## BIO-GEOCHEMICAL SIMULATION FOR SOLUTE TRANSPORT IN PIYUNGAN LANDFILL, YOGYAKARTA SPECIAL PROVINCE, INDONESIA

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#### Abstract

*Piyungan Landfill is the largest in Bantul Regency.* According to water quality sampling taken from a leachate pond, there are significant contaminant issues resulting from landfill leachate. The objectives of this research were achieved by applying a twodimensional bacteria mediated reduction numerical model was applied. Method of characteristic was applied to solve the advection part of the solute transport equation. Three bacteria  $(X_1, X_2, and X_3)$ groups were defined in the redox model. In the conceptual model, bacterial X<sub>1</sub> utilizes oxygen under aerobic conditions and nitrate, NO<sub>3</sub><sup>-</sup> under aerobic conditions as electron acceptors. Consequently, under aerobic conditions bacteria X<sub>2</sub>, and X<sub>3</sub> uti*lize MnO*<sub>2</sub>, and Fe(OH)<sub>3</sub> respectively as electron acceptors. In the redox model organic carbon which was defined as CH<sub>2</sub>O was considered as the electron donor for all bacteria mediated reduction reactions. The results of research are to improve the understanding of biogeochemical processes in aquifer. Keywords: landfill leachate, hydrogeological factors, numerical model, redox modeling.

#### 1 Introduction

Piyungan landfill is a dumping site of waste disposal from human activities' products, which has operated since 1995 and is the biggest landfill in Bantul Regency. Day by day the wastes are increasing in Yogyakarta and dumped in Piyungan landfill that can produce leachate (wastewater from leachate), which gives value of contaminant concentration. Based on Putra *et al.* (2001), the research for two-dimensional horizontal solute transport had been done; the plume was extended along the flow which moves from southern to northern part (Putra, 2001).

Furthermore, the existing of sanitary landfill still can bring a negative impact to the surrounding environment. The problems of groundwater contamination from leachate leakage always occur on the aquifer on the landfill area. Issue of groundwater contamination such as heavy metal and dissolved organic carbon, caused by leachate leakage in Piyungan Landfill has been reported by the local communities of Banyakan village (Putra *et al.*, 2001). Banyakan village is located more or less 700 m northern of Piyungan landfill (Figure 1).

Beside that there are still many researches of groundwater in Yogyakarta which are being performed. Some graduation papers of undergraduate and postgraduate students of Gadjah

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Figure 1: Location map and tree-dimensional view of the study area (North-South)

Mada University are contributed to groundwater problem in Bantul regency. Although many researches of groundwater had been done in this landfill, the study about contaminant transport from Piyungan landfill must be continued to give more understanding about redox reaction in groundwater systems.

Therefore, in order to give more understanding of contaminant transport from landfill, the modeling of redox from landfill will be focused on this research works must be simulated.

## 2 Site condition

This study area occupies the morphology of the river plains of the railroad. It is the result of fluvial deposition of Opak river. This unit spreads widely about 10.58% of the total mapped area. As well as alluvial deposits, outcrops of this unit found along river banks of Opak (Putra, 2001). Description and the characteristics of Opak fluvial deposit is as follows: spreading along the flow of Opak river, browngray, clayey sand-sized further down the coarse grain size and gravel, sorting is quite good, layered with changes not clear, and below are the tuffaceous sandstones and agglomerates. The thickness on the river bank of Opak river reachs approximately 12-15 meters (see Figure 2).

## 3 Theoretical Background

Based on the research of Perera *et al.* (2009), the model explains the utilization of  $O_2$ ,  $NO_3^-$ ,  $MnO_2$ , Fe(OH)<sub>3</sub> and  $SO_4^{2-}$  as electron acceptors for oxidation of organic carbon in the aquifer under aerobic and anaerobic conditions. The conceptual model consists of three different phases named as bio phase, mobile phase and matrix phase. Model parameters are adopted from literature on bacteria mediated multi-component modelling and bioremediation processes. Monod kinetic equation is used to formulate the bacterial growth (Perera *et al.*, 2009). The model explains the behaviours of aerobic and anaerobic bacteria under the avail-



Figure 2: Geological map of study area (redrawn and modified from Putra, 2001)

ability of organic carbon. The redox simulation of this research focuses on the possibility of the simulation of the formation of reduced environments in groundwater aquifers which purpose to know and understand the redox reaction in groundwater environment (Figure 3).

Bacteria group  $X_1$  uses oxygen under aerobic conditions and  $NO_3^-$  under anaerobic conditions as electron acceptor. Anaerobic bacterial groups  $X_2$ ,  $X_3$  and  $X_4$  use  $MnO_2$ , Fe(OH)\_3 and  $SO_4^{2-}$  as electron acceptors respectively (Perera *et al.*, 2009). Organic carbon which behaves as the electron donor is considered as the most important factor for the bacteria mediated reduction processes (Figure 3). On the Figure 3 shows the chemical species considered in the redox model and species exchange between different phases. The redox model describes the interactions between  $O_2$ ,  $NO_3^-$ ,  $MnO_2$ , Fe(OH)\_3,  $SO_4^{2-}$ , organic carbon concentrations, bacterial growth, precipitation of  $Fe(OH)_3$  in the mixing zone of the water region source.



Figure 3: Conceptualization of redox model (Perera *et al.,* 2009)

Exchange processes considered between the different model phases can be summarized as; (a) the mobile phase and the bio phase; (b) the mobile phase and the matrix phase; and (c) the bio phase and the matrix phase. The redox model describes the biological degradation of organic carbon by different bacterial groups and the precipitation of Fe(OH)<sub>3</sub> and FeS (Perera *et al.*, 2009). Microbial mediated redox sequences of reactions as shown in Figure 4, (aerobic oxidation,  $NO_3^-$  reduction,  $MnO_2$  reduction, Fe(OH)<sub>3</sub> reduction and  $SO_4^{2-}$  reduction) are modelled with four bacteria groups ( $X_1$ ,  $X_2$ ,  $X_3$ , and  $X_4$ ).

The general form of contaminant transport equation for the mobile components in the aqueous phase with the reaction term can be written as follow (Perera *et al.*, 2009):

$$\frac{\partial C_{mob}}{\partial t} + \frac{u'\partial C_{mob}}{\partial x} + \frac{v'\partial C_{mob}}{\partial y} = \\ \frac{\partial}{\partial x} \left( D_{xx} \frac{\partial C_{mob}}{\partial x} + D_{xy} \frac{\partial C_{mob}}{\partial y} \right) + \\ \frac{\partial}{\partial y} \left( D_{yy} \frac{\partial C_{mob}}{\partial y} + D_{yx} \frac{\partial C_{mob}}{\partial x} \right) + \sum_{i=1}^{3} S_{i}$$
(1)

Where Cmob is the mobile phase species concentration. u' and v' are the components of real

Aerobic respiration $\rightarrow$ Denitrification $\rightarrow$ MnO <sub>2</sub> reduction $\rightarrow$ Fe(OH) <sub>3</sub> reduction $\rightarrow$ SO <sub>4</sub> <sup>2-</sup> reduction $\rightarrow$ Precipitation of Fe(OH) <sub>3</sub> , FeS, MnS.							
	Bacteria	Reaction					
	X1(aerobis/anaerobis basteria)	$CH_2O+O_2 \rightarrow HCO_3'+H^+$					
		$5CH_2O+4NO_3 \rightarrow 5HCO_3 + H^*+2N_2+2H_2O$					
	X2(manganese reducing bacteria)	$\mathbb{C}\mathrm{H_2O}\text{+}2\mathrm{Mn^{4+}2H_2O} {\rightarrow} \mathrm{HCO_3^{-+}5H^{+}\text{+}2Mn^{3+}}$					
	X <sub>3</sub> (iron reducing bacteria)	$\mathrm{CH_{2}O+4Fe^{3+2}H_{2}O{\rightarrow}\mathrm{HCO_{3}^{-}+5H^{+}+4Fe^{2+}}}$					
	$X_4$ (sulfate reducing bacteria)	$\mathbb{C}\mathrm{H}_{3}\mathrm{O}+\mathrm{SO}_{4}{}^{2}  2\mathrm{H}\mathrm{CO}_{3}{}^{*}+\mathrm{H}^{+}+\mathrm{H}\mathrm{S}^{*}$					

Figure 4: Sequence of major biological reactions considered in the biogeochemical model (Perera *et al.*, 2009)

pore velocities in x and y directions.  $S_i$  is the source/sink term which represents the solute exchange between different phases.

The dispersion coefficients, dependent on the real pore velocity, are represented as follows (in Perera *et al.*, 2008):

$$D_{xx} = \frac{\alpha_L u'^2}{V} + \frac{\alpha_T v'^2}{V} + \tau \cdot D_M$$
  

$$D_{yy} = \frac{\alpha_T u'^2}{V} + \frac{\alpha_L u'^2}{V} + \tau \cdot D_M$$
  

$$D_{xy} = D_{yx} = \frac{(\alpha_L - \alpha_T)u'v'}{V}$$
(2)

Where  $\alpha_L$  and  $\alpha_T$  are the microscopic dispersion lengths related to the sand particle diameter,  $v = \sqrt{(u'^2 + v'^2)}$ , and  $D_M$  is the fluid molecular diffusion coefficient.  $\tau$  is the tortuosity (Perera *et al.*, 2008).

The source-sink term ( $S_i$ ) of Equation (3) describes the solute exchange between model phases. The solute exchange driving force is the difference between the concentrations of the species in different phases. The following equations describe the solute exchange between model phases (Perera *et al.*, 2009).

The exchange of solute between the mobile phase and the bio phase  $S_1$  is defined as:

$$S_{1} = \frac{\alpha(1-\varepsilon_{0})}{a} \cdot \frac{\theta_{bio}\sqrt{D_{L}}}{\theta_{bio}+\theta_{w}} (C_{bio} - C_{mob})$$
  
$$= \alpha' (C_{bio} - C_{mob})$$
  
$$\alpha' = \frac{\alpha(1-\varepsilon_{0})}{a} \cdot \frac{\theta_{bio}\sqrt{D_{L}}}{\theta_{bio}+\theta_{w}}$$
(3)

The exchange of solute between the mobile phase and the matrix phase  $S_2$  is defined as:

$$S_{2} = \frac{\beta(1-\varepsilon_{0})}{a} \cdot \frac{\theta_{mat}\sqrt{D_{L}}}{\theta_{mat}+\theta_{w}} (C_{mat} - C_{mob})$$
  
$$= \beta' (C_{mat} - C_{mob}) \qquad (4)$$
  
$$\beta' = \frac{\beta(1-\varepsilon_{0})}{a} \cdot \frac{\theta_{bio}\sqrt{D_{L}}}{\theta_{bio}+\theta_{w}}$$

The exchange of solute between the bio and matrix phases  $S_3$  is defined as:

$$S_{3} = \frac{\gamma(1-\varepsilon_{0})}{a} \cdot \frac{\theta_{mat}\sqrt{D_{L}}}{\theta_{bio}+\theta_{mat}} (C_{bio} - C_{mat})$$
  
$$= \alpha' (C_{bio} - C_{mat}) \qquad (5)$$
  
$$\gamma' = \frac{\gamma(1-\varepsilon_{0})}{a} \cdot \frac{\theta_{bio}\sqrt{D_{L}}}{\theta_{bio}+\theta_{w}}$$

Where  $C_{mob}$ ,  $C_{bio}$  and  $C_{mat}$  are the concentrations of solute in the mobile phase, bio phase and matrix phase respectively.  $\alpha$  is the exchange coefficient between bio phase and mobile phase.  $\beta$  is the exchange coefficient between matrix phase and mobile phase.  $\gamma$  is the exchange coefficient between bio phase and matrix phase.  $\alpha'$ ,  $\beta'$  and  $\gamma'$  are the simplified exchange coefficients.  $\varepsilon_0$  is the initial porosity, *a* is the diameter of uniform soil particle and bio, mat and w are the specific volume of bio, matrix and mobile phases respectively.  $D_L$  is the dispersion coefficient (Perera *et al.*, 2009).

#### 4 Methods

Finite different approximation is used to solve the problem. The finite difference approximations are derived from the fundamental definition of derivative and Taylor series expansion of a function f(x). When a function f(x) and its derivatives are single valued, finite, and continuous function of x, then by Taylor's series (in Jinno *et al.*, 2001).

The method of characteristic is one of the solution technique used to solve the solute transport in porous media. It was originally applied by Garder *et al.* (1964) for calculation of miscible displacement in reservoir simulation

#### 5 Results and Discussions

The some parameters to input the redox model were show in Keophousone *et al.* (2010a). Based on Keophousone *et al.* (2010b), the research of two-dimensional groundwater flow was simulated; therefore the flow modeling has been simulated again for satisfying with parameters data and site situation. Beside the flow simulation, the solute transport has been simulated either (in Keophousone *et al.*, 2010c), and the calibration of parameter were edited afterward to make the model in reality.

The redox model was applied to the selected cross section under the following chemical species distribution which is shown in Table 1. Redox model was applied for two years of simulation and results are shown for bacteria growth, mobile phase species and matrix phase species along the borehole BH1 were chosen. Thus, the organic carbon supply from the matrix phase was kept constant and the velocity profile was followed as done for flow simulation (Keophousone et al., 2010). Moreover, constant supplies of MnO<sub>2</sub> are also assigned. The organic carbon (1.0 mg/L; Perera et al., 2009) is continuously supplied from the top soil layer where cocist of sandy silt. Oxygen is infiltrated from the ground surface at the constant concentration of 8 mg/L with the infiltrated water (Perera et al., 2009).

On Table 1 shows the initial conditions assigned for redox model and the assigned boundary conditions for the simulation. The initial bacterial concentrations for aerobic and anaerobic bacteria were kept as 0.001 mg/L. Redox model was applied for two years simulation and results are shown for bacteria growth, mobile phase species and matrix phase species along the borehole BH1. As shown on Figure 6 to 8, the obtained numerical results show the formation of two reduced zones. One reduced zone has been formed at depths between 0.0 m and 0.7 m, and another one has been formed below 2.3 m depth, this result can be observed by matrix phase (Figure 7). The reason for the redox zone is obvious.

## Aerobic Oxidation, Denitrification and Growth of Bacteria $X_1$

According to Figure 6, bacteria  $X_1$  growth is dominant in the freshwater region (Figure 6) and no or less growth can be seen in the anaerobic source region of the aquifer. The gradual decrease of CH<sub>2</sub>O in the aerobic freshwater region is the main reason for the decrease of bacteria  $X_1$  growth after 30 days.

Due to the consumption of CH<sub>2</sub>O in the freshwater region of bio phase by bacteria X<sub>1</sub>, CH<sub>2</sub>O in the freshwater region of the matrix phase is decreased and it is correctly simulated and shown in Figure 7. The growth of bacteria  $X_1$  in the freshwater region is controlled by the available CH<sub>2</sub>O. As a result of continuous infiltration of rainwater with the 8 mg/L of O<sub>2</sub> concentration, there is enough  $O_2$  for bacteria  $X_1$  to grow. However, there is no constant CH<sub>2</sub>O supply for the freshwater region. Therefore, when the available CH<sub>2</sub>O is consumed, the growth of bacteria  $X_1$  hinders. Moreover, in Figure 8,  $NO_3^-$  concentration decreases gradually due to the denitrifying (NO<sub>3</sub><sup>-</sup> reduce to bacteria  $X_1$ ) process of bacteria  $X_1$ . The concentration variations of  $O_2$  and  $NO_3^-$  with the metabolism of bacteria  $X_1$  imply that the redox model can simulate the growth of aerobic and denitrify bacteria under the available CH<sub>2</sub>O appropriately.

#### Reduction of MnO<sub>2</sub> and growth of bacteria X<sub>2</sub>

Formation of  $Mn^{2+}$  and  $Fe^{2+}$  could be observed as a result of the reduction of  $MnO_2$  and  $Fe(OH)_3$  below 2.3 m. Bacteria  $X_2$  reduce  $MnO_2$  to  $Mn^{2+}$  under anaerobic conditions. Figure 7 shows the growth of  $MnO_2$  reducing bacteria  $X_2$ . The decrease of  $MnO_2$  in the matrix phase (Figure 7) corresponds well with the bacteria  $X_2$  growth which is shown in Figure 6.

Chamical Spacing	Mobile phase (mg/L)		Bio phase	Matrix phase
Chemical Species	Fresh water	Source water	(mg/L)	(mg/L)
O <sub>2</sub>	0.0	3.4	0.0	-
NO <sub>3</sub> -	0.05	0.002	0.0	-
MnO <sub>2</sub>	-	-	0.0	5.0
Fe(OH) <sub>3</sub>	-	-	0.0	1.5
Fe <sup>2+</sup>	0.0	0.0	0.0	-
Mn <sup>2+</sup>	0.0	0.0	0.0	-
CH <sub>2</sub> O	0.6	0.5	0.0	0.5
SO4 <sup>2-</sup>	12.0	500	-	-

Table 1: Initial chemical species distribution for redox model



Figure 5: Initial and boundaries of chemical species distribution for redox model



Figure 6: Numerical results for aerobic and anaerobic bacterial growth (at BH1)



Figure 7: Numerical results for concentration variations of the matrix phase species (at BH1)





Figure 8 shows the concentration of formed  $Mn^{2+}$  in mobile phase. Bacteria  $X_2$  growth is significant in the anaerobic source region of the aquifer. Constant CH<sub>2</sub>O layer provides abundant electron donor environment which is favorable for bacterial growth. According to Figure 6 growth of bacteria  $X_2$  increases up to 2 years. The reason for that is the availability of  $MnO_2$ . Bacteria  $X_2$  use available  $MnO_2$  in the organic carbon region. Availability of CH2O and MnO<sub>2</sub> under anaerobic condition is favorable for the growth of bacteria X<sub>2</sub>. Availability of CH<sub>2</sub>O and MnO<sub>2</sub> in the anaerobic region are the determining factors for bacteria  $X_2$ . Redox model is able to simulate the growth of bacteria X<sub>2</sub> appropriately under the assigned conditions.

# **Reduction of Fe(OH)\_3, the growth of bacteria** $X_3$ and precipitation of $Fe(OH)_3$

The reduction of Fe(OH)<sub>3</sub> occurs in the anaerobic region due to the metabolism of bacteria  $X_3$  in the redox model. On Figure 8 shows the formation of Fe<sup>2+</sup> in mobile phase due to the reduction of Fe(OH)<sub>3</sub> in the bio phase. Figure 6 shows the bacteria  $X_3$  growth for ten years while in Figure 7 shows the concentration changes of Fe(OH)<sub>3</sub> in the matrix phase.

The decrease of the growth of bacteria  $X_3$  in the constant CH<sub>2</sub>O layer is due to the consumption of  $Fe(OH)_3$  in the bottom of the aquifer. Therefore, bacteria X<sub>3</sub> grows in that region until all the  $Fe(OH)_3$  is reduced by bacteria  $X_3$ . The precipitation of formed Fe<sup>2+</sup> takes place in the mixing zone area, where O<sub>2</sub> is available. The reduction of Fe(OH)3 takes place in the lower part of mixing zone and anaerobic source region of the aquifer at different rates according to the availability of CH<sub>2</sub>O and Fe(OH)<sub>3</sub>. High concentration of dissolved Fe<sup>2+</sup> is a common water quality problem associated with groundwater. Therefore, the reduction of  $Fe(OH)_3$  is an important process to be considered in numerical simulations to enhance the understanding of biological involvement in groundwater.

The formed  $Fe^{2+}$  moves to the mixing zone with the advective flow and meets  $O_2$  and precipitates as  $Fe(OH)_3$ . The precipitation of  $Fe(OH)_3$  is highlighted as the increase of Fe(OH)<sub>3</sub> concentration in the matrix zone. In Figure 7, the gradual increase of Fe(OH)<sub>3</sub> concentration in the mixing zone region (elevation between 2.1 m to 2.3 m) depicts the precipitation of Fe(OH)<sub>3</sub>. The Fe<sup>2+</sup> concentration increases around the 2.3 m elevation in the mobile phase (Figure 8) is due the reduction of precipitated Fe(OH)<sub>3</sub> in bottom edge of the mixing zone.

#### 6 Conclusions

The calculation result shows that biochemical processes are important and must be taken to be considered in groundwater quality problem. The simulation results show the trends of groundwater quality alteration such as denitrification,  $MnO_2$  reduction and  $Fe(OH)_3$  reduction. Therefore, due to the limitations in the field measurements and difficulties in the biogeochemical parameter identification, it was impossible to simulate the exact situation which is prevailing in the Piyungan landfill area. Further efforts are needed to modify the redox model parameters in the case where more accurate groundwater chemistry is needed.

## Acknowledgements

This study has been supported financially by AUN/SEED-Net, JICA. The authors would like to acknowledge and express their sincere thanks and appreciations to Prof. Kenji Jinno. Our gratitude is further expressed to Regency of Bantul, Yogyakarta Special Province for their kind supporting and kind helps during data collection.

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