EFFECTIVENESS OF PERMEABLE REACTIVE BARRIER (PRB) ON HEAVY METAL TRAP IN AQUIFER AT SOLID WASTE DUMPSITE: A SIMULATION STUDY

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ABSTRACT: Open dumping of solid waste causes a serious environmental impact on groundwater due to contamination of landfill leachate rich in heavy metals. Identification of contaminant flow and implementation of remediation technologies, such as permeable reactive barriers (PRB) are highly demanded and applicable. In this study, a groundwater model, Geo-Environmental Risk Assessment System (GERAS) has been used to simulate two-dimensional heavy metal transport in an aquifer at solid waste dumpsite and to estimate the effectiveness of virtual PRB on trap heavy metals. First, two cases were examined: 1) Open dumping of waste located above the aquifer and 2) Buried dumping of waste into the aquifer. Concentration changes of heavy metals (Cd and Pb) inside the aquifer beneath the waste unit, inside PRB, upstream and downstream points to PRB were examined by changing the hydraulic gradient, distribution coefficient, and pollution load. Results showed the numerical simulations well captured the wash-out process of heavy metals from the pollutant source. The time required for full wash-out was highly dependent on the hydraulic gradient, distribution coefficient, pollution load and the way of waste dumping. In Case 2, a sudden pollution plume was observed with high heavy metal concentrations, by creating greater risk at the downstream. Next, a virtual PRB was set in downstream of the aquifer by installing a section with a high heavy metal adsorption capacity (based on previous studies). Results showed that in both cases, the virtual PRB well trapped the target metals and reduced the contamination level less than the effluent water quality standards.

Keywords: Solid waste dumpsite, Groundwater, Heavy metal, Permeable reactive barrier, Numerical simulation

1. INTRODUCTION

Open dumping is a common method used for the final disposal of municipal solid waste in most developing countries [1]. Landfill leachate generates at the process of the waste degradation and may contaminate with several toxic materials, including heavy metals. According to the [2],[3], the heavy metals such as Pb, Cd, Cu, Zn, Ni were detected in the landfill leachate and in some cases the heavy metal concentrations were exceeded their maximum effluent water quality standards [3]. The management practices at the open solid waste dumpsites are primitive in level, no landfill liners, leachate collection systems, no final covers, consequently the leachate is directly open into the original topsoil. The permeation of leachate causes serious soil and groundwater pollution in surroundings [1]. The contaminated water plume may spread through the aquifer along with the hydraulic gradient. Thus, the public and wildlife at the down streams become soft targets of this contaminated groundwater [1]. Once the heavy metals reach to the groundwater bodies, the bioaccumulation may occur and it leads to several health issues such as kidney diseases, cancers, mental retardation in children, gastrointestinal disorders etc.

In this regard, identification, and mitigation of groundwater contamination at open solid waste, dumpsites have been highly demanded. Recently, Permeable Reactive Barrier (PRB) system, became an option to treat contaminated groundwater at open solid waste dumpsites, based on their easiness of the installation and management. The PRBs are one of the in-situ water treatment technology, which can immobilize the targeted contaminants in the polluted water plume. PRB system does not interrupt the groundwater flow in the treatment process, thus highly suitable to use as in-situ pollutant immobilization technique [4]. In this study, a numerical simulation software, Geo-Environmental Risk Assessment System (GERAS) [5], has been used to simulate a two-dimensional groundwater
flow coupled with heavy metal transport. In the simulations, the effectiveness of PRB on the heavy metals (Cd and Pb) trap in an aquifer at solid waste dumpsite has been examined.

2. MATERIALS AND METHODS

2.1 Selection of Modeling Area

A conceptual contaminated site was classified based on data collected from an open solid waste dumpsite located in the Central province of Sri Lanka. The dumpsite has been used for 7 years of waste dumping [2]. Fig. 1 illustrates the arrangement and setting of the conceptual open solid waste dumpsite model. The conceptual open solid waste dumpsite (waste unit) was fixed as 100m in length and 40m in width and the area of the aquifer was set to be 400m x 100m in x and y-direction. The thicknesses of the aquifer and waste unit were set to be 2m. A 4m width and 45m length virtual PRB was installed in the downstream (20m away from the end point of the dumped waste in the x-direction). In the simulations, two cases were studied: Case 1: An open dump of waste (pollutant source) located above the aquifer and direct permeation of rainwater into the aquifer through the waste layer (Fig. 1a). Case 2: A buried waste dump inside the aquifer, no effect of rainfall, groundwater flow affects the washing out of contaminants (Fig. 1b). Spatial and temporal variability of contaminants and the effectiveness of the PRB were evaluated in both cases by analyzing the changes in heavy metal concentrations at upstream, downstream and inside the PRB.

2.2 Governing Equations

GERAS simulation model is used to estimate contaminant concentration (heavy metals) in the groundwater as a function of time and space. The model simulation is considered to be two dimensional in the horizontal plane. The governing equations have applied to three main zones of the contaminated sites.

2.2.1 Zone 1: Aquifer

The model follows the flow equations as shown in Eqs. (1) and (2) assuming water is an incompressible fluid, the fluid pressure created by groundwater is low, and the aquifer is homogeneous, and isotropic porous media [6].

\[ \frac{\partial^2 \Phi}{\partial x^2} + \frac{\partial^2 \Phi}{\partial y^2} = 0 \]  

\[ v_x = -\frac{k_x}{n_a} \frac{\partial \Phi}{\partial x} \quad \text{and} \quad v_y = -\frac{k_y}{n_a} \frac{\partial \Phi}{\partial y} \]  

where \( \Phi [L] \) is the pressure head, \( v_x \) and \( v_y \) [LT\(^{-1}\)] are the groundwater velocities in x and y directions, \( k_x \) and \( k_y \) [LT\(^{-1}\)] are the hydraulic conductivities in x and y directions, \( n_a [-] \) is the porosity of aquifer.

A solute transport equation used in the simulations was as follows [7]:

\[ \frac{\partial C}{\partial t} + \nabla \cdot (D \nabla C) = S + \nabla \cdot (v C) \]
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\[
\frac{\partial C}{\partial t} = \frac{\partial}{\partial x} \left[ D_x \frac{\partial C}{\partial x} \right] + \frac{\partial}{\partial y} \left[ D_y \frac{\partial C}{\partial y} \right] - \left[ V_x \frac{\partial C}{\partial x} - V_y \frac{\partial C}{\partial y} \right] - \frac{1-n_a}{n_a} \rho_s \frac{\partial S}{\partial t}
\]

where \(C\) [ML\(^{-3}\)] is the solute concentration, \(t\) is the time, \(D_x\) and \(D_y\) [L\(^2\)T\(^{-1}\)] is the diffusion coefficient in \(x\) and \(y\) directions, \(S\) [MM\(^{-1}\)] is the saturated adsorption of the contaminant into soil particles. The \(D_x\) and \(D_y\) are as follows:

\[
D_x = \varepsilon D_M + \tau_L V_x
\]

\[
D_y = \varepsilon D_M + \tau_T V_y
\]

where \(D_M\) [L\(^2\)T\(^{-1}\)] is the molecular diffusion coefficient of metal (Cd and Pb) in free water and \(\varepsilon\) is the tortuosity (used Millington and Quirk model [8] in this study). The longitudinal and transverse dispersivities, \(\tau_L\) and \(\tau_T\) [L], were determined with respect to the scale of the aquifer in the simulation and \(\tau_T\) was considered as 0.1 times of \(\tau_L\) [9]. In the aquifer, linear adsorption model was used for the adsorption of heavy metals into solid phase:

\[
S_s = k_{d-w} C_w
\]

\[
S_s = k_{d-w} C_w
\]

where \(k_{d-w}\) [L\(^3\)M\(^{-1}\)] is the distribution coefficient of target heavy metal in the aquifer.

2.2.2 Zone 2: Waste unit

Few assumptions were made when applying the governing equations to waste unit: 1) heavy metal component in solid waste stocked within the waste unit, 2) dissolution/ desorption of heavy metal due to rainfall permeation and groundwater flow in Case 1, and only by groundwater flow in Case 2. Saturated adsorption of heavy metals into waste particles: \(S_{ws}\) [MM\(^{-1}\)] were determined following linear adsorption model using the distribution coefficient of target heavy metal in the waste unit: \(k_{d-ws}\) [L\(^3\)M\(^{-1}\)].

\[
S_{ws} = k_{d-ws} C_{ws}
\]

The concentration of the heavy metal component in porous media of waste (\(C_{ws}\)) [ML\(^{-3}\)] is considered to be equal to the heavy metal concentration of adjacent soil porous.

Inflow flux of heavy metal in Case 1 follows the Eq. (8).

\[
q_{hm} = R q_t C_w
\]

where \(q_{hm}\) [MT\(^{-1}\)] is the inflow flux, \(R\) is the permeation ratio, \(q_t\) [L\(^3\)T\(^{-1}\)] is the rainfall intensity, and \(C_w\) [ML\(^{-3}\)] is the equilibrium concentration of heavy metal in the water phase. The inflow of heavy metal in Case 2 is controlled by Eq. (7) [10].

2.2.3 Zone 3: Permeable Reactive Barrier (PRB)

Langmuir adsorption model was applied to characterize the heavy metal adsorption at PRB in the simulations:

\[
\frac{C_{ws}}{S} = \frac{1}{b Q_m} + \frac{C_w}{Q_m}
\]

where \(S\) is the adsorbent amount of heavy metal into the reactive material, \(b\) [L\(^3\)M\(^{-1}\)] is the Langmuir constant and \(Q_m\) [MM\(^{-1}\)] is the maximum adsorption capacity [11].

2.3 Setting of Input Parameters

Table 1 summarizes parameters used in the simulations and fig. 2 illustrates the general framework of the model simulation.

2.3.1 Aquifer

The hydraulic gradient was calculated based on groundwater level measured at the studied dumpsite [12]. The model uses the single value for the calculation and it is assumed that the hydraulic gradient is constant throughout the simulation period. The distribution coefficient of heavy metal in the water phase \(k_{d-w}\) was determined considering the soil type of studied area. The soil type of the studied dumpsite is Reddish brown latosolic and the distribution coefficient calculated for the same soil type was used [13].

2.3.2 Waste unit

The initial concentration of heavy metal was set by considering monitored metal concentrations at studied dumpsite [2]. Referred values [14] were used for the distribution coefficient of heavy metal in waste. The weight of the dumped waste was considered as 1.07E+07 kg, according to the in-situ dry density of waste (= 0.6 kg/m\(^3\)) and specific gravity (= 2.3) [15].

2.3.3 Permeable reactive barrier (PRB)

The PRB material was selected based on the previous study [16]. In the simulations, the hydraulic conductivity of PRB is assumed to be equal to the hydraulic conductivity of aquifer.
Fig. 2. The general framework of the model simulation.

Table 1 Input parameters used for model simulation

| Zone               | Description                                      | Symbol | Unit    | Value          | Reference |
|--------------------|--------------------------------------------------|--------|---------|----------------|-----------|
| Aquifer            | Porosity                                        | $n_a$  |         | 0.4            | [15]      |
|                    | Hydraulic conductivity                           | $k_x, k_y$ | m/s    | 1E-03          | [15]      |
|                    | Hydraulic gradient                               | $i$    | -       | 0.06 (0.006, 0.12)* | [12]      |
|                    | Molecular diffusion coefficient                  | $D_M$  | m$^2$/s | Cd: 6.0E-10    | [15]      |
|                    |                                                   |        |         | Pb: 7.9E-10    |           |
|                    | Dispersivity                                     | $\tau_x$ | m      | Cd: 10         | [15]      |
|                    |                                                   |        |         | Pb: 10         |           |
|                    | Distribution coefficient                         | $k_{d-w}$ | m$^3$/kg | Cd:1.3E-02     | [14]      |
|                    |                                                   |        |         | Pb:1.4E-01     |           |
| Waste unit         | The initial concentration of the heavy metal component | $S_{ws}$ | mg/kg  | Cd:3.63E-02    | [2]       |
|                    | Distribution coefficient                         | $k_{d-ws}$ | m$^3$/kg | Cd:1.3E-03     | [17]      |
|                    |                                                   |        |         | Pb:2.7E-03     |           |
| PRB                | The maximum adsorption capacity of the heavy metal | $Q_m$  | mg/kg   | Cd:3.02E+03    | [16]      |
|                    | Langmuir isotherm constant                        | $b$    | m$^3$/mg| Pb:9.94E+02    | [16]      |
|                    |                                                   |        |         | Pb:4.54E-05    |           |
| Meteorological parameters | Rainfall intensity | $q_r$  | mm/yr   | 3.53 E+03      | [2]       |
|                    | Permeation ratio                                 | $R$    | -       | 1              | [18]      |

*Used for the sensitivity analysis

Rainfall data were collected from a rain-gauge station nearby studied dumpsite [2].

2.4 Sensitivity Analysis

Sensitivity analysis was conducted to analyze the effect of various input parameters. The selected parameters are some of the primitive characters which greatly depends on the site location, hydrology, and management practices etc. The values chosen were 1, 0.1, 2 times of the actual input parameter for hydraulic gradient (0.06, 0.006, 0.12). Similarly, the scale of the dumpsite (waste unit) was changed from 100 m x 40 m to 40 m x 40 m in order to evaluate the effect of pollutant loads. The model simulations were also conducted by considering the distribution coefficient of the aquifer $k_{d-w}$. The $k_{d-w}$ for each metal greatly varies with the soil type of the aquifer. According to the Alumaa et al., (2001) [19] the $k_{d-w}$ depends on the different characteristics of the soil such as organic matter content (OMC), clay content and soil mineralogy etc. As an example, $k_{d-w}$ of Pb positively correlates with the OMC, whereas for the Cd a great positive correlation was observed with the soil mineralogy. The $k_{d-w}$ may greatly differ from site to site. Thus the effect of the $k_{d-w}$ was also examined in the sensitivity analysis. The values chosen for the $k_{d-w}$ were 1, 0.1, 10 times of the $k_{d-w}$ of the study area. (Cd: 1.3E-02, 1.3E-03, 1.3E-01 and Pb: 1.4E-01, 1.4E-02, 1.4 m$^3$/kg). The
time taken for peak heavy metal concentration in the monitoring points was compared.

3. RESULTS AND DISCUSSION

3.1 Model Outputs of Sensitivity Analysis

Table 2 exemplifies the model outputs of sensitivity analysis for the pollution load and hydraulic gradient. In both cases output values (peak time and concentration inside the PRB) were highly sensitive to the hydraulic gradient for both heavy metals. For example, the time for peak concentration was mainly controlled by $i$: the peak time for $i = 0.006$ become 4-6 times higher than that for $i = 0.06$ except for the Pb with low pollution load. On the other hand, the effect of pollution load was significant only for the outputs from Case 2: the peak concentration for the low pollution load became ~20% of the peak concentration for the high pollution load.

Figure 3 shows the effect of distribution coefficient on the washing out of the heavy metals in the aquifer. The effect of $k_{d-w}$ was significant for the heavy metal concentration and the rate of the washing out process. The peak heavy metal concentration was higher and the less time was taken to reach breakthrough with the decreasing of $k_{d-w}$ According to Alumaa et al., (2001) [19] the soil rich with organic matter may have high $k_{d-w}$ for Pb which encourage the slow releasing, whereas the soil rich with Ca may enhance the Cd immobilization (high $k_{d-w}$) and slow washing out process.

As elaborated in Table 2, the washing out of contaminants (reduction in metal concentration) was much faster in Case 2 than Case 1 for both Cd and Pb. This attributes to the direct exposure of waste unit into the groundwater flow in Case 2. On the other hand, the washing out of heavy metals is controlled by the rainfall precipitation in Case 1. These results suggest that rapid and higher pollution migration easily occurs in the waste dump site with a buried waste.

3.2 Evaluation of the Effectiveness of PRB

Figure 4 illustrates the temporal variation of the heavy metal concentration in different places of the aquifer. In most cases, the PRB trapped well heavy metals in the aquifer and reduced the peak concentration at the outflow from PRB. For both Cd and Pb, the heavy metal concentrations observed at the outflow from PRB were less than those of the effluent water quality standards in Sri Lanka which is <0.1 mg/L for Cd and Pb (CEA 2005) [20]. Furthermore, the heavy metals trapped by the PRB in Case 2 were greater than those in Case 1 due to the higher concentrations of heavy metals in the inflow to PRB.

3.3 Pollution Migration Process in the Aquifer

Figure 5 shows an example of pollutant migration process in the aquifer. The simulation was carried out to observe the distribution of Cd concentration in the aquifer, with respect to the Case 2. A rapid washing out process was observed in the upstream to the PRB and the pollutant plume reached the PRB within six months. Due to the adsorption at the PRB, a drastic reduction of Cd concentration was observed in the outflow from the PRB. However, even after 10 years, contaminants can be found in the aquifer which is 300 m away from the buried waste unit.

Table 2 Model outputs of sensitivity analysis for the pollution load and hydraulic gradient

|                 | $i$   | Cd (Time (Year), Concentration (mg/L)) | Pb (Time (Year), Concentration (mg/L)) |
|----------------|------|--------------------------------------|---------------------------------------|
| High pollution load | Case 01 | 0.060 (5.50, 2.30E-03) | 0.120 (3.70, 2.18E-03) | 0.006 (24.0, 2.35E-03) |
|                 | Case 02 | 0.060 (0.90, 1.16E-02) | 0.120 (0.50, 1.22E-02) | 0.006 (5.30, 1.14E-02) |
| Low pollution load | Case 01 | 0.060 (7.50, 1.44E-03) | 0.120 (4.60, 1.30E-03) | 0.006 (41.9, 1.60E-03) |
|                 | Case 02 | 0.060 (2.80, 2.54E-03) | 0.120 (1.70, 3.14E-03) | 0.006 (17.7, 2.44E-03) |

*N.B.: No breakthrough.
Case 1: High pollutant load                       Case 1: Low pollutant load                       Case 2: High pollutant load                        Case 2: Low pollutant load

Fig. 4 Temporal variation of the heavy metal concentration at the different locations of the aquifer for Cd [(a), (b), (c), (d)] and for Pb [(e), (f), (g), (h)]

- Inside the waste unit
- Inflow to PRB
- Inside PRB
- Outflow from PRB
- Effluent water quality standards

Fig. 4 Temporal variation of the heavy metal concentration at the different locations of the aquifer for Cd [(a), (b), (c), (d)] and for Pb [(e), (f), (g), (h)]
4. CONCLUSIONS

In this study, numerical simulations were used to understand the washing-out process of heavy metals from the pollutant source (waste unit) and to evaluate the effectiveness of a virtual PRB set in the downstream of the contaminant plume. The model well captured the washing out process at the aquifer and the time period needed for the full washing-out was highly dependent on the range of hydraulic gradient, distribution coefficient of the aquifer, pollution load and the way of waste dumping (Case 1 or Case 2). The PRB well trapped the targeted heavy metals and reduced the contamination level less than the effluent water quality standard.

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