao Pong. Natural groundwater flows from the ore bodies in the C-H mining pit in a southwest direction to the adjacent areas (Fig. 1). The shallow groundwater wells in the nearby villages were mainly dug into the aquifer of the lateritic layer at depths between 1.5 and 7 meters. The soil profile in the surrounding area consists of a 20-40 cm top soil layer over the whole area, a 1.5 to 7 meter thick Lateritic layer and a thick clay layer of between 4.3-11 m depth formed at the bottom of the soil sequence.

The parameters of the unsaturated-saturated hydraulic were indirectly derived by input percentage of sand, silt, clay and bulk density into pedotransfer function in HYDRUS-2D. Such sorption parameters derived from equilibrium models with linear and Langmuir isotherms and chemical non-equilibrium two-site models, have been taken from lateritic soil column experiments showed in Table 1. Bulk Lateritic soils were collected from a depth of between 0 and 2 m below the ground surface from the Akara mine area (Fig. 1). The column experiments were conducted to investigate manganese fate and transport during February to September 2008. The simulations assumed that heavy metals were leached under acidic conditions (pH 5) from the tailing storage facility passing through the cracked liners and then through the lateritic soil reaching into the shallow groundwater system. Although this exact scenario may not happen in the real situation, the result can, however, be used for general monitoring and prevention plans regardless of the exact site and cause of the leaching.

HYDRUS-2D was used to simulate water flow in variably saturated soil. The Richard's equation (Hardie *et al*., 2011) can be written in the following simplified form (Eq. 1).

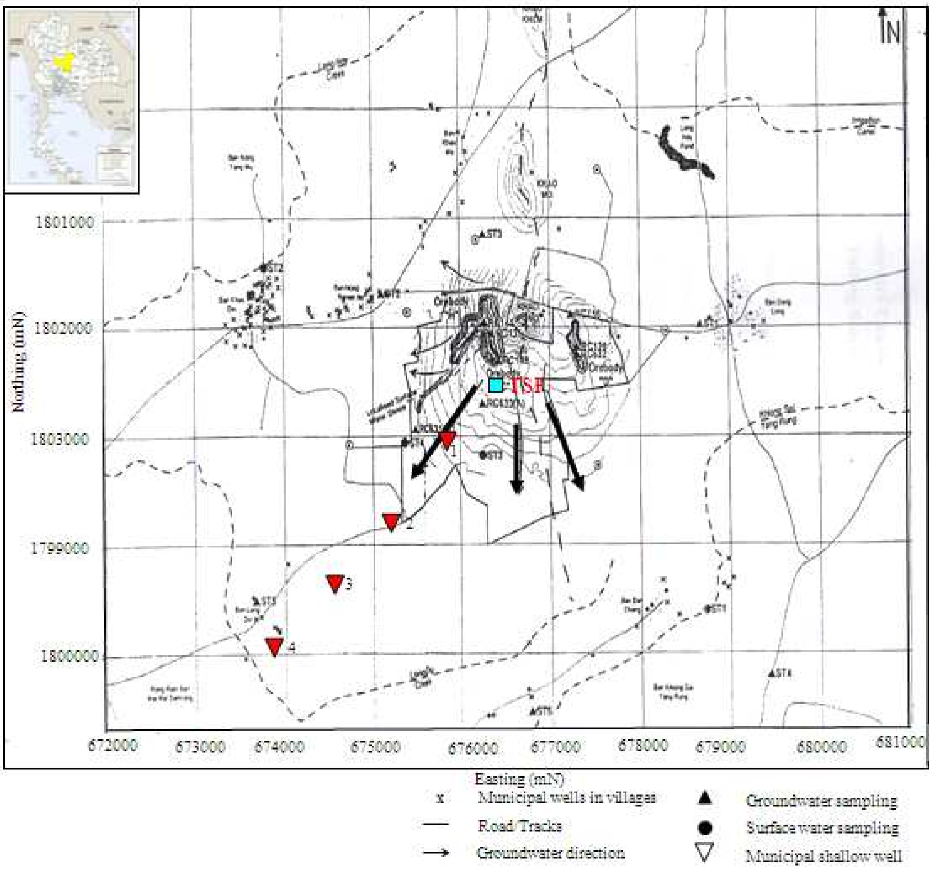

Fig. 1: Map showing the location and groundwater flow in the Akara gold mining site

Table 1: Input parameters for the HYDRUS-2D simulation of heavy metal transport

Parameter	Value
Longitudinal dispersivity#, m	500
Transverse dispersivity#, m	50
Bulk density# (g cm^{-3})	1.23
Residual water content#	0.104
Saturated water content#	0.523
Saturated hydraulic conductivity#, m day^{-1}	0.76
Water flux, mm day^{-1}	0.020 and 0.026 (30% increase)
Chemical nonequilibrium model parameters#,	0.12, 0.80, 0.33 and 0.02 for Mn^{2+} (single system)
Q_{max}, mM g^{-1} and b, L mM^{-1}, f (-) and α (hr^{-1})	0.04, 0.88, 0.48 and 0.05 for Mn^{2+} (multi-metal system)
Equilibrium convection-dispersion model parameters with	0.08 and 1.26 (single system)
1.Langmuir model#, Q_{max}, mM g^{-1} and b, L mM^{-1}	0.03 and 1.39 (multi-metal system)
2. Linear model#, K_d, L g^{-1}	11.53 (single system)
	5.05 (multi-metal system)
Initial concentration#, C_0, mg L^{-1}	40 for Mn^{2+}
Thailand drinking water standard, mg L^{-1}	0.3 for Mn^{2+}

#Derived from previous study (Chotpantarat *et al*., 2011)

Water flow equation:

$$\frac{\partial\theta}{\partial t} = \frac{\partial}{\partial z}\left[k(\theta)\left(\frac{\partial h}{\partial z}+1\right)\right] \qquad (1)$$

Where: θ(h) is the volumetric water content (L^3L^{-3}), h is the suction pressure head (L), k(θ) is the unsaturated hydraulic conductivity (LT^{-1}), t is the time (T), z is the vertical distance (L)

Soil hydraulic properties: The Soil Water Characteristics Curve (SWCC) is the relationship between water content, $\theta(h)$ and suction pressure head, h. The formulation often used to describe unsaturated soil is van Genuchten (VG) formulation as presented in Eqs. 2-3. The unsaturated hydraulic conductivity function, K(h) are given by the the Mualem-van Genuchen model (Palla *et al*., 2009) as show below in Eqs. 4-5:

van Genuchten equation:

$$S_e = \frac{\theta(h)-\theta_r}{\theta_s-\theta_r} = \left[\frac{1}{1+(\alpha h_m)^n}\right]^m \quad h<0 \qquad (2)$$

$$=1 \quad h \geq 0 \qquad (3)$$

$$\begin{aligned} k(\theta) &= K_s k_r(\theta) \quad h<0 \\ &= K_s \quad h \geq 0 \end{aligned} \qquad (4)$$

$$k_r(\theta) = (S_e)^k \left[1-\left(1-(S_e)^{1/m}\right)^m\right]^2 \qquad (5)$$

Where: $\theta(h)$ is the soil water content at the suction head (-), h_m is the soil suction head (L), θ_r is the residual water content (L^3L^{-3}), θ_s is the saturated water content (L^3L^{-3}), K_s is the saturated hydraulic conductivity (LT^{-1}), k_r is the relative hydraulic conductivity (LT^{-1}), S_e is the relative water saturation (-), n is constant of soil k is the pore-connectivity parameter (-) (k = 0.5 in this case), m, n, α are fitting parameters of soil water retention curve; m = 1-(1/n)

The parameters of the unsaturated-saturated hydraulic were indirectly derived by input percentage of sand, silt, clay and bulk density into pedotransfer function in HYDRUS-2D

Solute transport equation: Chemical non-equilibrium model or the two-site model with Langmuir sorption model as presented by Eq. 6:

$$\begin{aligned} &\left(1+\frac{f\rho}{\theta}\left[\frac{Q_{max}b}{(1+bC)^2}\right]\right)\frac{\partial c}{\partial t} = D\frac{\partial^2 c}{\partial x^2} \\ &-v\frac{\partial c}{\partial x} - \frac{\alpha\rho_b}{\theta}\left[(1-f)\frac{Q_{max}bC}{1+bC} - s_2\right] \end{aligned} \qquad (6)$$

Where: C is the solute concentration in solution (ML^{-1}), S is the sorbed solute concentration on soil (MM^{-1}), T is time (T), V is the average linear groundwater velocity (LT^{-1}), P_b is the soil bulk density (ML^{-1}), θ is the volumetric moisture content or porosity for saturated media, D is the hydrodynamic dispersion coefficient (L^2T^{-1}), f is the fraction of equilibrium sites (-), α is a first-order kinetic rate coefficient (T^{-1}), S_2 is the solid phase concentration at site 2.

RESULTS

In order to forecast of the long term impacts on groundwater quality, various simulations of flow and heavy metals transport have been simulated under different environmental conditions as shown below.

Effect of different sorption models under single and multiple metal systems on groundwater quality: To investigate the importance of the proper sorption parameters obtained from column studies (Chotpantarat *et al*., 2011) under different environmental conditions, the sorption parameters (Table 1) derived from different environmental conditions were applied to the field scenario where the compacted clay liner was assumed to be cracked.

Figure 2 shows the schematic simulation of heavy metal transport from the TSF. For simplicity, it was assumed that the lateritic layer has a uniform thickness of 7 meters and the hydraulic gradient is 0.001 m m^{-1}. It was assumed that the maximum concentrations of heavy metals leached from the column desorption experiments were transported continuously to the lateritic soil. Under these conditions, the input parameters used are listed in Table 1. Moreover, the background concentration of Mn^{2+} in the lateritic aquifer layer was assumed to be zero. With the assumed initial concentration C_0 and the sorption parameters obtained from the equilibrium models with linear and Langmuir isotherms, as well as chemical non-equilibrium two-site model (Table 1), the predicted (simulated) Mn^{2+} metal concentration in the four municipal shallow wells, located at 1-4 km from the source, respectively, were evaluated using the HYDRUS-2D computer simulation software (Fig. 3).

The simulation also shows the influence of the simultaneous presence of other metals on the transport of Mn^{2+} in lateritic soil (Fig. 3), depicted as the Mn^{2+} concentration detected in each well (y-axis) versus the time to reach that Mn^{2+} concentration (x-axis). The time needed to reach the Thailand drinking water standard of Mn^{2+} at well no. 1 (the nearest well from the source) under multi- and single-metal systems were about 70 and 130 years, 133 and 322 years and 70 and 150 years for chemical nonequilibrium two-site model, equilibrium models with Langmuir and linear

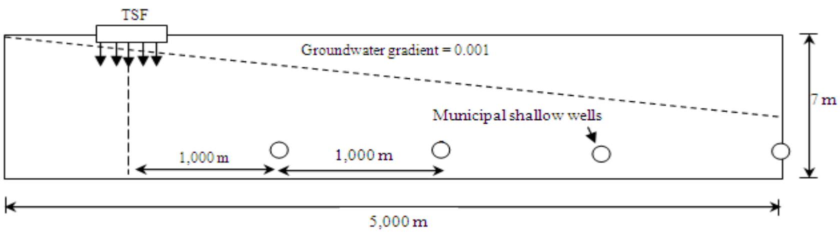

Fig. 2: Schematic of the heavy metal transport leached from the TSF through the lateritic aquifer and the positions of the assumed ground wells nos. 1- 4 with a 1 km distance between each well

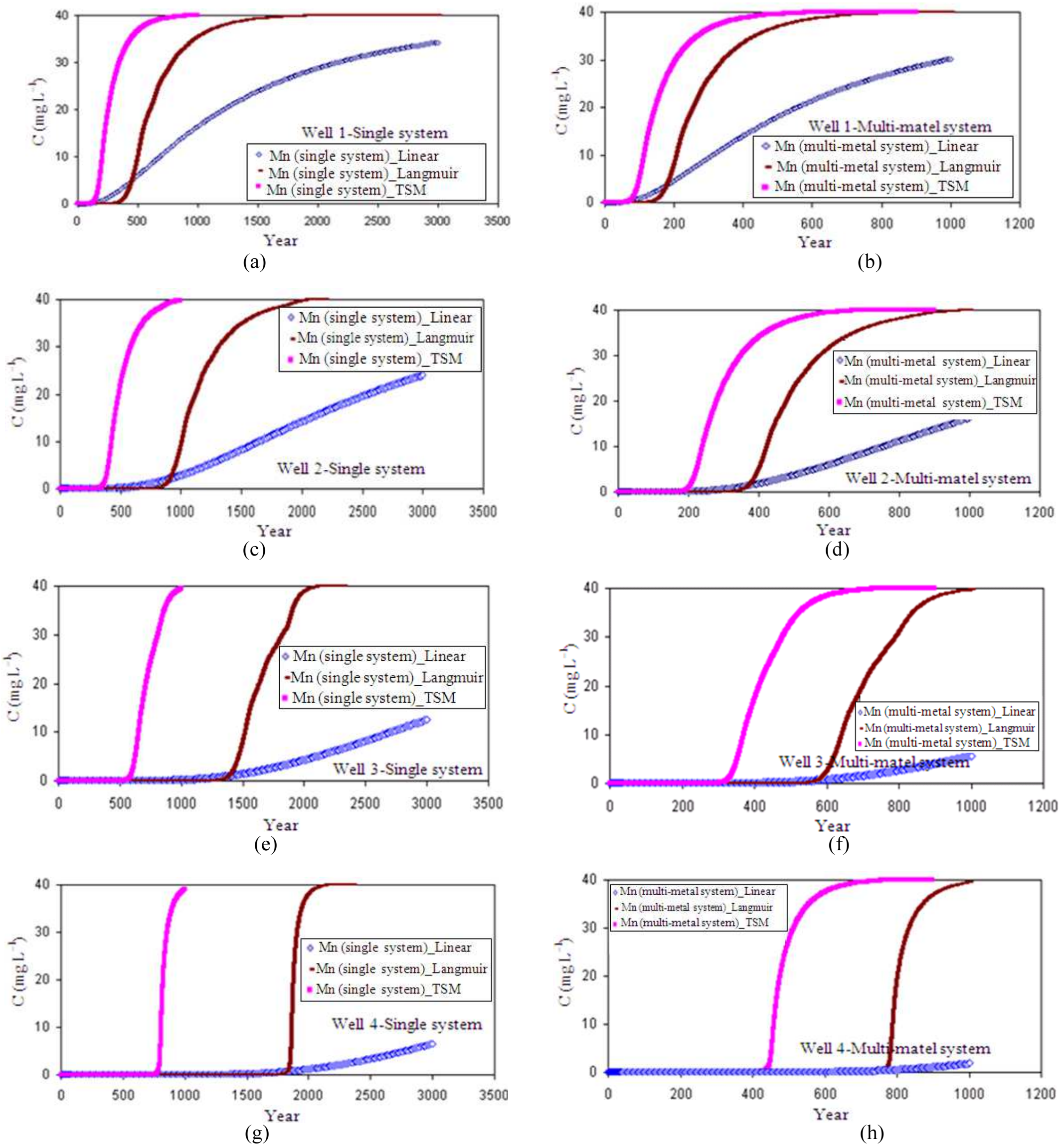

Fig. 3: Concentrations of Mn^{2+} in the lateritic aquifer simulating under equilibrium convection-dispersion models with linear and Langmuir isotherm and the chemical nonequilibrium two-site model for single metal and multi-metal systems in four wells at increasing 1 km distances from the contaminant source (Fig. 1)

Table 2: Required time, at well nos. 1-4 under single-,and multiple-metal systems, for Mn^{2+} concentrations to exceed the drinking water standards of Thailand with equilibrium convection-dispersion model with linear and Langmuir isotherm and chemical nonequilibrium Two-Site Model (TSM)

	Time to exceed the Thailand drinking water standard for Mn^{2+} (years)					
	Single metal systems			Multiple metal systems		
Well	Linear	Langmuir	TSM	Linear	Langmuir	TSM
1	150	322	130	70	133	70
2	540	816	340	250	340	190
3	1070	1325	560	490	557	310
4	1565	1790	770	715	757	430

Table 3: Required time, at well nos. 1-4 under single-,and multiple-metal systems, for Mn^{2+} concentrations to exceed the drinking water standards of Thailand at a basal (f = 0.002 m/day) or 30% elevated (f = 0.0026 m day^{-1}) leachate rate

	Time to exceed the Thailand drinking water standard for Mn^{2+} (years)			
	Single metal system		Multiple metal system	
Well	Basal	30% Elevated	Basal	30% Elevated
1	130	106 (-19%)	70	58 (-17%)
2	340	271 (-20%)	190	149 (-22%)
3	560	444 (-21%)	310	248 (-20%)
4	770	602 (-22%)	430	340 (-21%)

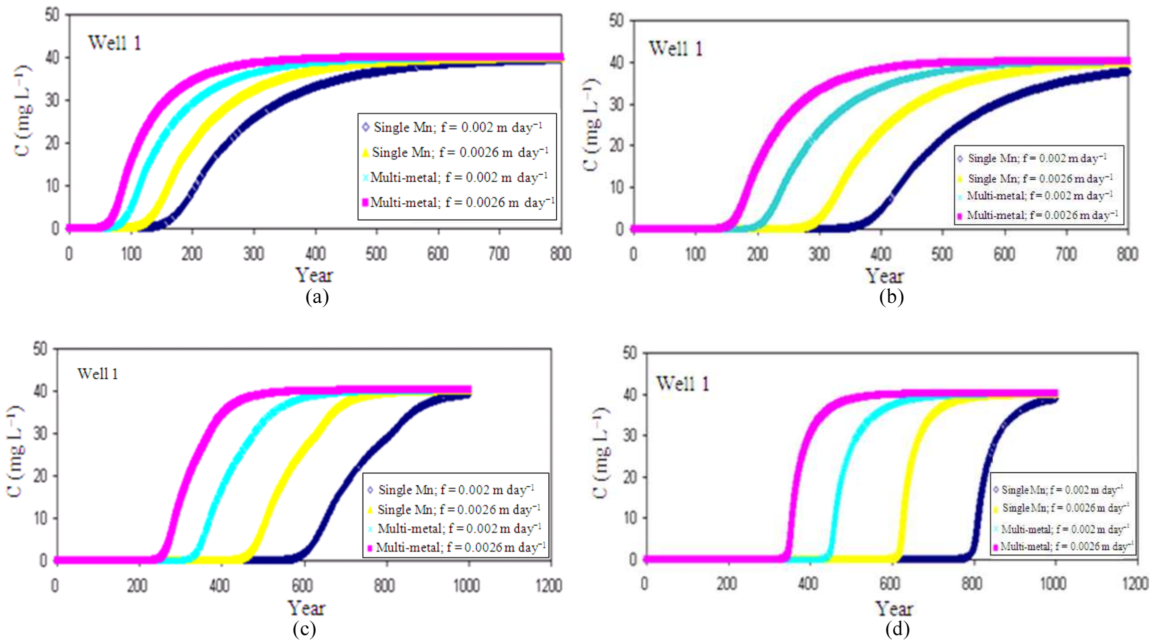

Fig. 4: Concentrations of Mn^{2+} in the lateritic aquifer under single- and multi-metal systems under variable flux in four wells which represent increasing 1km distances from the contaminant source (Fig. 1), with a 30% elevated water flux rate of 0.026 mm day^{-1}

isotherms, respectively (Table 2). Regarding the effect of distance from the source (TSF) and the time to reach the standard, the time for well nos. 2-4 increased respectively. This means that the time to reach required concentrations were significant difference for applying the different approaches to explain heavy metal transport through the aquifer.

Effect of variable leachate fluxes on groundwater quality: During the mining processing, an enormous amount of mine tailings are typically generated and dumped into the TSF, potentially causing the clay liner to crack. As a consequence, when it rains, the water flux infiltrates through the mine tailings with a relatively high potential increase in the Mn^{2+} concentration in the mine tailings leachate. In addition, during the post-mine closure, the increase in the head difference between the leachate in the TSF and the water table might be one of the major factors that affect the increasing water flux through the clay liner. With parameters derived from chemical nonequilibrium two-site model, the computer simulation results of variable fluxes of leachate with a 30% increase from the base case (0.002 m day^{-1}) infiltrating through lateritic aquifer in case of cracking of compacted clay liner are summarized in Fig. 4. Increasing the water flux 30% from the base case decreased the time required to reach the Thailand drinking water standard of Mn^{2+} at well no. 1 (nearest well from the source) by about 58 (17% decrease) and 106 years (19% decrease) under multi- and single-metal systems, respectively (Table 3). Likewise, for well nos. 2-4, the time required to reach the Thailand drinking water standard of Mn^{2+} decreased about 20% compared with those of the base case in both single-and multi-metal systems.

DISCUSSION

According to previous results, the transport of Mn^{2+} simulated with sorption parameters derived from chemical nonequilibrium model move faster than those derived from Langmuir model, which might cause from the binding coefficients, b (1.26 and 1.39 for single and multiple metal systems, respectively), derived from Langmuir model were higher than those from chemical nonequilibrium model (0.80 and 0.88 for single and multiple metal systems, respectively). Moreover, Mn^{2+} under multi-metal systems seems to move faster than those under single metal systems and would exceed the standard in the first well (1 km from source) in approximately 70 years. In the multi-metal system, the time to reach the standard for Mn^{2+} at specific locations appeared to be reduced by about 1.8-fold compared to that of single systems, respectively. This potential impact to the shallow groundwater should be of concern in monitoring/prevention strategies. As mentioned, the transport of Mn^{2+} in a multi-metal system was faster than that observed in the single metal systems, which may well have resulted from the competition of heavy metals for limited sorption sites and nonequilibrium processes (Oh *et al*., 2009; Fernandez-Calvino *et al*., 2010). This notion and data are in good agreement with the values of maximum sorption capacity (Table 1) and retardation factor of Mn^{2+} onto lateritic soil. Retardation factor (R_f = 6.95) of Mn^{2+} in a multi-metal system was lower than those in the single- (R_f = 19.22) and binary- (R_f = 10.40) metal systems derived from column data previously (Chotpantarat *et al*., 2011). Du and Hayashi (2006) found results in line with our study that the potential impacts of the assumed landfill on the groundwater environment are significantly dependent on the partition coefficients of the Ariake clay liner, K_p, which were derived from different sorption of soil: metal solution ratios.

Du and Hayashi (2006) found results along with our simulations when increasing water level in tailing pond, the migration of Cd^{2+} through Ariake clay liner into aquifer underneath increased. In the multi-metal system, the times needed to reach the standard for Mn^{2+} at specific locations under variable fluxes appear to be reduced by about 1.8-fold compared with those in the single-metal system, which is a very similar degree of difference between the single- and multiple-metal systems as that seen with the basal leachate level, suggesting changes in the leachate rate do not display differential effects between the single- and multiple-metal systems. The study of Ngoc *et al*. (2009) give a agreement with Mn^{2+} movement in subsurface environment that the increasing of water layer in paddy field from 1-30 cm can enhance the leaching rate of heavy metal up to 36%.

As mentioned above, the selection of the proper sorption parameters, such as those derived from column studies with the simultaneous presence of other heavy metals, is very crucial. Moreover, to provide a sufficient level of accuracy about the infiltrated water from tailing ponds, the monitoring wells underneath the compacted clay liner of the tailing ponds should be monitored even after the mine closure. The construction design of the compacted clay liner is of great concern due to the high loading of mine tailings into and the long-term aging of the clay liner. When the clay liner cracks, it will probably lead to a high level of metal leachates through the subsurface environment. Moreover, the head difference in the TSF after mine closure might increase the risk that leachates migrate through the underlying aquifer. Therefore, predicting the potential impact with variable water fluxes and proper sorption parameters will provide a visible description of the heavy metal transport and further act as a tool for the design and installation of monitoring wells in field conditions. However, it should be noted that these predicted simulations were initiated for rather simple cases. The simulation would be more accurate in its prediction when information for the actual field conditions was used for the model. For example, water levels or metal concentrations in the monitoring wells should be taken into account. Moreover, the other processes, such as precipitation and dissolution of Fe-oxides and other chemical and biological processes may affect the heavy metal transports. Further study should be carried out in some detail for formulating a better predictive ability of the heavy metal transport under specific field conditions.

CONCLUSION

The application of the HYDRUS-2D model provided visible descriptions of heavy metal movement in the lateritic aquifer under different environmental conditions. The sorption parameters from column studies and variable fluxes derived can be used as important parameters to predict heavy metal transport in field conditions. However, the actual sorption parameters obtained from different models and different environmental conditions are the critical factors for accurate and useable predictions.

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