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Mathematical modeling of the formation of reduced environments in a coastal aquifer under the bacterial mediation

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Abstract: A numerical study of the bio-geochemical aspects of the coastal aquifers is an interesting subject to address. So far, numerical simulations which discuss about the bacteria mediated reduction processes are not common in the scientific literature. Snyder et al., (2004) made detailed surveys on the bio-geochemical oxidation-reduction approach for the mixing zone formed in a shallow aquifer at the estuary of Sapelo Island, Georgia, USA. Based on the bio-geochemical approaches, they have shown that the distribution of manganese, iron and sulphate ions are affected by the availability of natural organic carbon and bacteria mediated reducing processes, which take place at the both sides of fresh and seawater transition zone. Hiroshiro et al., (2006) analyzed the redox (oxidation-reduction) potential distribution in the monitoring wells and found out that the redox potential drastically decreased below the mixing zone from fresh water to reduced seawater.

In this paper, a numerical investigation (redox model) of the formation of reduced environments under the aerobic and anaerobic bacterial mediation at the availability of organic carbon is discussed. Developed redox model is applied to the selected cross section of Kujyukurihama beach of Chiba prefecture, Japan. Field investigations conducted in Kujyukurihama beach have found out that the measured oxidation-reduction potentials (ORP) at the seawater region of the aquifer show -380 mV, -99 mV and -66 mV at 8.0 m, 7.0 m and 5.0 m respectively. Moreover, the measurements conducted at different depths of this aquifer show high concentrations of reduced species in the anaerobic seawater region. The availability of oxygen and other electron acceptors such as NO_3^-, MnO_2, $Fe(OH)_3$ and SO_4^{2-} with the organic carbon as the electron donor encourage different bacteria to activate and create reduced environments in the subsoil. The present numerical simulation considers four bacterial groups named as X_1, X_2, X_3 and X_4 to simulate the reduction processes. Bacteria X_1 consumes oxygen in the aerobic condition and reduces NO_3^- in the anaerobic condition while X_2, X_3 and X_4 bacterial groups reduce MnO_2, $Fe(OH)_3$ and SO_4^{2-} respectively at the anaerobic condition utilizing the available organic carbon.

At the initial stage of modeling, a density dependent two dimensional model is used to obtain the steady state seawater and freshwater distribution of the selected cross section of the Kujyukurihama beach. The initial chemical species concentrations are assigned according to the distribution of seawater and freshwater in the aquifer. A 4.0 m thick organic carbon layer is assumed at the bottom of the aquifer in the numerical simulation. Then bacteria mediated redox model is applied to simulate the formation of reduced environment in the aquifer. In the redox model, three different phases are considered named as mobile phase, matrix phase and bio phase. All the bacterial activities are taken place in the bio phase. The species allocated in three phase can transfer between phases according to the concentration gradients created by bacterial metabolism take place in the bio phase. The numerical results are presented for the different chemical species and four bacterial groups which are considered in model. Numerical results show that the availability of organic carbon is an important factor which determines the formation of different reduced environments in the aquifer. Moreover, the results of this study show that, generally it is possible to simulate the formation of reduced environments at the subsurface of coastal aquifer using the bacteria mediated redox model. So far, this kind of numerical studies are very limited in the field of numerical modeling of coastal aquifer related phenomena. Therefore, in future more detailed numerical modeling studies are essential to explain the biogeochemical processes of coastal aquifers.

Keywords: *Bacteria mediated reduction, chemical transport/reaction, coastal groundwater, organic carbon*

1. INTRODUCTION

Geochemical properties of the groundwater in a coastal aquifer would change with the depth from aerobic state to anaerobic state. The bacteria mediated redox reactions are significant in the geochemical property changes of the subsurface water in a coastal aquifer. The availability of oxygen and other electron acceptors such as NO_3^-, MnO_2, $Fe(OH)_3$ and SO_4^{2-} with the organic carbon (CH_2O) as the electron donor encourage the different bacteria to activate and form reduced environments in the subsoil. The gradual decrease of oxygen with the depth allows bacteria to use other electron acceptors and form Mn^{2+}, Fe^{2+} and HS^- rich reduced environments. The seawater below the mixing zone is almost anaerobic in the coastal aquifer, and bacteria mediated reduction processes are dominant. Depending on the availability of organic carbon, a sequence of redox zones of increasing redox potential may develop at the down gradient of the aquifer; zones of oxygen reduction, NO_3^- reduction, MnO_2 reduction, $Fe(OH)_3$ reduction and SO_4^{2-} reduction can be developed if the organic carbon and corresponding electron acceptors are present in the aquifer under the bacterial mediation.

Jinno et al., (2001) researched on the geochemical properties of groundwater of the unconfined aquifer at Kujyukurihama coast in Chiba prefecture, Japan by analyzing the major ions. Seawater in the unconfined aquifer at Kujyukurihama coast has -380 mV oxidation-reduction potential at 8.0 m below the surface of the sand pit and the groundwater environment in the seawater region of the aquifer was considerably reduced. The measurements conducted at different depths of this particular aquifer show high concentrations of reduced species in the anaerobic seawater region (Table 1). As shown in Table 1, location (5) shows high DOC concentration. Few decades ago this area was sea and the sea bed was covered by sea weeds. After the continuous reclamation, the present sand pit has been formed. The sea weeds buried in the subsurface made such a large biodegradable organic carbon. The behaviors of different bacteria groups under aerobic and anaerobic conditions at the availability of electron donors (CH_2O) and electron acceptors (O_2, NO_3^-, MnO_2, $Fe(OH)_3$ and SO_4^{2-}) are numerically simulated by developing a bacteria mediated reduction model. The density difference between freshwater and seawater is one of the significant feature which is needed to be considered in the groundwater flow modeling of coastal aquifers. Therefore, the combination of density dependent flow with the bacteria mediated redox reactions is considered in this numerical simulation. Figure 1 shows the location of the selected place for the study. Due to the practical constrains, it is not possible to validate the model through a direct comparison between the measured data and numerically simulated results. Some of the practical constraints can be listed as: (i) those processes have been taking place for thousands of years. (ii) history of the geological formation of the coastal aquifer is unknown. (iii) continuous measurement of chemical species which reside in that environment, is costly. Due to those reasons, validation of the numerical model through a direct comparison is not discussed. Instead of that, authors tried to simulate the possible trends of bacteria mediated reduction processes. The availability of organic carbon is the major factor which influences the formation of reduced environments under the mediation of aerobic and anaerobic bacteria. In this numerical study, the behaviors of aerobic and anaerobic bacteria under the assigned organic carbon distribution and the formation of Mn^{2+}, Fe^{2+} and HS^- at the reduction of MnO_2, $Fe(OH)_3$ and SO_4^{2-} are discussed for the selected cross section of the site. In the first part of the simulation, a density dependent two-dimensional solute transport model is used to obtain the steady state conditions for the chloride distribution and velocity distribution of the unconfined aquifer. The validity of the two dimensional density dependent model was confirmed by Jinno et al., 2001 and Nakagaga et al., 2002. The concentration distributions of chemical species in the freshwater and seawater are different due to the physical, chemical and biological processes in subsurface environments. For this reason, it is initially necessary to obtain the chloride distribution of the selected cross section of the aquifer. The initial distribution of chemical species in freshwater, mixing zone and seawater regions are assigned according to the obtained chloride distribution. These species distribution and velocity distribution are input to the two-dimensional mass transport and bacteria mediated reduction model (redox model).

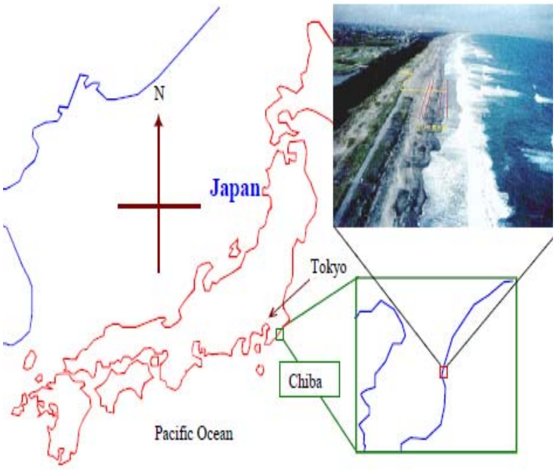

Figure 1. Location map of the site

2. NUMERICAL SIMULATION

2.1 Bacteria mediated redox model

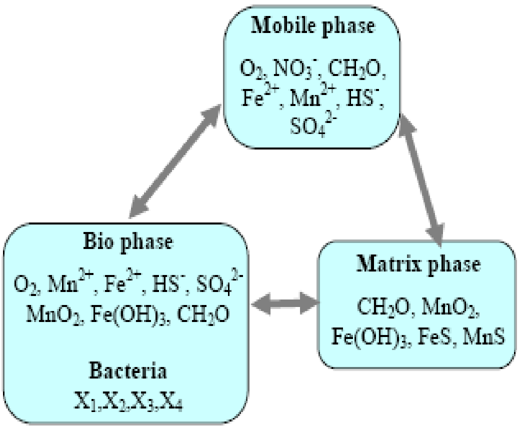

Figure 2. Chemical species considered in the model and mass transfer processes between different phases.

The redox model describes the biological degradation of dissolved organic carbon (CH_2O) by different bacteria groups. Bacteria mediated redox sequence of reactions (aerobic oxidation, nitrate reduction, Mn(IV) reduction, Fe(III) reduction and SO_4^{2-} reduction) are modeled with four bacteria groups named as X_1, X_2, X_3 and X_4. Bacteria X_1 uses oxygen under aerobic conditions and nitrate under anaerobic conditions. Under anaerobic conditions bacterial groups X_2, X_3 and X_4 use Mn (IV), Fe (III) and SO_4^{2-} as electron acceptors respectively and reduce them to Mn^{2+}, Fe^{2-} and HS^- .The redox model takes into account three different phases: mobile pore water phase, immobile bio phase and matrix phase. The bio phase is assumed to include all bacterial growth and biological processes. The species located in three phases can transport between different phases according to the concentration gradients. Bacterial activities in bio phase generate concentration gradients between different phases. Redox model simulates the concentration variations due to the bacterial activities taking place in bio phase. Figure 2 shows three phases, which are considered in the redox model and the chemical species those reside in each phase. The two-dimensional partial differential equation for the advective-dispersive solute transport with the chemical reaction term for the mobile phase species can be written as:

$$\frac{\partial(\theta C_{mob})}{\partial t}+\frac{\partial(uC_{mob})}{\partial x}+\frac{\partial(vC_{mob})}{\partial y}=\frac{\partial}{\partial x}\left(\theta D_{xx}\frac{\partial C_{mob}}{\partial x}+\theta D_{xy}\frac{\partial C_{mob}}{\partial y}\right)+ \frac{\partial}{\partial y}\left(\theta D_{yy}\frac{\partial C_{mob}}{\partial y}+\theta D_{yx}\frac{\partial C_{mob}}{\partial x}\right)+\sum_{i=1}^{3}S_i \tag{1}$$

where C_{mob} is the mobile phase species concentration. S_i is the source/sink term which represents the solute exchange between different phases. D_{xx}, D_{xy}, D_{yy}, D_{yx} are the dispersion coefficients, θ is the volumetric moisture content. The solute exchange processes are: (i) mass transfer between the mobile phase and the bio phase, $S_1=\alpha(C_{bio} - C_{mob})$; (ii) mass transfer between the mobile phase and the matrix phase, $S_2=\beta(C_{mat} - C_{mob})$; and (iii) mass transfer between the bio phase and the matrix phase, $S_3=\gamma(C_{bio} - C_{mat})$. C_{bio} and C_{mat} are the concentrations of chemical species in bio phase and matrix phase, α, β and γ are the exchange coefficients between the different phases. Bacteria can utilize several substrates simultaneously. The specific growth rate of bacteria is assumed to be a function of the concentration of the substrates. As described by the ecological redox sequence, microbially mediated redox processes are described by following reactions:

Aerobic respiration (bacteria X_1) => $CH_2O + O_2 \rightarrow CO_2 + H_2O$
Denitrification (bacteria X_1- anaerobic condition) => $CH_2O + 4/5NO_3^- + 4/5H^+ \rightarrow CO_2 + 2/5N_2 + 7/5H_2O$
Manganese dioxide-reduction (bacteria X_2) => $CH_2O + 2MnO_2 \rightarrow 2Mn^{2+} + 3H_2O + CO_2$
Iron hydroxide-reduction (bacteria X_3) => $CH_2O + 4Fe(OH)_3 + 8H^+ \rightarrow 4Fe^{2+} + 11H_2O + CO_2$
Sulphate reduction (bacteria X_4) => $CH_2O + 1/2SO_4^{2-} + 1/2H^+ \rightarrow CO_2 + 1/2HS^- + H_2O$

The growth of bacteria is described by the Double Monod kinetic equation as (Schafer et al., 1998):

$$\frac{\partial X}{\partial t}=v_{\max}\frac{C_1}{K_{s1}+C_1}\cdot\frac{C_2}{K_{s2}+C_2}X \tag{2}$$

where v_{max} is the maximum growth rate, C_1 is the electron donor concentration in bio phases, C_2 is the electron acceptor concentration in bio phases, K_{s1} is the primary substrate half-saturation constant, K_{s2} is the secondary substrate half-saturation constant, and X is the bacterial concentration.

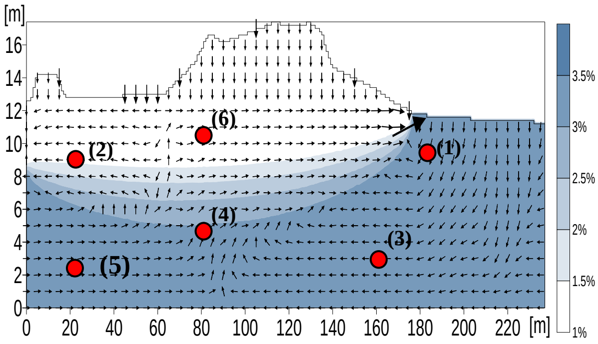

Figure 3. Steady state chloride distribution (%) and velocity distribution for the two-dimensional density dependent solute transport model. (*Selected cross section of Kujyukurihama coast, Chousei village, Chiba, Japan*)

Table **1** Measured ground water chemistry of Kujyukurihama beach

Location	(1)	(2)	(3)	(4)	(5)	(6)
pH	8.1	7.9	8.2	7.2	6.9	7.6
Na^+ (mg/l)			4500	5900	5500	120
K^+ (mg/l)			170	220	210	15
Ca^{2+} (mg/l)			190	240	220	45
Fe^{2+} (mg/l)			<0.1	0.2	<0.1	<0.1
Total – Mn (mg/l)			0.06	1	0.17	0.04
Cl- (mg/l)			7300	9500	9100	230
SO_4^{2-} (mg/l)			1000	1200	980	40
DO (mg/l)	1.7	<0.5	(4.1)	(2.9)	<0.5	3.5
DOC (mg/l)			5	5	11	<1
TOC (mg/l)			7	14	20	1
ORP (mV)			-66	-99	-380	78
S^{2-} (mg/l)			<0.5	<0.5	11	<0.5

Table **2** Parameters used for the simulation.

Parameter		Value
Exchange coefficient	α	3 day^{-1}
	β	0.5 day^{-1}
	γ	0.005day^{-1}
Half-saturation constant	K_{CH2O}	0.10 mmol/L
	K_{O2}, K_{NO3}, K_{MnO2}, $K_{Fe(OH)3}$, K_{SO4}	1.0 x 10^{-3} mmol/L
Aerobic bacteria X_1	Max. growth v_{max}	3.0 day^{-1}
	Decay rate v_{X1dec}	0.45 day^{-1}
Denitrification bacteria X_1	Max. growth v_{max}	2.25 day^{-1}
	Decay rate v_{X1dec}	0.34 day^{-1}
Mn(II)-reduction bacteria X_2	Max. growth v_{max}	0.5 day^{-1}
	Decay rate v_{X2dec}	0.075 day^{-1}
Fe(III) reduction Bacteria X_3	Max. growth v_{max}	0.5 day^{-1}
	Decay rate v_{X3dec}	0.075 day^{-1}
Sulfate reduction Bacteria X_4	Max. growth v_{max}	0.45 day^{-1}
	Decay rate v_{X4dec}	0.067 day^{-1}

Figure 3 shows the numerical results obtained for the steady state of chloride distribution and velocity distribution for the selected cross section. In this study, the finite difference method and the method of characteristics are used as numerical solution techniques to solve the model equations The model for the biologically mediated redox processes is highly complex, as it needs to involve a large number of parameters. Monod kinetic and stoichiometric parameters are taken from several studies related to modeling and simulation of redox processes (Lensing et al.,1993). The parameters are adjusted to obtain the realistic result sets. The values of the kinetic and stoichiometric parameters are listed in Table 2. The initial and boundary conditions are selected concerning the existing geological conditions of the Kujyukurihama coast. The initial concentration of 0.001 mg/l is assigned for the bacteria X_1, X_2, X_3 and X_4 reside in the bio phase. The growth of bacteria, consumption of CH_2O and formation of Mn^{2+} ,Fe^{2+} and HS^- in the reduced conditions are presented. 8 mg/l of oxygen is infiltrated from to ground surface with the infiltrated freshwater.

2.2 Numerical results and discussion

In the numerical simulation, a uniform 10 mg/l of organic carbon (as shown in Figure 4) layer of 4.0 m thick is assumed at the bottom part of the coastal aquifer. In the redox model, matrix phase provides CH_2O for bacterial metabolism. Constant CH_2O layer provides abundant electron donor environment which is favorable for bacterial growth. Since the assumed CH_2O layer is located in the oxygen low bottom part of the aquifer, anaerobic bacteria growth is significant. Table 3 shows the initial concentrations assigned for the species reside in mobile, bio and matrix place of the redox model. Figure 4 shows the schematic diagram for the selected cross section of the numerical model. The numerical results obtained are explained below for the borehole No 4. The calculation is carried out for 2 years. Figure 5 shows the growth of bacteria X_1, X_2, X_3 and X_4 under the assigned distribution of CH_2O. The concentration distributions of O_2, NO_3^-, Mn^{2+}, Fe^{2+}, and HS^- in the mobile phase along the depth of bore hole No. 4 for 2 years are shown in Figure 6. Figure 7 explains the concentration variation of CH_2O, MnO_2 and $Fe(OH)_3$ in the matrix phase.

2.2.1 *Numerical Results of Bacteria X_1 Growth and Reduction of O_2 and NO_3^-*

According to the graph (a) of Figure 5, bacteria X_1 growth is dominant in the freshwater region and zero growth can be seen in the anaerobic seawater region of the aquifer. Until 30 days, bacteria X_1 grow rapidly. After that due to the gradual decrease of CH_2O due to consumption in the aerobic freshwater region, bacteria X_1 growth decrease. The growth of bacteria X_1 in the freshwater region is controlled by the available CH_2O.

Table **3** initial background distributions of species for the redox model

Chemical Species	Mobile phase (mg/l)		Bio phase (mg/l)	Matrix phase (mg/l)
	Fresh water	Sea water		
O_2	3.5	0.0	0.0	
NO_3^-	0.05	0.0	0.0	
MnO_2			0.05	5.0
$Fe(OH)_3$			0.05	5.0
Fe^{2+}	0.0	0.0	0.0	
Mn^{2+}	0.0	0.0	0.0	
CH_2O	0.5	10.0	0.05	5.0
SO_4^{2-}	40.0	2300.0	0.0	
HS^-	0.0	0.0	0.0	

As a result of continuous infiltration of rainwater, O_2 concentration close to the ground surface increases. Elevation between 4.0 m and 14.0 m, O_2 concentration decreases and forms O_2 low region. Moreover, in Figure 6 (b), NO_3^- concentration decreases gradually due to the denitrifying process of bacteria X_1 under low O_2 condition. Denitrification is rapid with decrease of O_2.

2.2.2 *Numerical Results of Bacteria X_2 Growth and Reduction of MnO_2*

Reduction of MnO_2 is carried out by the anaerobic bacteria X_2. Bacteria X_2 growth is significant in the anaerobic seawater region of the aquifer up to 30 days (Figure 5 (b)). After that bacteria X_2 growth in the CH_2O layer becomes lower. The reason for that is that the gradual decrease of MnO_2 due to the consumption by bacteria X_2.

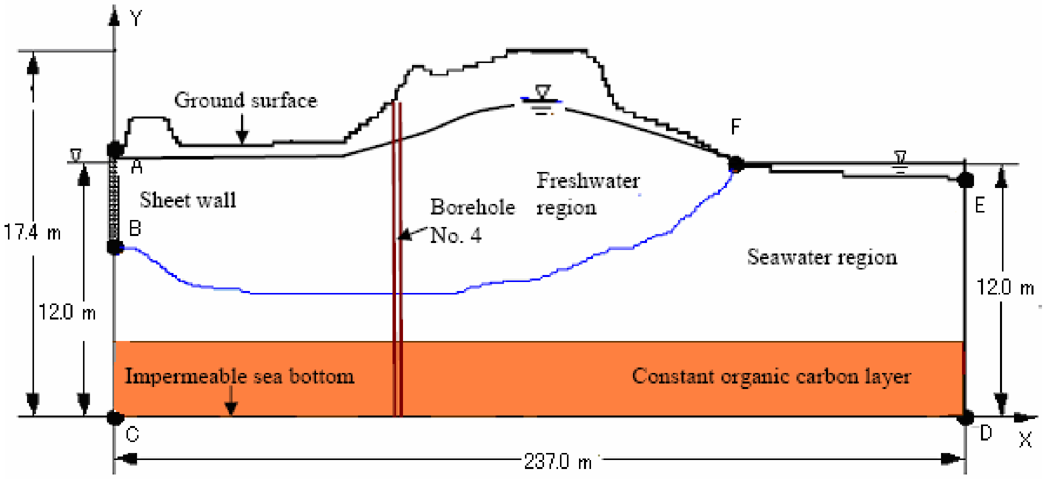

Figure 4. Schematic diagram for numerical simulation

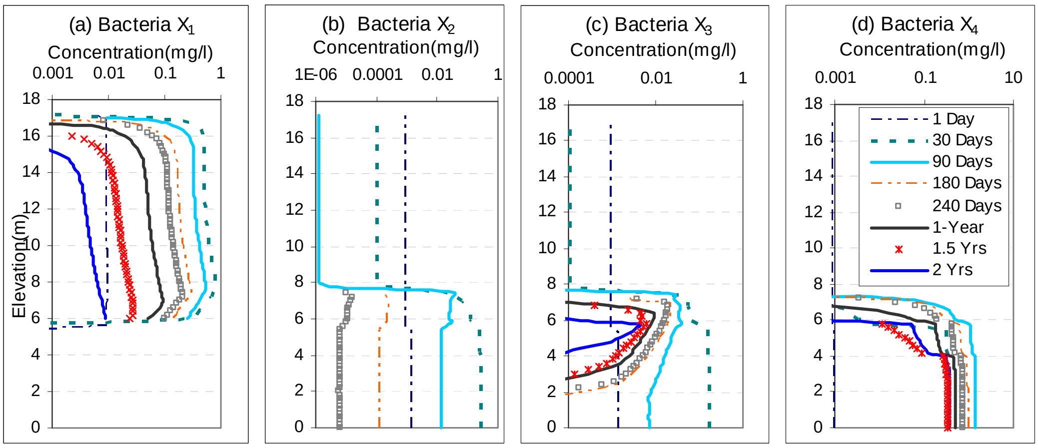

Figure 5. Aerobic and anaerobic bacterial growth

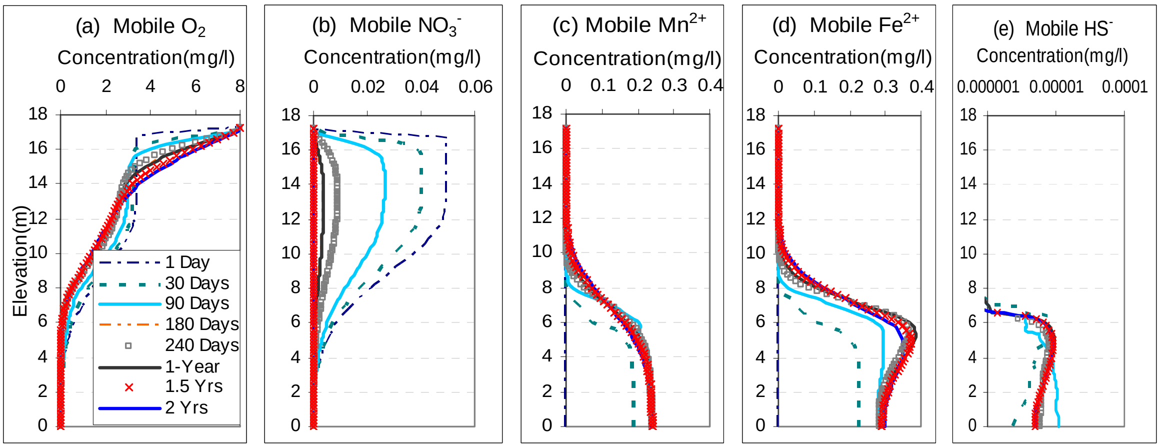

Figure 6. Concentration variations of mobile phase species

However, gradual consumption of MnO_2 in the CH_2O layer forms a MnO_2 low region in the CH_2O layer, then the bacteria X_2 growth in this area become low while in the region just above the CH_2O layer bacteria X_2 growth becomes significant until the total extinction of MnO_2. In Figure 5 (b) after 30 days, the bacteria X_2 growth in the elevation between 6.0 m and 8.0 m is little bit larger than in the constant CH_2O layer. After the consumption of all available MnO_2, the growth of bacteria X_2 becomes zero. The concentration of formed Mn^{2+} in the mobile phase has also become steady after 90 days showing no further generation of Mn^{2+}.

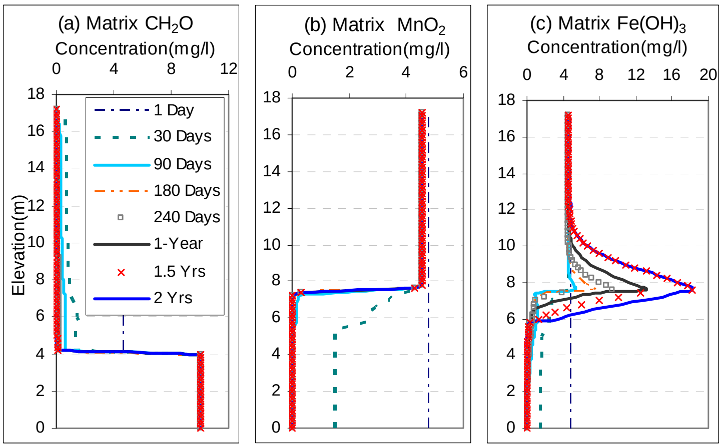

Figure 7. Concentration variations of matrix phase species

2.2.3 Numerical Results of Bacteria X_3 Growth and Reduction of $Fe(OH)_3$

The reduction of $Fe(OH)_3$ is occurred in the anaerobic region due to the activation of bacteria X_3 in the redox model. Figure 6 (d) shows the formation of Fe^{2+} in mobile phase due to the reduction of $Fe(OH)_3$ in the bio phase. Figure 5 (c) shows the bacteria X_3 growth for 2 years while in Figure 7 (c) shows the concentration changes of $Fe(OH)_3$ in the matrix phase. Up to 30 days the growth of bacteria X_3 increases and after that it starts to decrease in the constant CH_2O layer region due to the decrease of $Fe(OH)_3$ in the matrix phase in the bottom of the aquifer. Even though the $Fe(OH)_3$ in the CH_2O layer becomes zero after 90 days, still there are some $Fe(OH)_3$ above the CH_2O layer. Therefore, bacteria X_3 grows in that region until all the $Fe(OH)_3$ is reduced by bacteria X_3. The region above constant CH_2O layer, the determining factor is CH_2O while in the CH_2O layer bacteria growth is determined by the availability of $Fe(OH)_3$. The precipitation of formed Fe^{2+} takes place in the mixing zone area, where O_2 is available. The reduction of $Fe(OH)_3$ takes place in the mixing zone and anaerobic seawater region of the aquifer at different rates as explained above. The formed Fe^{2+} moves to the mixing zone with the flow and meets O_2 and precipitates as $Fe(OH)_3$. The precipitation of $Fe(OH)_3$ is highlighted in Figure 7 (c). As the concentration of $Fe(OH)_3$ increases of in the matrix phase particularly the elevation between 6.0 m to 10.0 m. The Fe^{2+} concentration increase around the 6.0 m elevation in the mobile phase (Figure 6 (d)) is due the reduction of precipitated $Fe(OH)_3$ in bottom edge of the mixing zone. The numerical results for the bacteria X_3 and reduction/precipitation of $Fe(OH)_3$ convince that the redox model simulates the bacteria X_3 growth and its related processes correctly..

2.2.4 Numerical Results of Bacteria X_3 Growth and Reduction of SO_4^{2-}

Bacteria X_4 responsible to the reduction of SO_4^{2-} in the anaerobic seawater in the aquifer. In the seawater region of the aquifer, abundant amount of SO_4^{2-} is available (Table 1). Therefore the reduction of SO_4^{2-} is determined by the availability of CH_2O. However, reduction of SO_4^{2-} is a slow process and it follows bacteria X_2 and X_3. Figure 6 (e) shows the gradual formation of HS^- as a result of SO_4^{2-} reduction by bacteria X_4. The continuous supply of CH_2O and SO_4^{2-} make the bacteria X_4 growth in the CH_2O layer almost steady state after 1.5 years. If the calculation continuous for a long time period the reduction of SO_4^{2-} would continuously take place forming a HS^- rich reduced environment. The numerical results of bacteria X_4 also suggest that the redox model is able to simulate the formation of reduced environments in the costal aquifer.

3 CONCLUSION

This study combined two numerical models to simulate the bio-geochemical reduction processes which are taking place in a coastal aquifer at Kujyukurihara coast in Chiba Prefecture, Japan. This type of numerical study is necessary at the occasions where complex hydro and bio-geological processes are involved. Density dependent flow with the bacteria mediated reduction processes is a new study area in the field of numerical simulations. Therefore, the authors' objective is to encourage the numerical modelers to focus on the development of numerical models which can address the coastal aquifer bio-geochemical processes extensively. The results from this study show that it is generally possible to simulate the formation of redox environments at the subsurface of coastal aquifer using the bacteria mediated solute transport model. The model is able to simulate the behaviors of various chemical species present in the coastal subsurface under the availability of organic carbon as electron donor. Numerical results show that the distribution of organic carbon governs the bacterial activities when the suitable electron acceptors are available. In the natural environment the formation of reduced environments involves large number of chemical reactions and it takes very large time period also. It is difficult to include all the reactions taking place in a coastal aquifer to a numerical model. However, in this simulation the basic reduction reaction sequence has been tested by assuming a CH_2O layer at the bottom of the aquifer. The results show that the model responds well to the CH_2O and other electron acceptor distribution. Moreover, this type of model can be used to test the bioremediation methods such as injection or infiltration of secondary waste water into a coastal aquifer. Therefore this type of numerical model is very important in understanding the coastal water quality formation. Even this model is based on the hydro-geological features of the Kujyukurihara coast, with appropriate modifications this model can be used for other coastal aquifers too.

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