

Effect of Climatic Change on Groundwater Quality Around the Subsurface Dam

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ABSTRACT: The effect of climatic change on groundwater quality around subsurface dam area has been observed using numerical simulation. The study is related to sustainable groundwater development from subsurface dams constructed in the south islands of Japan where limestone is the underground geology. The model includes dispersion/diffusion, advection, ion exchange, formation of complexes in the aqueous phase, and the dissociation of water. The mass action, transport, and site action equations are expressed in a differential/algebraic form and solved by FEM. Results reveal that dissolution of limestone is proportional to the acidification of rainwater i.e. inversely proportional to the pH of rainwater. The resulting increase in calcium ion concentration is expected to block the filter of the pumping well and deteriorate the quality of groundwater as well. Again, dissolution of limestone is proportional to the increase of intensity of rain that leads to the increase of velocity of water. Dissolution of limestone was inversely proportional to the temperature.

Keywords: Limestone dissolution; Climatic change;
Groundwater quality; Subsurface dam

1. INTRODUCTION

A subsurface dam is a system to store groundwater by a "cut-off wall" (dam body) set up across a groundwater channel. It is similar to a "surface dam" in its function of water storage by a dam body, but in contrast to a surface dam that stores surface water (river water), a subsurface dam stores groundwater. Also, since groundwater is stored in geological strata, a subsurface dam artificially recharges natural aquifers [9]. Half the number of subsurface dams now known to be present in the world are located on the African continent, and a large portion of these dams are located in semi-arid regions [8].

In Japan too, a lot of subsurface dams have been constructed so far and these are located in islands and isolated peninsular regions [5]. However, the quality of the natural water supplied by subsurface dams depends critically on the chemical reactions undergoing in their environment, in particular, the chemical dissolution of bed rock/minerals (limestone). Rocks coming in contact with water constitute the major sources of dissolved species to natural water. They start the water-rock reaction and this reaction moves forward towards equilibrium by dissolving or leaching bedrock minerals in water. Solute concentration in the water is

proportional to the reactivity of bedrock minerals forming the catchments [26].

A recent acceleration of climate change of great concern to mankind is global warming: the increase in global temperatures due to man made emissions of greenhouse gases, in particular, carbon dioxide (CO₂), which has not only raised the ambient temperature but has also lowered the pH of water-bodies and changed precipitation intensities all over the world [20]. All of these factors affect reactions favoring dissolution of carbonaceous stones [22]. Several numerical and experimental studies of limestone dissolution have been carried out, taking this aspect into consideration. Reference [24] conducted a research on implications of future climate change for stone deterioration and said that chemical weathering process will be affected by changing atmospheric composition, increased temperatures, and alternations of rainfall amounts. Kump *et al.* (2000), [13] showed that higher concentrations of carbon dioxide in the future global atmosphere will further encourage mineral dissolution and higher air temperatures and more rainfall will favour chemical weathering in general. Sjöberg and Richard (1984), [19] studied temperature dependence of calcite dissolution kinetics in aqueous solutions with pH 2.4-8.4, from 1°C to 62°C. Noireil *et al.* (2005), [15] conducted an experiment on hydraulic properties and micro-geometry evolution accompanying limestone dissolution by acidic water. Liang *et al.* (1996), [14] investigated the dissolution kinetics at the calcite-water interface. Saharawat *et al.* (2011), [18] conducted a research and showed that at low pH recharged water Ca ion concentration was more than high pH of recharged water.

In this scenario of global warming and consequent acidification of water bodies, it has become very important to investigate the long term stability of subsurface dams related to sustainable groundwater development, as also the quality of groundwater stored in them that is affected by changes in climate through limestone dissolution. In other words the sustainability of such large scale economic and social investments has to be carefully assessed.

In our previous research [1], we studied dissolution of limestone due to change in pH, water velocity and temperature from reference values using a model for numerical simulation and assessed sustainability of groundwater development from underground dam area. In the present communication, we have used that numerical model developed by us to investigate the specific environmental impacts of greenhouse gas emission on groundwater quality around subsurface dams. In this study subsurface dam at

Miyako Island, where limestone is the underground geology, has been chosen. The objective of our research is to identify to what extent the enhancement of CO₂ emission due to fossil fuel burning affects pH, intensity of rain, temperature, the main factors causing dissolution of limestone, through simulation studies and then to investigate the water quality of groundwater surrounded by subsurface dam. Besides this, the results of numerical simulation studies of spatiotemporal distribution of Ca²⁺ ion concentration in the vicinity of the subsurface dam are presented and compared with our previous results [1].

2 THEORETICAL BACKGROUND

2.1 Seepage Analysis

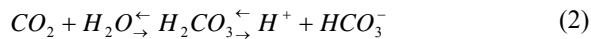
The permeability of all elements is estimated by the Kriging [11]. Based on the data, the groundwater flow analysis is carried out. Since the depth of the aquifer is smaller than the horizontal distance of the region, the quasi-three dimensional model was used for the analysis. The basic equation is written as (1).

$$D\lambda \frac{\partial h}{\partial t} = kD \left(\frac{\partial^2 h}{\partial x^2} + \frac{\partial^2 h}{\partial y^2} \right) + \varepsilon \quad (1)$$

where λ is the effective porosity, k is the permeability, D is the thickness of aquifer, h is the total head, and ε is the groundwater recharge. Equation (1) is solved by using the finite element method at an interval of 10 days.

2.2 Dissolution Chemistry

In CO₂-H₂O system, dissolved inorganic carbon exists in one of four major forms: CO_{2(aq)}, H₂CO₃, HCO₃⁻, and CO₃²⁻. The main chemical reactions involving these species are (1) the conversion of carbon dioxide into hydrogen and bicarbonate, (2) the dissociation of bicarbonate into hydrogen and carbonate ions, and (3) the dissolution of limestone. Since almost all of these reactions depend on the ambient pH and temperature, distribution or relative concentration of these species is also a function of pH and temperature. Dissolution of CO_{2(aq)} takes place according to the following reactions:



The equilibrium constants for (2) are:

$$(H_2CO_3) = 10^{-\log K_{CO_2}} \times P_{CO_2}$$

where, (H_2CO_3) = concentration of H₂CO₃, K_{CO_2} = Equilibrium constant for CO₂ dissolution, and P_{CO_2} = Pressure of CO₂, and

$$\frac{(HCO_3^-)(H^+)}{(H_2CO_3)} = \frac{(HCO_3^-)(H^+)}{10^{-\log K_{CO_2}} \times P_{CO_2}} = 10^{-\log K_{H_2CO_3}}$$

Hydrogen carbonate or bicarbonate ion dissociates as

$$HCO_3^- \rightleftharpoons H^+ + CO_3^{2-} \quad (3)$$

The equilibrium constant for (3) is:

$$(HCO_3^-) = \frac{10^{-\log K_{H_2CO_3} - \log K_{CO_2}} \times P_{CO_2}}{(H^+)}$$

$$-\log (HCO_3^-) = -(\log K_{H_2CO_3} + \log K_{CO_2}) - \log P_{CO_2} - \text{pH}$$

$$-\log (CO_3^{2-}) = -(\log K_{CO_2} + \log K_{H_2CO_3} + \log K_{HCO_3^-}) - \log P_{CO_2} - 2\text{pH}$$

$$\text{pH} = \frac{r}{r_{\max}} (\text{pH}_r - \text{pH}_0) + \text{pH}_0 \quad (4)$$

where pH_r is the pH in rain, pH₀ is assumed to be 8, r_{\max} is the maximum rain data in the duration, and r is the rain data at each time step. pH in groundwater is assumed to be 8. We assume pH in rainwater is small, but when rainwater seeps into the ground, it changes the groundwater pH and this has a considerable effect on dissolution of CaCO₃. After this dissolution, groundwater pH quickly returns to 8. The chemical composition of the bedrock tends to stabilize (buffer) the pH of the groundwater. The longer the contact time, the larger the effect of the rock chemistry on the composition and pH of the groundwater. Actually groundwater pH is from 7 to 8, so we assume groundwater pH is 8. The treatment of pH in the groundwater is introduced to reduce the large effects of pH in the chemical reactions.

Dissolution of calcium carbonate (limestone) is expressed by the equation which is



The equilibrium constant for (5) is

$$(Ca^{2+}) = \frac{10^{-\log K_{CaCO_3}}}{(CO_3^{2-})}$$

$$(Ca^{2+})(CO_3^{2-}) = 10^{-\log K_{CaCO_3}}$$

The equilibrium constants are calculated by those equations dependent on temperature, which have been suggested by [17] as:

$$\log K_{CO_2} = 108.3865 + 0.01985076T - 6919.53/T$$

$$- 40.45154 \log T + 669365/T^2$$

$$\log K_{H_2CO_3} = -356.3094 - 0.06091964T + 21834.37/T$$

$$+ 126.8339 \log T - 1684915/T^2$$

$$\log K_{HCO_3^-} = -107.8871 - 0.03252849T + 5151.79/T$$

$$+ 38.9256 \log T - 563713.9/T^2$$

$$\log K_{CaCO_3} = -171.9065 - 0.077993T + 2839.319/T$$

$$+ 71.595 \log T$$

where T is absolute temperature (K), which is assumed to be the same as actual temperature.

2.3 Transport Analysis

The governing equation is used to describe concentration distributions in the rocks. The transport equations include the effects of advection and hydrodynamic dispersion.

$$\frac{\partial C}{\partial t} = (Cv_i)_{,i} + (D_{ij}C_{,j})_{,i} + R = 0 \quad (6)$$

where C is the concentration of the components (solutes) which are Ca^{2+} and CO_3^{2-} , v_i is the Darcy velocity vector, D_{ij} is the hydrodynamic dispersion coefficient tensor, and R is the rate of dissolution of components. For solving the transport Eq. (6), firstly we need the R value of CO_3^{2-} , in this case we have got this value from Eqs. (3) and (4) for every time steps, where Eq. (3) is a function of both pH and temperature. Then we have got the distribution of CO_3^{2-} by solving transport equation. Again, for calculating of Ca^{2+} , we used the solved CO_3^{2-} values and put in the Eq. (5) and we have got the R value of Ca^{2+} and then by using Eq. (6) finally we got the distribution of Ca^{2+} . Summation convention is applied in (6). D_{ij} is the following relations with velocity:

$$\left. \begin{aligned} D_{xx} &= \frac{\epsilon_L v_x^2}{|v|} + \frac{\epsilon_T v_y^2}{|v|} + D_0 \\ D_{yy} &= \frac{\epsilon_L v_y^2}{|v|} + \frac{\epsilon_T v_x^2}{|v|} + D_0 \\ D_{xy} &= D_{yx} = (\epsilon_L - \epsilon_T) \frac{v_x v_y}{|v|} \end{aligned} \right\} \quad (7)$$

where ϵ_L and ϵ_T are longitudinal and transverse dispersivities, respectively. In this analysis, $\epsilon_L = 1.0$ m and $\epsilon_T = 0.1$ m are assumed because dispersivities are from 10^{-2} to 1 cm for laboratory experiments and range from 10^{-1} to 10^2 m for tracer test in the more heterogeneous porous materials typically encountered in the field [12]. D_0 is ignored in respect of high velocity. Equation (6) is multiplied by the thickness of strata and solved by upstream FEM.

3. Numerical Conditions of Chemical Transport

The region analyzed is around the subsurface dam at Miyakojima Island in Japan, shown in Fig. 1, while Fig. 2 shows the actual rainfall and temperature data, averaged for every 10-day over the site, taken over a period of 450 days from March, 1994 to May, 1995. The region is about 2 km from east to west and about 3 km from south to north and was divided into 1,680 elements with 1,764 nodes. Three points are chosen as representative points of actual pumping area for analysis: while A and C are points near the dam body and are chosen for variations over a short length scale, B is chosen away from the dam body and upstream. The same finite element mesh is used for seepage and transport analysis.

3.1 Ranges of Variation of the Parameters

As a reference case, it is assumed that pH of recharge water (rainwater) is 5. The initial groundwater pH is assumed to be

8 and initial temperature is 18°C. Change of pH in groundwater due to rain is governed by (4) and pressure of carbon dioxide is assumed to be 0.003 atm.

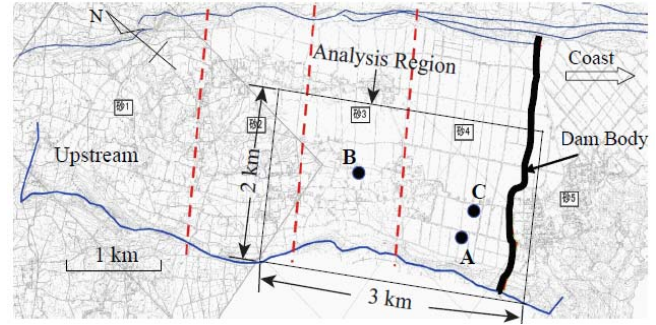


Fig. 1 Region analyzed

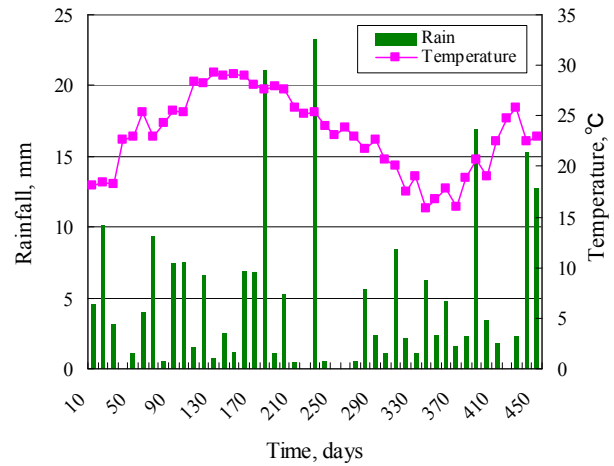


Fig. 2 10-day averaged rainfall and temperature

Generally the pH of acid rain is less than 5.6 and can go down to values as low as 2.1 [2], [16], [25], [27], [3]. To have a better evaluation of the effect of acidity of the water on dissolution of limestone, the pH is set at 4 and 6, assuming the variance of pH values to be about 1. Again, Frank *et al.* (2007), [4] reported that more rain occurred from the increase in surface temperatures due to global warming. United States Environmental Protection Agency (2000), [23] has also correlated more precipitation with global warming. In this study, variations of 10 percent rainfall above and below normal are taken for checking the effect of rain intensity on limestone dissolution. Of course, the direct effect of global warming, i.e., the effect of increase in average daily temperature on limestone dissolution was also assayed. Global temperature increment for next century has been estimated to vary from 1.1°C to 6.4°C [6], [10], [7]. In this study, the increase of temperature is assumed to be 1°C and temperature in the entire region is changed with every 10-day average temperature to examine the effect. To get a fuller range of variation, examination is also carried out with 1°C

decrease in temperature from every 10-day average temperature.

4. Results and Discussion

4.1 Effect of pH

Table 1 summarizes the effects of pH, rainfall intensity and temperature on concentration of Ca^{2+} at 450 days. Over these days, Ca^{2+} concentrations increased 4863.68 percent with the decrease of one unit pH and decreased 93.46 percent with increase of one unit, respectively from the reference value. Fig. 3 shows the spatial distribution of Ca^{2+} concentration at 450 days after changing the pH to 6. Dissolution of limestone decreases on increasing the pH and the highest concentration reaches 0.27 mol/L, while the opposite result is obtained on decreasing the pH and in this case the highest concentration increases up to 205.14 mol/L. Variations of concentration with time at the sample points for pH=4 and 6 are shown in Fig. 4. It shows that the concentration starts going up after 180 days indicating the onset of acid rain, and continues increasing up to 450 days. Concentrations are highest at A (0.13 mol/L for pH=6 and 76.70 mol/L for pH= 4) and lowest at B (0.07 mol/L for pH=6 and 29.80 mol/L for pH= 4) and at C it has intermediate values. As Ca^{2+} increases with acidification, it will accumulate on the pumping well screen and block the filter severely. Excess of Ca^{2+} will deteriorate the quality of groundwater and it can definitely be concluded that the change in pH has a large effect on the operation of subsurface dam.

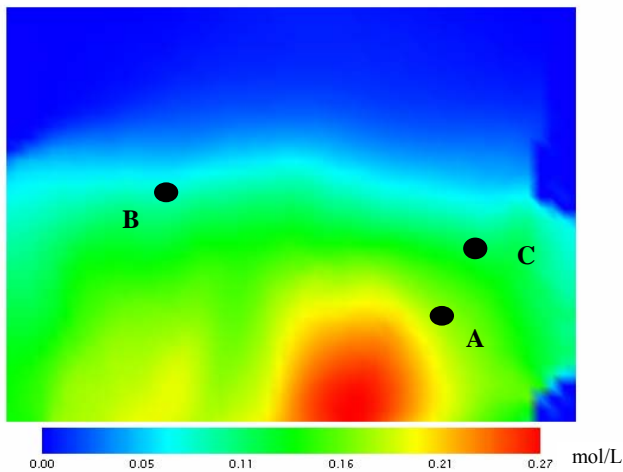


Fig. 3 Distribution of Ca^{2+} (450 days) for pH= 6

4.2 Effect of Rainfall Intensity

Fig. 5 shows the distribution of Ca^{2+} concentration in 450 days for 10 percent less rainfall from reference case. The results show that the decrease of intensity of rainfall, which coincides with the decrease of velocity, caused the decrease in the dissolution of limestone. For comparison, concentrations of Ca^{2+} with time, at the sample points for 10 percent less and more rainfall, are shown in Fig. 6. The figure also shows that the concentrations are highest at point A (1.72 mol/L and 2.61 mol/L for less and more rainfall, respectively)

and lowest at point B (0.79 mol/L and 1.27 mol/L for less and more rainfall, respectively) and at point C concentrations are 1.06 mol/L for less rainfall and 1.72 mol/L for more rainfall.

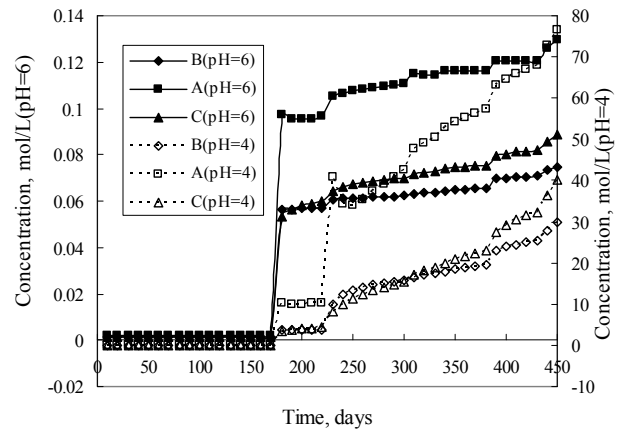


Fig. 4 Concentrations of Ca^{2+} with time for sample points for pH=6 and pH=4

Table 1 Change of Ca^{2+} concentrations with climatic changes

Environmental Parameters		Ca^{2+} concentrations (mol/L)	% change of Ca^{2+} concentrations from reference
pH	Reference pH=5	4.13 [§]	-
	pH=4	205.16 [§]	4863.68
	pH=6	0.27	- 93.46
Rainfall	Reference (10 days average)	4.13 [§]	-
	10 percent increased	5.41 [§]	30.99
	10 percent decreased	4.25	-21.38
Temperature	Reference (10 days average)	4.13 [§]	-
	1°C increased	4.06 [§]	- 1.69
	1°C decreased	4.20	1.69

[§]data from [1]

Ca^{2+} concentration is 21.38 percent decreased due to 10 percent less rainfall and Ca^{2+} concentration is 30 percent increased due to 10 percent more rainfall, respectively from reference (Table 1). In this case, Equation (4) has no effect, because rainfall is increased 10 percent both in the maximum rain data in the duration and in the rain data at each time step..

4.3 Effect of Temperature

The distribution of Ca^{2+} at 450 days after decreasing temperature by 1°C is shown in Fig. 7. The results show that the highest concentration is 4.20 mol/L. Fig. 7 also shows that the dissolution of limestone increases very little from

reference with temperature decrease. Ca^{2+} concentrations are found to increase 1.69 percent with 1°C decrease in temperature while the same percentage decreased with 1°C increase in temperature, respectively from reference (Table 1).

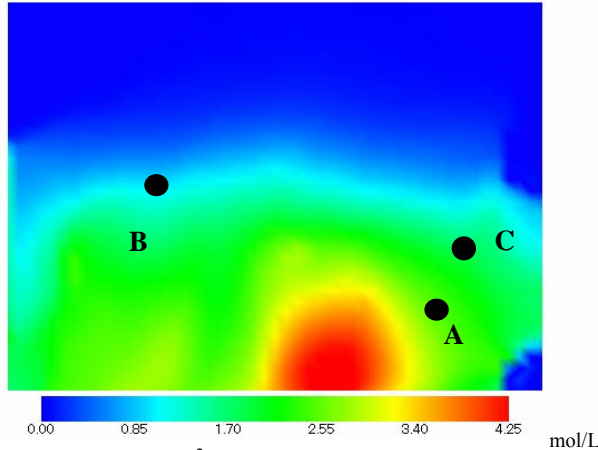


Fig. 5 Distribution of Ca^{2+} (450 days) for 10 percent less rain

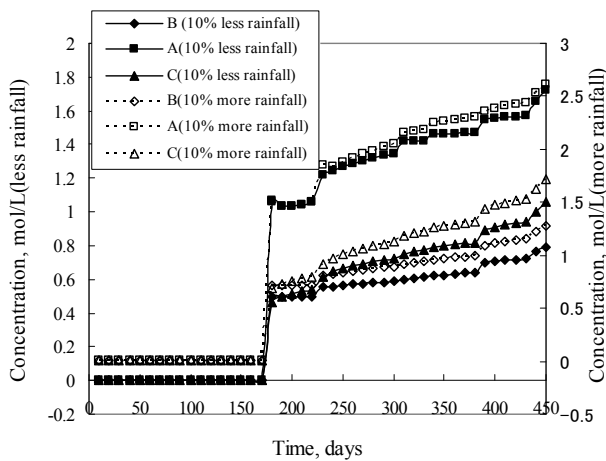


Fig. 6 Concentrations of Ca^{2+} with time for sample points for changed rainfall

The variations in concentration of Ca^{2+} with time for the sample points at decreased and increased temperatures are shown in Fig. 8. The figure also shows that the concentrations are highest at point A (1.70 mol/L and 1.65 mol/L for decreased and for increased temperature, respectively) and lowest at point B (0.78 mol/L and 0.76 mol/L for decreased and for increased temperature, respectively) and at point C concentration is 1.05 mol/L for decreased temperature and 1.01 mol/L for increased temperature.

In this case, dissolution depends on the equilibrium constant of the chemical reactions and those constants are only dependent on temperature. Changing the temperature causes a small change in the equilibrium constants of dissolution reactions and this is the reason behind the small change

observed [1]. Due to 1°C decrease in temperature, the equilibrium constants K_{CO_2} increases by 2.12 percent and K_{CaCO_3} increases by 1.20 percent, while the equilibrium constants $K_{\text{H}_2\text{CO}_3}$ and $K_{\text{HCO}_3^-}$ decrease by 1.37 and 2.13 percents, respectively. Temperature effect on dissolution is very small compared to other parameters like pH of rainwater and rainfall intensity.

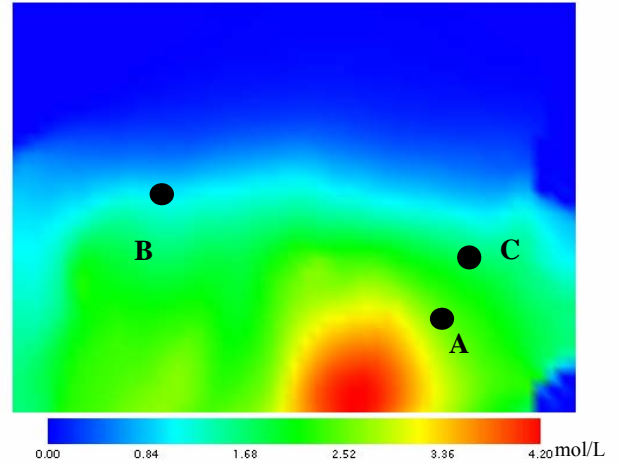


Fig. 7 Distribution of Ca^{2+} (450 days) for 1°C decrease in temperature

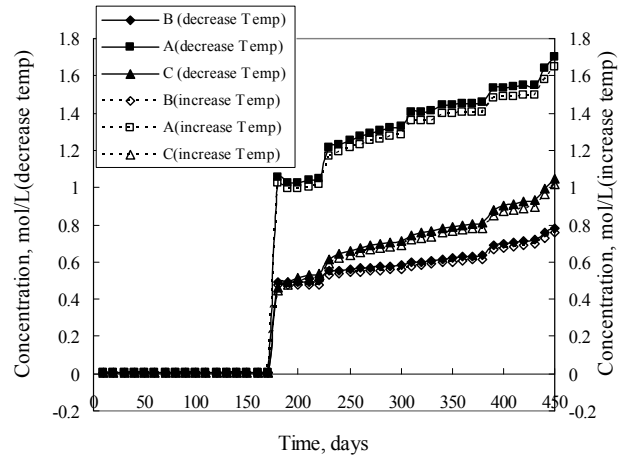


Fig. 8 Concentrations of Ca^{2+} with time for sample points for changed temperature

5. Conclusions

The effect of climatic change on groundwater quality around the subsurface dam area was observed using numerical simulation. From the results the following conclusions can be arrived at.

1. The dissolution of limestone is proportional to the acidification of rainwater i.e. inversely proportional to the pH of rainwater.
2. Dissolution of limestone is proportional to the increase of intensity of rain that leads to the increase of velocity of

water. Dissolution of limestone is inversely proportional to the temperature but this dependence is weak.

The enhanced dissolution of calcium ions is expected to block the filter of the pumping well and deteriorate the quality of groundwater as well. Hence, the general conclusion of this research is that the quality of groundwater from subsurface dam area depends on several environmental factors. Among the factors that are included in this research, pH of water in contact has the maximum effect on the dissolution of limestone. Increasing emission of CO₂ in terrestrial atmosphere due to fossil fuel burning and industrial activity causes global warming as well as acidification of water bodies. Any reduction in CO₂ emission would go to reduce dissolution of limestone and, consequently, subsurface dams can operate smoothly over a long term.

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International Journal of GEOMATE , Oct. 2011, Vol. 1, No.1 (Sl. No. 1)
MS No. 1m received September 15, 2011, and reviewed under
GEOMATE publication policies.

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