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Scoping calculations for the Matrix Diffusion Experiment

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November 1993

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SCOPING CALCULATIONS FOR THE MATRIX DIFFUSION EXPERIMENT

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Abstract

The migration of radionuclides in the rock is envisioned to take place in individual permeable fractures forming a fracture network. One important entity that will influence the migration rate for the radionuclides is the surface area in contact with flowing water which is available for sorption and diffusion into the rock matrix. This area is often referred to as the "flow wetted surface" and has not been accurately determined in earlier tracer experiments. Furthermore, the migration from the water in a fracture, through the fracture filling material and into the pore system within the rock matrix has a significant influence on the migration rates for radionuclides. It is therefore important to demonstrate that this effect takes place in natural fractures.

The main objectives with the proposed Matrix Diffusion Experiment are

- to demonstrate the significance of matrix diffusion as retardation mechanism for transport of sorbing tracers in a natural fracture
- to determine the flow path geometry and the flow wetted surface in a single fracture plane
- to determine in-situ values of diffusion and sorption coefficients

The scoping calculations presented in this report include practical considerations and tracer migration calculations relevant for the proposed Matrix Diffusion Experiment.A mixture of tracers having different sorption capacities should be injected simultaneously. These tracers should have sorption capacities, $K_d\rho_p$ values, ranging from, at least, 1 m^3/m^3 to 10000 m^3/m^3. Most of the injected tracers should however have sorption capacities in the range $K_d\rho_p$=1 to 100 m^3/m^3, since the concentration profile into the rock matrix can not be determined for stronger sorbing tracers. The tracers should have linear sorption isotherms.

The report also includes some practical considerations. Different injection methods and the need for sampling holes in the vicinity of the injection hole has been discussed. We prefer if the tracers were injected using a constant flowrate method. This has significant advantages compared to the other considered methods when the experiment is evaluated. Furthermore, we do not at present think that it is motivated to have sampling holes in the fracture plane. The cost associated with these holes should instead be used to investigate more fractures. There are however some advantages with sampling holes in the fracture plane so the discussion will certainly continue and we might also change our opinion.

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APPENDIX

1 INTRODUCTION

The migration of radionuclides in the rock is envisioned to take place in individual permeable fractures forming a fracture network. One important entity that will influence the migration rate for the radionuclides is the surface area in contact with flowing water which is available for sorption and diffusion into the rock matrix. This area is often referred to as the "flow wetted surface" and has not been accurately determined in earlier tracer experiments. Furthermore, the migration from the water in a fracture, through the fracture filling material and into the pore system within the rock matrix has a significant influence on the migration rates for radionuclides. It is therefore important to demonstrate that this effect takes place in natural fractures.

Experiments aiming at investigating the diffusion of tracers into the rock matrix have been performed in Stripa. The "In-situ Diffusion Experiment" [Birgersson et al, 1988] clearly showed that non-sorbing tracers were able to diffuse cm´s-dm´s into the rock matrix. The tracers were however injected from boreholes into more or less unfractured rock giving somewhat different conditions compared to escaping radionuclides. In another experiment, sorbing tracers were injected into a fracture plane that later was excavated in the "Stripa 2-D Experiment" [Abelin et al, 1985]. Some evidence that these sorbing tracers could migrate from the fracture and into the rock matrix was obtained, but the time for this experiment was quite short, giving short penetration depths into the matrix. Another complicating factor was the high and varying background concentrations in the rock of the used tracers.

The main objectives with the proposed Matrix Diffusion Experiment are

- to demonstrate the significance of matrix diffusion as retardation mechanism for transport of sorbing tracers in a natural fracture
- to determine the flow path geometry and the flow wetted surface in a single fracture plane
- to determine in-situ values of diffusion and sorption coefficients

The scoping calculations presented in this report include practical considerations and tracer migration calculations relevant for the proposed Matrix Diffusion Experiment. The main objectives of the scoping calculations have been to:

- propose an experimental layout and suitable sorption properties for the tracers to be used
- discuss how the results from the experiment can be used for determination of parameters controlling transport of sorbing tracers in single fractures

2 SHORT PRESENTATION OF THE MATRIX DIFFUSION EXPERIMENT

The proposed Matrix Diffusion Experiment is more or less a combination of two Stripa experiments, the "In-situ Diffusion Experiment" and the "2-D Experiment". A mixture of tracers with different sorption capacities will be injected into a fracture that a long time later will be excavated and examined for tracers on the surfaces as well as in the rock matrix. A detailed characterization of the tracer abundance on the fracture surfaces is envisioned to give information on the flowpaths (channel pattern) and the migration distance for the tracers. The subsequent sampling of the rock matrix is envisioned to provide information on matrix diffusion effects.

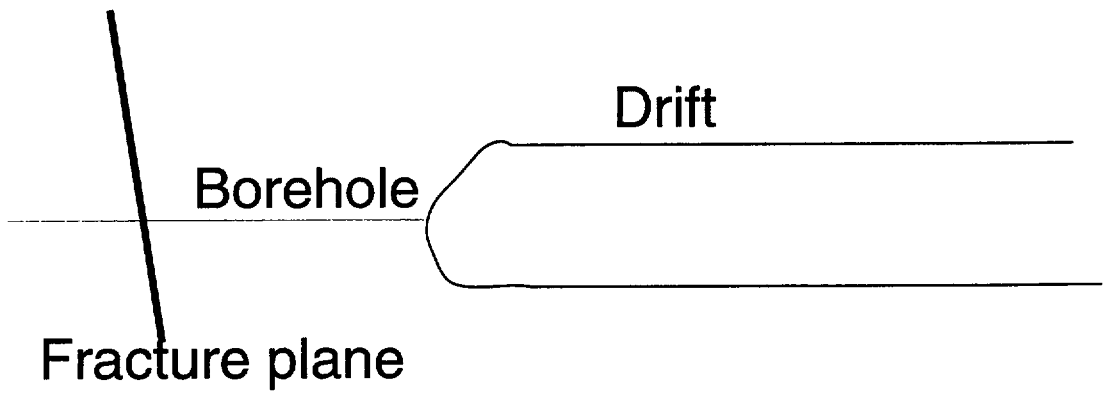

Figure 1. Schematic experimental layout.

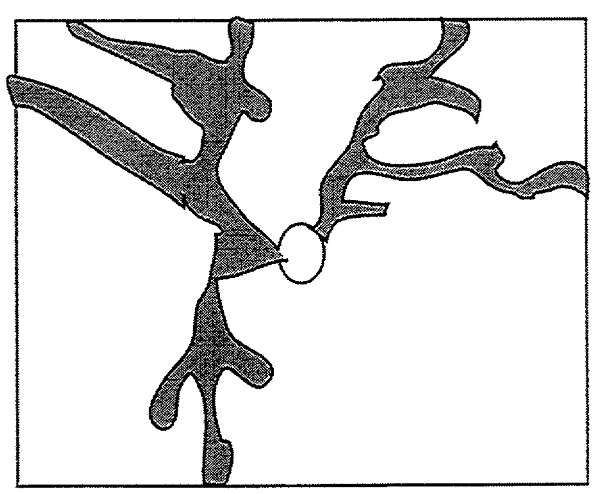

Figure 2. Possible flowpath pattern within the fracture plane.

3 PERFORMED CALCULATIONS

The flow geometry is unknown and will be one of the entities that will be determined from the experiment. The calculations have been made with two geometrically simple "extreme" cases on the flow geometry and one "realistic" description using KTH/KAT´s stochastic variable aperture distribution model. The following flow geometries have been used in the scoping calculations

- channelized linear flow (flow along a single channel with narrow and constant width)
- radial flow (flow within the entire fracture plane)
- KTH/KAT´s variable aperture distribution model

Schematic layouts of the flow geometries are given in Figure 3.

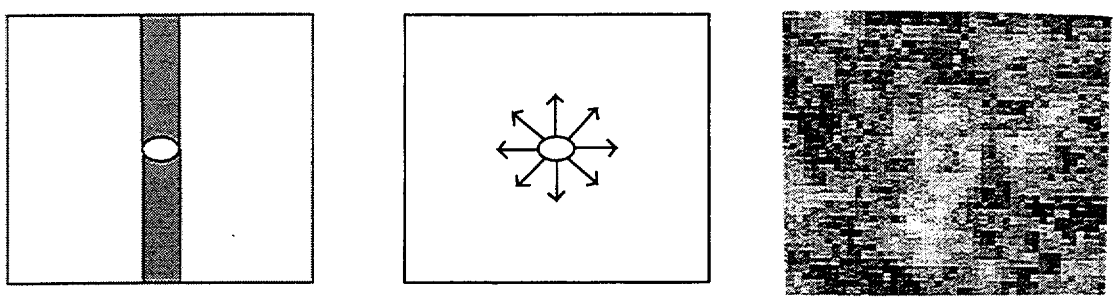

Figure 3. Considered geometries. Linear flow, radial flow and the variable aperture distribution model.

The migration calculations along the flowpaths as well as into the rock matrix has for the cases of linear and radial geometry been calculated as functions of

- sorption capacity ($K_d\rho_p$)
- effective diffusivity (D_e)
- fracture aperture
- channel width (linear flow)
- injection flowrate (Q)
- time

The calculations have been performed using a "best estimate" for each of the parameters. The impact of changing the different parameter values has then been studied by changing one parameter at a time a factor 10 up and down. The value of $K_d\rho_p$ has however been changed a factor 100 up and down in order to account for a wide range of sorbing tracers. The "best estimates" of the parameter values were:

	Best estimate	Variations
D_e	$2*10^{-13}$ m²/s	$2*10^{-14}$, $2*10^{-13}$, $2*10^{-12}$ m^2/s
Aperture	10^{-4} m	10^{-5}, 10^{-4}, 10^{-3} m

and in the calculations for the linear symmetry:

Channel width	76 mm	7.6, 76, 760 mm

It could be noted that the effective diffusivity (D_e) in most cases does not depend on the sorption properties of a tracer but rather on the size, shape and molecular weight of the tracer. Apart from the parameters above, some other parameters have to be included in the calculations. These parameters depend on the experimental conditions and the sorption properties of the chosen tracers.

	Base case	Variations
$K_d\rho_p$	100 m^3/m^3	1, 100, 10000 m^3/m^3
Flowrate	1 ml/h	0.1, 1, 10 ml/h
time	1 year	1, 5 years

$K_d\rho_p$ is equal to the porosity, ε_p, (approx. 0.005) for a non-sorbing tracer and the chosen values on $K_d\rho_p$ (1, 100, 10000) correspond to tracers that are weakly, medium and strongly sorbing. Sr and Cs have been used in other tracer experiments and have $K_d\rho_p$ values of about 40 and 400 respectively [SKB-91]. The sorption capacities as well as the effective diffusivities will depend on the chosen tracers,the properties of the Äspö granite and the Äspö groundwater chemistry.

The approach for the variable aperture distribution model is somewhat different. The 6 m x 6 m fracture plane has in this model been divided into 100x100 squares all having apertures stochastically generated from a log-normal distribution. A standard deviation of σ=1 and a correlation length of 0.3 m (5 squares) was used for the generation of apertures.

3.1 LINEAR FLOW

Figure 4 gives the conceptual model for the linear flow case.

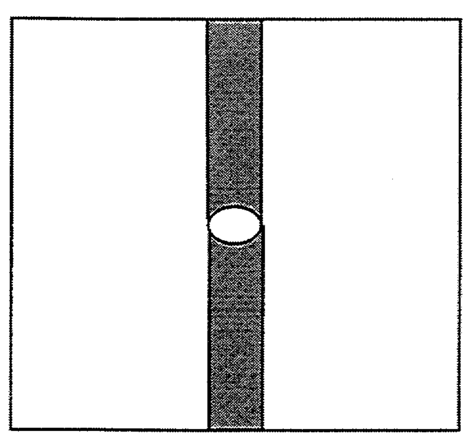

Figure 4. Linear flow. Fracture plane, injection hole and flowpath.

The migration of the tracer solution is along the flowpath that is intersected by the injection hole. The width of the flowpath is in the base case assumed to be the same as the diameter of the injection borehole (76 mm). The width and aperture of the flowpath is constant.

3.1.1 Migration along the fracture (channel)

The concentration profile along a channel having constant aperture and width can be calculated using Equation 1. Dispersion due to advection is omitted in the equation. Otherwise it is the "exact" solution for the assumption made (Moreno and Neretnieks, 1991).

$$\frac{C}{C_0} = erfc\left[\frac{xW}{Q}\sqrt{\frac{K_d\,\rho_p\,D_e}{t-t_wR_a}}\right] \qquad \text{Eq.(1)}$$

where x is the distance [m] from the injection section, W the channel width [m], Q the flowrate [m^3/s], $K_d\rho_p$ the sorption coefficient [m^3/m^3], D_e the effective diffusivity [m^2/s], t the time [s], t_w the water residence time [s] and R_a the surface retardation factor. C_0 is the injection concentration [kg/m^3] and C is the concentration in the water in the fracture plane at distance x [m] from the injection hole at time t [s].

The first part of the argument in Equation 1:

$$\frac{xW}{Q}$$

is the fracture surface (what we call the flow wetted surface) in contact with the flowing water divided with the water flow rate. The second part of the argument in the equation:

$$\sqrt{\frac{K_d\,\rho_p\,D_e}{t-t_wR_a}}$$

includes the sorption and diffusion coefficients as well as the time for the experiment.

The concentration profile along the fracture plane will be strongly influenced by the parameters included in the first part of the equation (x, W and Q) since they are outside the square root. The parameters within the square root ($K_d\rho_p$, D_e, t, t_w and R_a) have a much smaller impact on the concentration profile. Note that R_a has been assigned the value 1 in the examples below, i.e. surface sorption has been excluded. Furthermore, it is obvious from Equation 1 that a variation in for example W will give the same concentration profile as a variation in Q and that changing Q or W a factor 10 is the same as changing $K_d\rho_p$ or D_e or t a factor 100. Nevertheless, variations in all parameters are illustrated in the figures below.

Migration along the flow path - parameter variations

Figures 5 and 6 show the concentration profiles along the fracture planes after an injection time of 1 and 5 years respectively. The three curves in each figure correspond to $K_d\rho_p$=1, 100 and 10000 m^3/m^3.

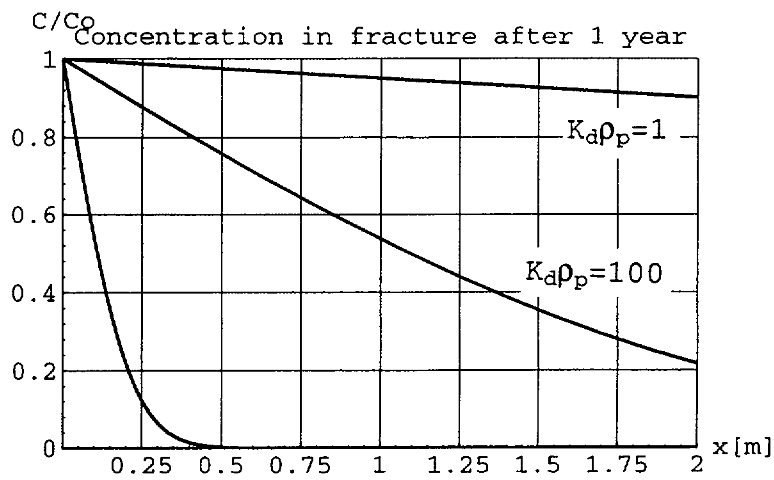

Figure 5. C/C_o as function of distance from the injection location and $K_d\rho_p$. Time =1 year and $K_d\rho_p$=1, 100 10000 m^3/m^3. (W=76 mm, Q=1 ml/h, $D_e=2*10^{-13}$ m^2/s and R_a=1)

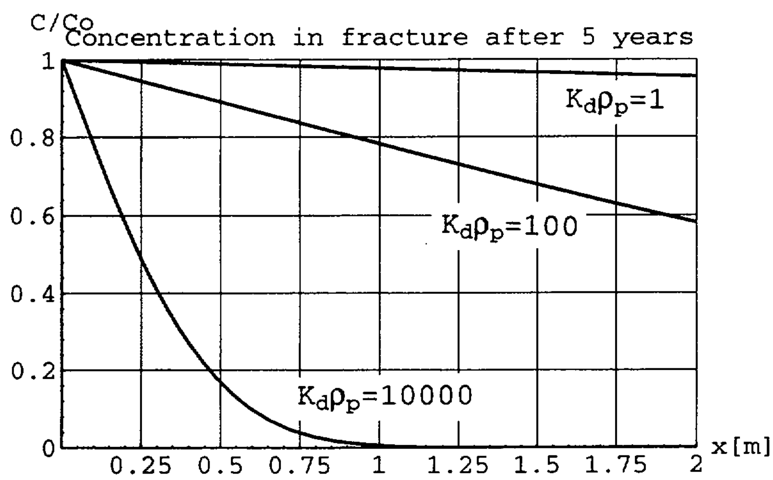

Figure 6. C/C_o as function of distance from the injection location and $K_d\rho_p$. Time =5 years and $K_d\rho_p$=1, 100,100000 m^3/m^3. (W=76 mm, Q=1 ml/h, $D_e=2*10^{-13}$ m^2/s and R_a=1)

Figures 5 and 6 show that $K_d\rho_p$ can not be less than about 10000 if all injected tracers are to be found within the square meter(s) of the fracture plane that will excavated. A $K_d\rho_p$ of 1 will result in a high and almost uniform tracer concentration in the excavated fracture plane resulting in only a small fraction of the injected tracers are expected to be found within the excavated volume. Figures 7 to 10 illustrate the impact on varying W, Q, the fracture aperture and D_e.

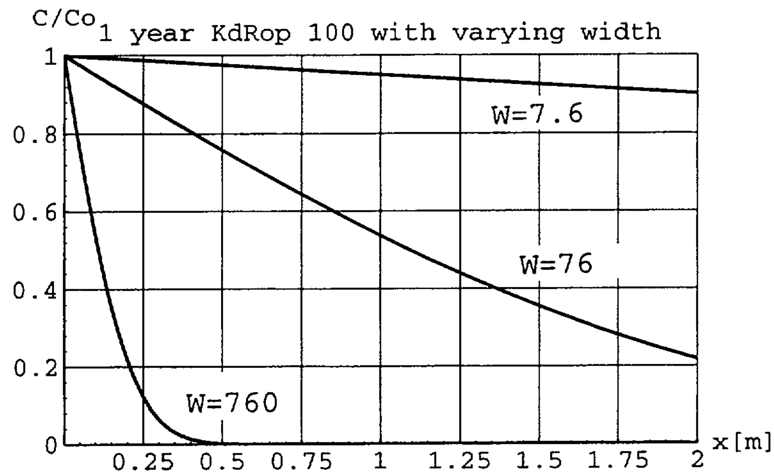

Figure 7. C/C_o as function of distance from the injection location and W. W=7.6, 76, 760 mm. ($K_d\rho_p$=100 m^3/m^3, Q=1 ml/h, D_e=2*10^{-13} m^2/s, time=1 year, fracture aperture=0.1 mm and R_a=1)

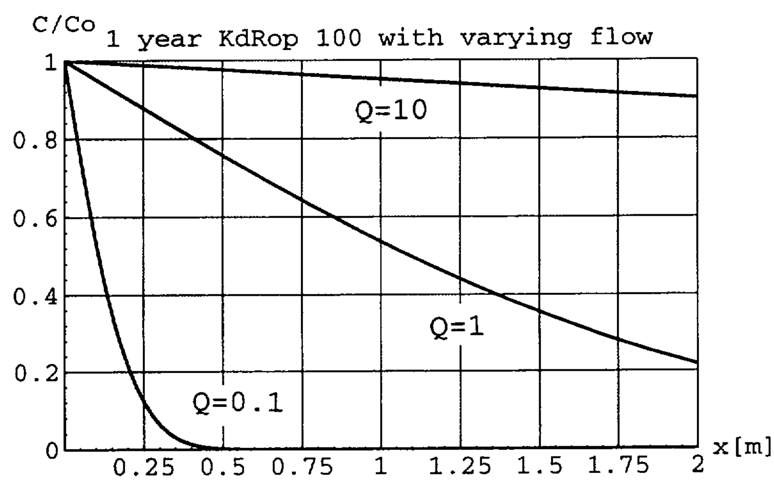

Figure 8. C/C_o as function of distance from the injection location and Q. Q=0.1, 1, 10 ml/h. ($K_d\rho_p$=100 m^3/m^3, W=76 mm, D_e=2*10^{-13} m^2/s, time=1 year, fracture aperture=0.1 mm and R_a=1)

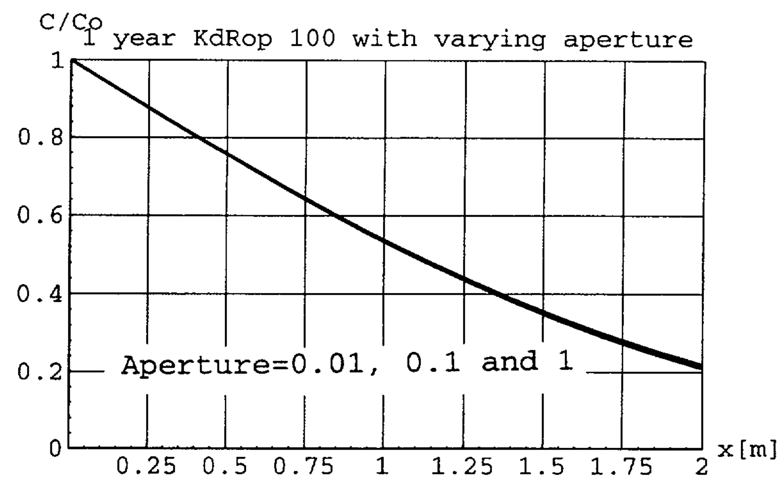

Figure 9. C/C_0 as function of distance from the injection location and fracture aperture. Fracture aperture=0.01, 0.1, 1 mm. ($K_d\rho_p$=100 m^3/m^3, W=76 mm, Q=1 ml/h, $D_e=2*10^{-13}$ m^2/s, time=1 year and R_a=1)

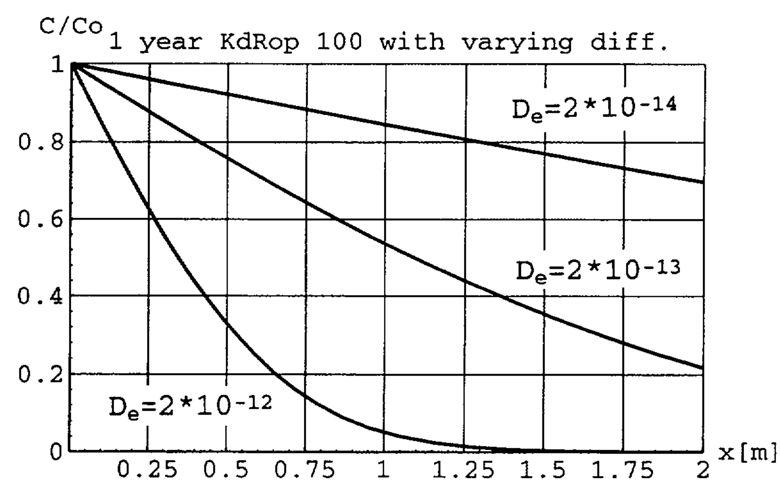

Figure 10. C/C_0 as function of distance from the injection location and D_e. $D_e=2*10^{-14}$, $2*10^{-13}$, $2*10^{-12}$ m^2/s. ($K_d\rho_p$=100 m^3/m^3, W=76 mm, Q=1 ml/h,fracture aperture=0.1 mm, time=1 year and R_a=1)

The impact of the fracture aperture is very small since a change will only influence the water travel time (t_w) that still will be several orders of magnitude lower than the time of interest for the sorbing tracer (t), see Equation 1.

3.1.2 Diffusion into the rock matrix

The analytical solution for the concentration profile into the rock matrix at different distances from the injection hole is given in Equation 2 (Gureghian, 1990).

$$\frac{C}{C_0} = erfc\left[\frac{\frac{2xW}{Q}\sqrt{K_d\rho_p D_e} + z\sqrt{\frac{K_d\rho_p}{D_e}}}{2\sqrt{t-t_w R_a}}\right] \quad \text{Eq. 2}$$

where z is the distance into the rock matrix [m] from the surface of the fracture.

The concentration profile into the rock matrix will be different at different distances from the injection hole since the concentration in the fracture plane is a function of distance from the injection hole and time. The concentration profile into the rock matrix has therefore been calculated using Equation 2 at 0.2, 0.4, 1 and 2 m distance from the injection hole. Samples taken 0.2 m from the injection hole have in this context been deemed to be fairly undisturbed due to mechanical and rock stress effects caused by the presence of the hole.

It should be noted that C in equation 2 denotes the concentration in the pore water in the rock matrix. The concentration in the rock is $K_d\rho_p$ times higher for a sorbing specie.

Considering Equation 2, it is rather difficult to see the impact of the effective diffusivity, sorption capacity and time on the concentration profile into the rock matrix. To illustrate the impact of these parameters one can consider the case where the tracer concentration in the fracture plane is constant, C_0, with time. This is almost true for a weakly sorbing tracer a short distance from the injection hole since the advective transport is then fast enough to achieve an almost constant concentration in the fracture plane. The concentration profile would in this case look like curve I in Figure 11. The area under curve I is the same as under rectangle II giving that rectangle II gives a value on how far into the rock matrix the tracer has migrated. This distance is called the penetration depth, η, and is calculated using the following expression (Neretnieks, 1979)

$$\eta = \frac{2}{\sqrt{\pi}}\sqrt{\frac{D_e t}{K_d\rho_p}} \quad \text{Eq. 3}$$

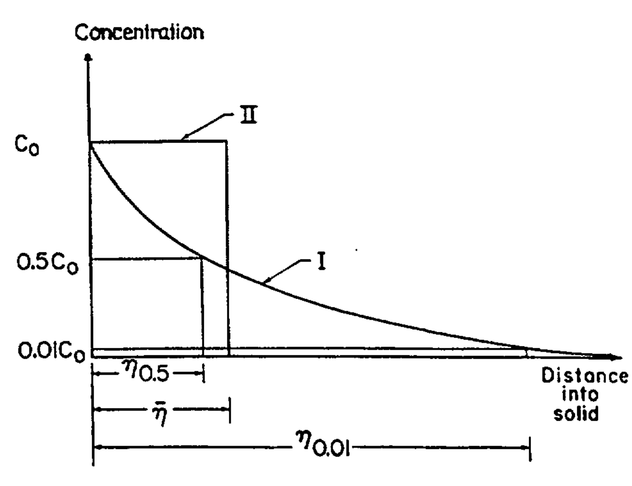

Figure 11. Diffusion profile into the rock matrix for a constant concentration in the fracture plane.

Diffusion into the rock matrix is a very slow process for a sorbing tracer. It can be seen in Equation 3 that a tracer with a sorption coefficient of $K_d\rho_p$=10000 will require 10000 times longer time to obtain the same concentration profile into the rock matrix as a tracer having $K_d\rho_p$=1.

The concentration profiles into the rock matrix have been calculated at 0.2, 0.4, 1 and 2 m from the injection hole using the same parameter numbers as for the previous calculations of the concentration profiles along the fracture plane.

	Base case	**Variations**
D_e	$2*10^{-13}$ m^2/s	$2*10^{-14}$, $2*10^{-13}$, $2*10^{-12}$ m^2/s
$K_d\rho_p$	100 m^3/m^3	1, 100, 10000 m^3/m^3
Aperture	10^{-4} m	10^{-5}, 10^{-4}, 10^{-3} m
Flowrate	1 ml/h	0.1, 1, 10 ml/h
time	1 year	1, 5 years
Width	76 mm	7.6, 76, 760 mm

Diffusion into the rock matrix - parameter variation

Figures 12 and 13 shows the concentration profiles into the rock matrix 0.2 m from the injection hole after an injection time of 1 and 5 years respectively. The curves in each figure corresponds to $K_d\rho_p$=1, 100, 10000 m^3/m^3 respectively.

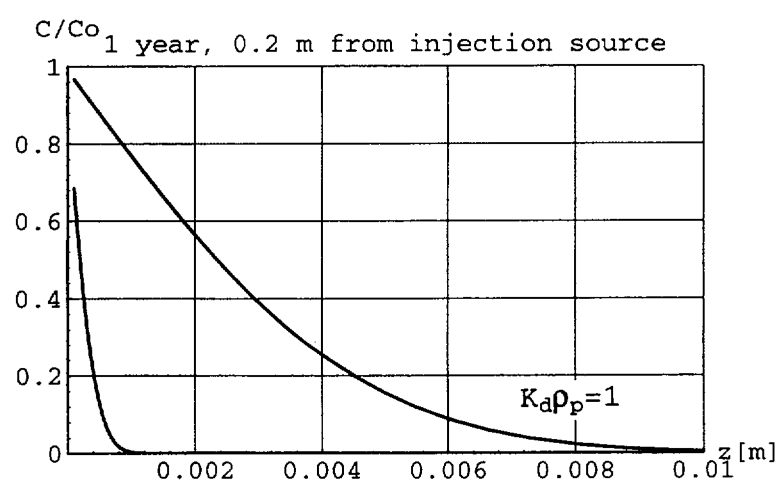

Figure 12. C/C_o into the rock matrix 0.2 m from the injection hole as function $K_d\rho_p$. Time=1 year and $K_d\rho_p$=1, 100 10000 m^3/m^3. (W=76 mm, Q=1 ml/h, D_e=$2*10^{-13}$ m^2/s and R_a=1)

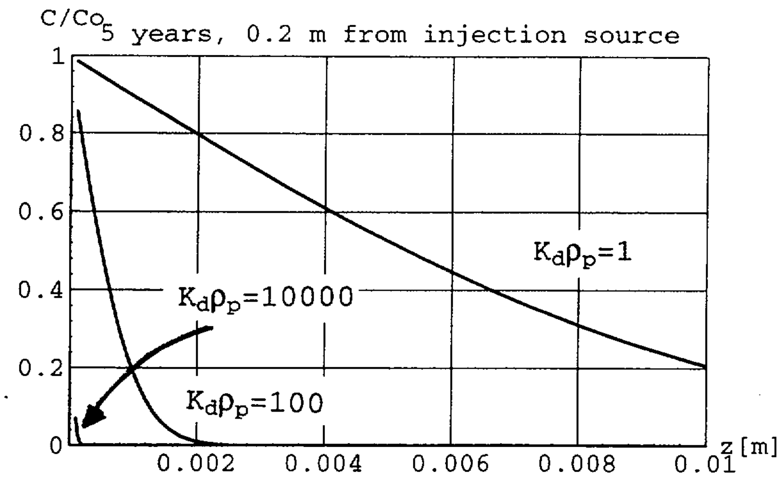

Figure 13. C/C_o into the rock matrix 0.2 m from the injection hole as function $K_d\rho_p$. Time=5 years and $K_d\rho_p$=1, 100 10000 m^3/m^3. (W=76 mm, Q=1 ml/h, $D_e=2*10^{-13}$ m^2/s and R_a=1)

It is from a practical point of view desirable if the tracers have migrated at least a few mm, preferably a cm, into the rock matrix. Figures 12 and 13 illustrate that this will be achieved for a tracer having a sorption coefficient of $K_d\rho_p$=1 m^3/m^3, while a tracer having $K_d\rho_p$=100 m^3/m^3 will not be found further in into the rock matrix than about 2 mm even if the experiment would continue for 5 years.

Figure 14 together with Figure 12 illustrate the impact of varying the sampling distance from the injection hole.

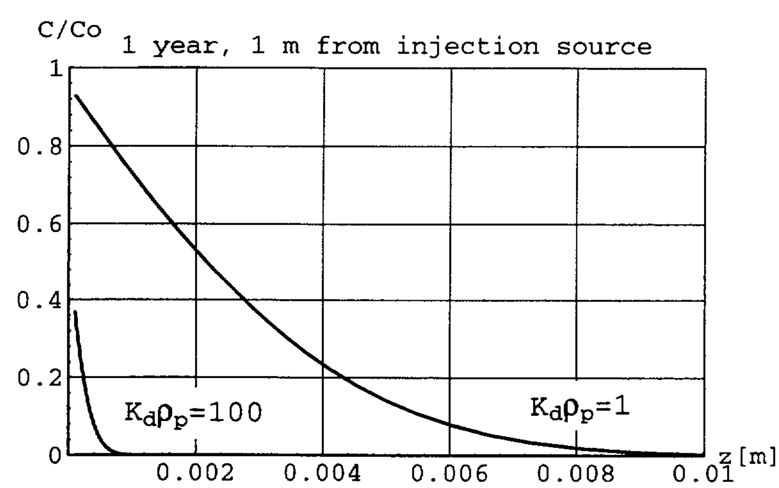

Figure 14. C/C_o into the rock matrix 1 m from the injection hole as function $K_d\rho_p$. Time=1 year and $K_d\rho_p$=1, 100, 10000 m^3/m^3. (W=76 mm, Q=1 ml/h, $D_e=2*10^{-13}$ m^2/s and R_a=1)

Figure 14 shows that the tracer having the lowest sorption capacity, $K_d\rho_p$=1 m^3/m^3, will get a concentration profile into the rock matrix at 1 m that is very similar to the one calculated at a distance of 0.2 m from the injection hole, Figure 12. This is because the concentration in the fracture plane as function of time is almost the same 0.2 m from the source as 1 m from the source. The difference for the tracer having a sorption coefficient of 100 is somewhat larger between the two distances from the source. The curve for $K_d\rho_p$=10000 can not be seen in the figures because of the small penetration depth.

Figure 15 shows the concentration profile into the rock matrix for different diffusivities.

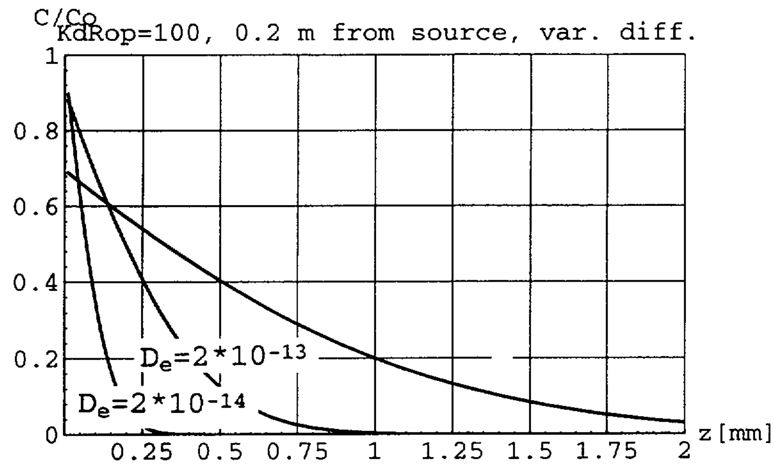

Figure 15. C/C_o into the rock matrix 0.2 m from the injection hole as function D_e. Time=1 year and D_e=2*10^{-14}, 2*10^{-13}, 2*10^{-12} m^2/s. (W=76 mm, Q=1 ml/h, $K_d\rho_p$ =100 m^3/m^3 and R_a=1)

The tracer having the highest diffusivity, 2*10^{-12} m^2/s, will have the largest penetration depth into the rock matrix but also a lower concentration close to the fracture surface because of the very strong interaction with the rock matrix.

3.2 RADIAL FLOW

The conceptual model for the radial flow case is given in Figure 16.

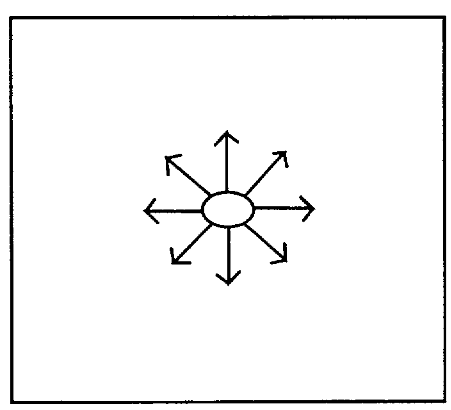

Figure 16. Radial flow. Fracture plane, injection hole and flow directions.

The migration of tracer is from the injection hole radially outward in all directions. This implies that the entire fracture plane is available for tracer migration and that the fracture surfaces do not have any contact points but rather hover over each other. Compared to the case of linear flow, the surface area available for sorption and diffusion into the rock matrix is rapidly increasing with increasing distance from the injection hole. The fracture aperture has been assumed to be constant.

3.2.1 Migration along the fracture

Concentration profiles have been calculated for this radial flow problem using the same set of parameters as in the calculations for linear flow. However, this problem has been solved numerically due to the rather complex situation with radial advection and simultaneous interaction with the rock matrix.

The migration distance along the fracture plane is significantly shorter for radial symmetry compared to the linear case for the same set of parameters because of the larger surface area in contact with the flowing water. Figure 17 illustrates the concentration profiles along the fracture plane for linear and radial flow for a tracer having $K_d\rho_p$=100 m^3/m^3 and a flowrate of 10 ml/h.

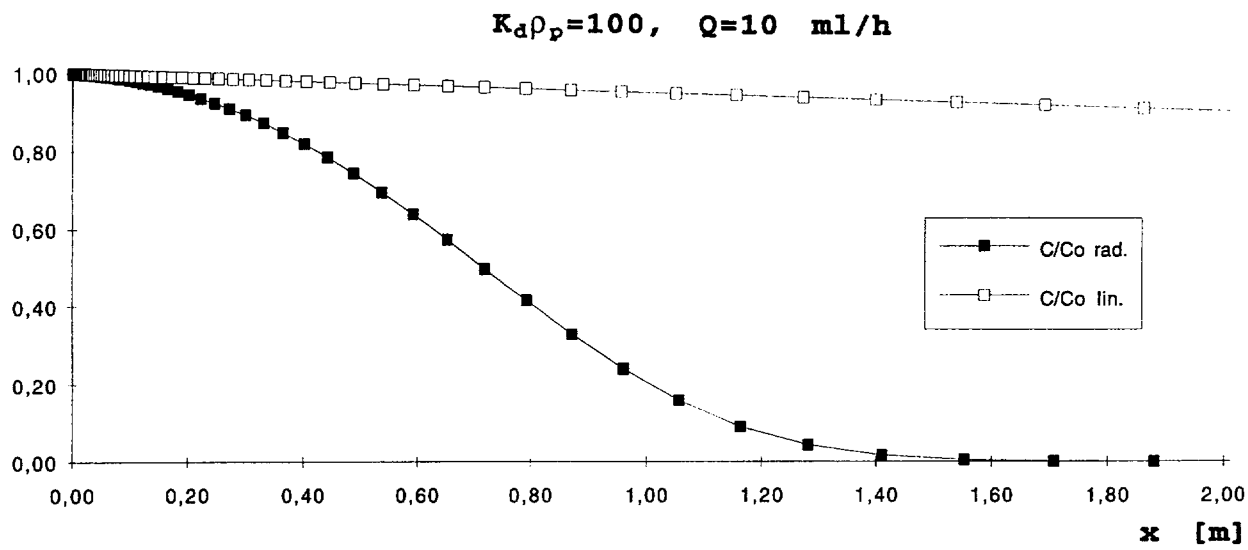

Figure 17. C/C_0 as function of distance from the injection location for linear and radial symmetry. Time=1 year, $K_d\rho_p$=100 m^3/m^3, D_e=2*10^{-13} m^2/s, Q=10 ml/h and R_a=1. W=76 mm for the linear case.

Increasing the time for the experiment from 1 year to 5 years gave a rather significant change in the concentration profiles along the fracture plane in the case of linear flow, see Figures 5 and 6. An increase in time will however only induce a minor change for the case of radial flow, see Figure 18.

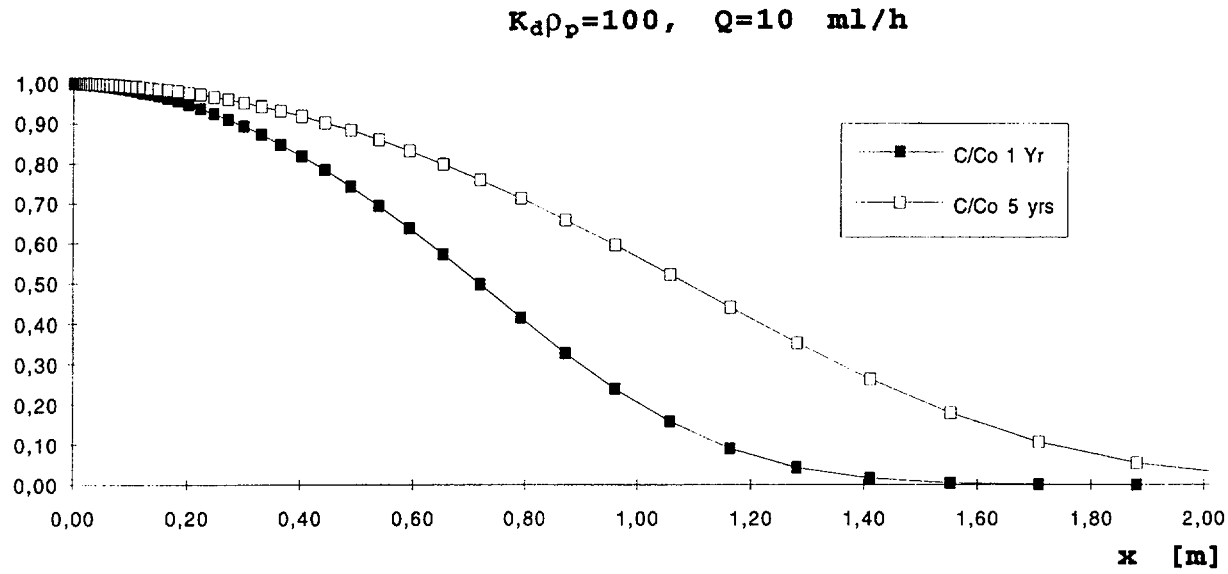

Figure 18. C/C_0 as function of distance from the injection location for 1 and 5 years respectively. $K_d\rho_p$=100 m^3/m^3, D_e=2*10^{-13} m^2/s, Q=10 ml/h and R_a=1.

Figures 5 and 6 illustrated that a tracer having a $K_d\rho_p$ of 10000 m^3/m^3 is expected to migrate 0.5 m along the fracture plane in 1 year and 1 m in 5 years in the case of linear flow. The same tracer would migrate significantly shorter if we have radial flow, see Figure 19. It should also be noted that a flowrate of 10 ml/h has been used for the radial migration, Figure 19, instead of 1 ml/h that was used in the calculations for linear flow. Applying the same flowrate for both cases would give an even larger difference in the concentration profiles.

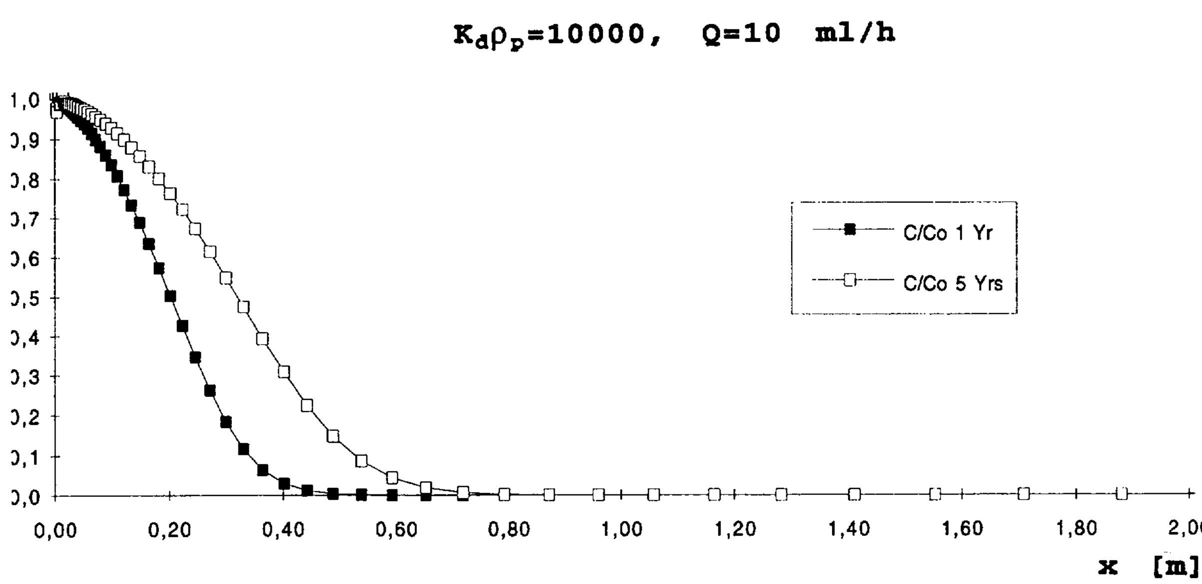

Figure 19. C/C_0 as function of distance from the injection location for 1 and 5 years respectively. $K_d\rho_p$=10000 m^3/m^3, D_e=2*10^{-13} m^2/s, Q=10 ml/h and R_a=1.

3.2.2 Diffusion into the rock matrix

There is no difference in the concentration profile into the rock matrix for the case of linear or radial flow **if** the concentration in the fracture plane as function of time has been the same. But since the tracers will migrate slower in the case of radial flow, the concentration in the fracture plane a distance from the injection section will be lower which implies shorter penetration depths into the rock matrix. The concentration profile into the rock matrix at different distances from the injection section is illustrated in Figure 20.

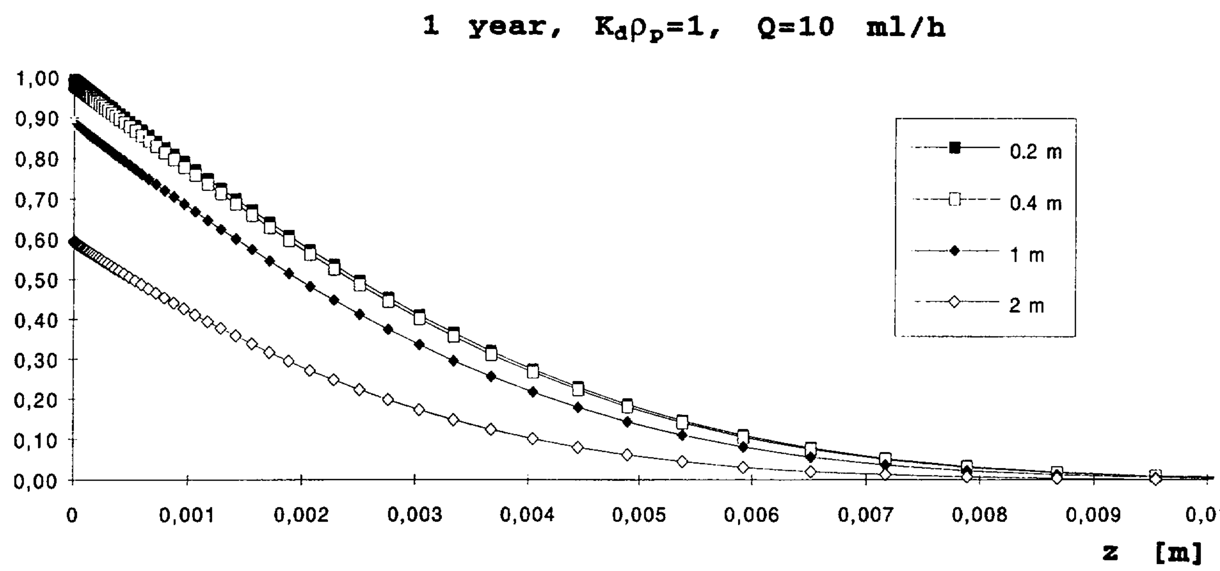

Figure 20. C/C_0 into the rock matrix at different distances from the injection hole. $K_d\rho_p$=1 m^3/m^3, D_e=2*10^{-13} m^2/s, Q=10 ml/h and R_a=1.

3.3 VARIABLE APERTURE DISTRIBUTION MODEL

The 6 m x 6 m fracture plane has in this model been divided into 100x100 squares all having apertures generated from a log-normal distribution. A standard deviation of σ=1 and a correlation length of 0.3 m (corresponding to 5 squares) were used for the aperture generation. The generated fracture plane is illustrated in Figure 21 where a darker color represents a smaller aperture.

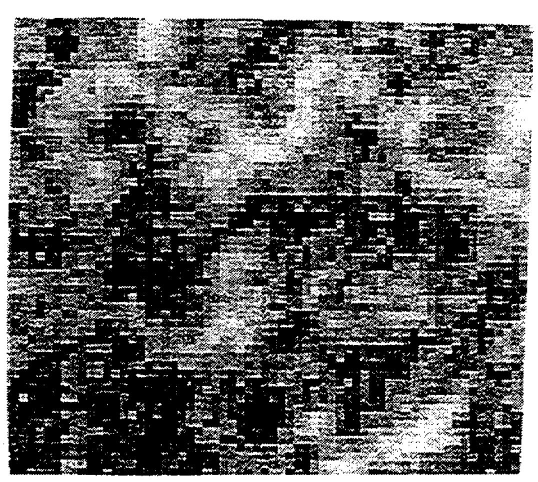

Figure 21. The generated fracture plane in the variable aperture model.

The water flow and the tracer migration will preferentially take place in the channels/flowpaths that will be formed by connected "squares" having large apertures. It should be noted that there will be some water flow and tracer transport in all squares since the apertures have been generated from a logarithmic distribution which prevents the apertures to become 0.

3.3.1 Migration in the fracture plane

The procedure for the tracer migration calculations with this model has been to first calculate the flowrate in all squares in the fracture plane assuming that water is injected in the center of the plane with a given flowrate and that the head is 0 at all four sides. The calculated flowrates and flowpaths are illustrated in Figure 22. A darker color represents a higher flowrate.

Figure 22. Flowrates and flowpaths in the generated fracture plane.

Once the system has been solved regarding the water flow, the tracers can be injected into the fracture plane. This is simulated by injecting particles, one at a time, in the center of the fracture plane and monitoring the migration of each particle as function of time.

Figures 23 and 24 gives the mass flow and concentration for a tracer that is injected for 1 year with a flowrate of 10 ml/h. The tracer has a sorption capacity, $K_d\rho_p$, of 100 m^3/m^3 and an effective diffusivity, D_e, of $2*10^{-13}$ m^2/s. The mass flow, Figure 23, is an illustration of the total number of particles that has passed the square. A darker color represents a larger number of particles. The concentration, Figure 24, illustrates the concentration in each square at the end of the experiment. A darker color represents a higher concentration.

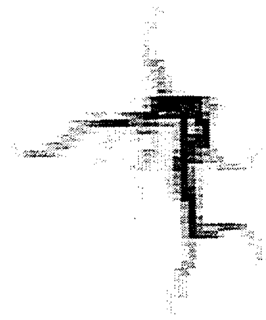

Figure 23. Mass flow in the generated fracture plane. Time=1 year, D_e=2*10^{-13} m^2/s, $K_d\rho_p$=100 m^3/m^3 and Q=10 ml/h.

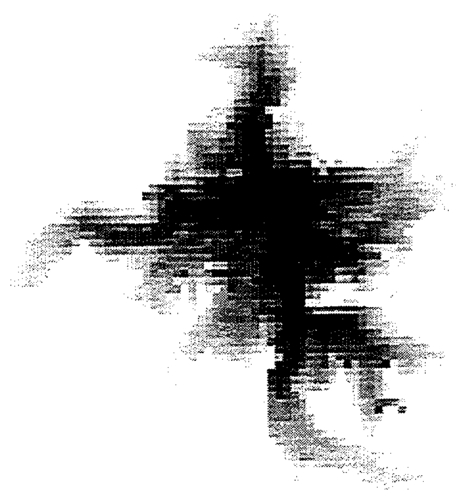

Figure 24. Tracer concentration in the generated fracture plane. Time=1 year, D_e=2*10^{-13} m^2/s, $K_d\rho_p$=100 m^3/m^3 and Q=10 ml/h.

The tracer mass flow, Figure 23, illustrate that the major part of the injected tracer has migrated along the largest water flow paths, see Figure 22. The tracer concentration, Figure 24, is somewhat smeared out since almost all squares close to the injection source will obtain a high concentration independent of aperture. The generated fracture plane has a size of 6x6 m and it can be seen in Figure 24 that the tracers have migrated 2-3 m outward from the injection source. This seems very reasonable since Figure 17 illustrated that a tracer would migrate about 1 m in the case of radial symmetry and several meter in the case of linear symmetry. The variable aperture model is, as expected, somewhere between these two extreme cases.

3.3.2 **Diffusion into the rock matrix**

There has not been any calculations made for diffusion into the rock matrix for this model. However, the concentration profiles into the rock matrix do not depend on the geometrical structure of the flowpaths in the fracture plane but rather on the tracer concentration in the fracture plane as function of time. Points that have the same concentration history in the fracture plane will obtain identical diffusion profiles into the rock matrix independent of the flow geometry.

4 SOME COMMENTS REGARDING THE EVALUATION OF THE EXPERIMENT

The Matrix Diffusion Experiment is expected to provide information regarding

- surface concentrations along the fracture plane
- concentration profiles into the rock matrix

It is furthermore assumed that all tracers that are to be used in Äspö will be carefully tested for their diffusivity and sorption capacity in supporting laboratory experiments.

Both qualitative and quantitative information will be evaluated from the experiment. The obtained qualitative information will be

- to demonstrate that tracers can diffuse into the rock matrix from a natural single fracture
- to demonstrate that sorbing tracers will be retarded due to interaction with the rock matrix
- to illustrate the flow path geometry in a single fracture plane

The first of the three points above will be based on the concentration profiles into the rock matrix and the two other points from the surface mapping.

The parameters that are expected to be evaluated from the experiment are

- the effective diffusivity, D_e
- the sorption coefficient, $K_d\rho_p$
- channel widths, W

The concentration profiles along the fracture plane as well as into the rock matrix will most certainly not be as smooth as those calculated in the previous chapters. Reality will certainly complicate the evaluation. Apart from practical problems as sampling techniques, tracer analysis etc, there will be some physical problems that will be hard (impossible?) to avoid such as

- mixing of waters
- particles, flakes and rough fracture surfaces

Mixing of the injected tracer solution with fresh water a distance away from the injection hole will result in a decrease in the tracer concentration in the fracture plane. This effect might be hard to separate from an increased sorption capacity which also has the effect of decreasing the tracer concentration a distance away from the source. The mixing is schematically illustrated in Figure 25.

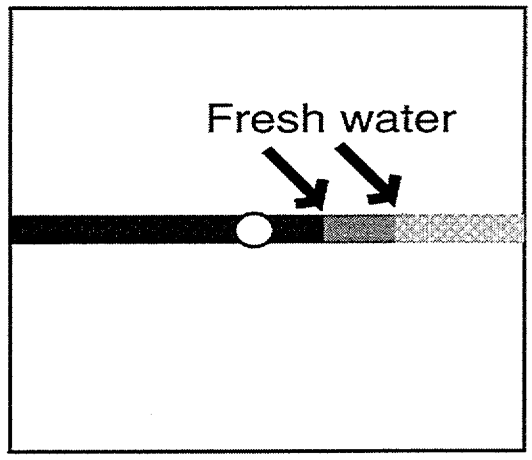

Figure 25. Schematic illustration showing the decrease in concentration due to mixing with fresh waters.

Mixing with fresh waters will affect and complicate the evaluation of the concentration profiles along the fracture plane as well as into the rock matrix. The probability for mixing increases with increasing distance from the injection hole which means that a large part of the evaluation of the experiment is envisioned to be based on the samples taken close to the injection source.

Particles, flakes and rough fracture surfaces will give an contact area for the tracers that will be significantly larger than the "flow wetted surface" that is relevant for radionuclides leaking from a waste repository, see Figure 26.

Figure 12 illustrates that a tracer with $K_d\rho_p$=100 m^3/m^3 will penetrate less then 1 mm into the rock matrix in 1 year. The penetration depth for a tracer having $K_d\rho_p$=10000 m^3/m^3 will be much less. These short penetration depths implies that tracers having these sorption properties will be affected by a surface area much larger than the "flow wetted surface". This area is however not of any importance since even though these irregularities gives an extra surface area, the volume is very small giving that they are not of any significant importance for radionuclides migrating in a fracture for 1000:s of years.

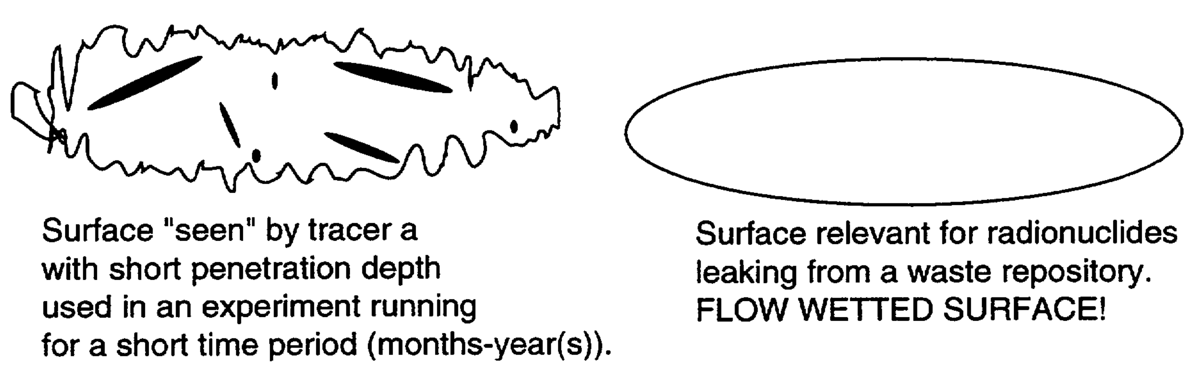

Figure 26. Cross section of a flowpath, channel. Surface "seen" by a tracer with short penetration depth in a "short" term experiment and surface relevant for radionuclides emerging from a leaking repository (="flow wetted surface").

These irregularities will primarily influence the evaluation of the concentration profiles along the fracture plane and into the rock matrix for medium and strong sorbing tracers, $K_d\rho_p$=100 and 10000 m^3/m^3.

4.1 EVALUATION OF PARAMETER VALUES

4.1.1 Sorption coefficient, $K_d\rho_p$

The concentration in the water in the fracture plane will be in equilibrium with the concentration at the surface. Knowing the concentration in the water and measuring the concentration on the fracture surface gives a value of the sorption coefficient, $K_d\rho_p$. The problem is to know the concentration in the water in the fracture plane when the injection is terminated. In the case of mixing of waters, the tracer concentration will be less than the injection concentration for all types of tracers and for all times. The most reliable number on $K_d\rho_p$ will therefore be obtained from samples taken close to the injection source, since the concentration in that water have most likely been equal to the injection concentration, C_o. Supporting laboratory measurements using the fracture surface can also be made.

4.1.2 Effective diffusivity, D_e

The effective diffusivity will be evaluated based on the concentration profiles into the rock matrix. The evaluation of the diffusivity requires that the concentration in the fracture plane as function of time is known. This can be achieved for a weakly sorbing tracer where the concentration in the fracture plane is close to C_o during almost the entire experiment unless there has been mixing of waters. The most reliable number on D_e will therefore be obtained by measuring the concentration profile into the rock matrix close to the injection source.

4.1.3 $K_d\rho_pD_e$

The parameters in this group can be evaluated separately as indicated above, but can as well be evaluated from the concentration profile along the fracture if the flowrate and channel width are known, see Equation 1. This evaluation is based on a decreasing concentration with distance which will eliminate the weakly sorbing tracers. The more strongly sorbing tracers will however be more or less influenced by the particles, flakes and rough surfaces as discussed above. Unfortunately, this makes any evaluation of the group $K_d\rho_p D_e$ very dubious for strongly sorbing species. Supporting laboratory measurements should also be made.

5 SUITABLE TRACERS

The main aim with these scoping calculations has been to suggest suitable sorption properties for the tracers that will be injected. Neglecting the difficulties one can expect in a real fracture, mixing of waters and particles, and instead considering the ideal concentration profiles given in Figure 5 it is obvious that tracers with higher $K_d\rho_p$ than 10000 is meaningless to use if the flow pattern in the fracture plane should be investigated. Tracers with $K_d\rho_p$=10000 or more has to be used if one wants to have the possibility to retrieve all injected tracer. A weakly sorbing tracer, $K_d\rho_p$=1, should have almost the same concentration in the channel independent of the distance, up to 2 m, from the injection source. This type of tracer could be very useful to inject since it could give information regarding mixing of waters.

When evaluating the effective diffusivity, it is desirable from a practical point of view that the tracers have migrated some millimeters or preferably a centimeter into the rock matrix. This "suitable distance" will however be dependent on the sampling technique, tracer analysis, surface roughness etc. Considering the concentration profile into the rock matrix, Figure 12, it is obvious that it is meaningless to try to evaluate the diffusivity for a tracer with $K_d\rho_p$=10000. A tracer with $K_d\rho_p$=100 might work, while a tracer with $K_d\rho_p$=1 should be perfect.

The above discussion about suitable tracers is based on the results from the calculations using linear flow. If the other extreme flow situation, radial flow, is considered it seems useless to inject tracers with higher $K_d\rho_p$ than 100 to investigate the flow pattern in the fracture plane, see Figure 17. On the other hand, the concentration profiles into the rock matrix in the vicinity of the injection source are not so dependent on the flow geometry. The same $K_d\rho_p$ range as given above for linear flow, $K_d\rho_p$=1-100, should therefore also be suitable for diffusion into the rock matrix in the case of radial flow.

5.1 SUMMING UP

Several different tracers should be injected simultaneously. These tracers should have $K_d\rho_p$ values ranging from, at least, 1 up to 10000 m^3/m^3. The majority of these tracers should however have $K_d\rho_p$ values in the range 1 to 100 since tracers having higher $K_d\rho_p$ values can not be used for determination of diffusion effects.

It has furthermore been assumed that the tracers have linear adsorption isotherms. Non-linear sorption would complicate the evaluation significantly.

6 PRACTICAL CONSIDERATIONS

6.1 TYPE OF INJECTION

There are a number of alternative ways to inject the tracers into the fracture. Those considered by us are given below

Constant flowrate	Used in the Tracer Migration Experiment in the Validation drift in Stripa
Constant pressure	Used in the In-situ Diffusion Experiment, the 2-D experiment, the 3-D experiment and the Channeling experiments in Stripa.
Dilution probe	
Soluble compound	

Injections using constant flowrate or constant pressure have been applied in several tracer experiments and will not be described here. It should be noted that both these methods are based on that the tracers are injected with a pressure higher than the natural pressure. The actual injection pressure will depend on the fracture and the injection flowrate, but is envisioned to be in the order of one or a few meters head.

The tracers will be injected without any overpressure in both the dilution probe method and the soluble compound method. The dilution probe injection is based on that a section of the borehole is isolated and filled with a tracer solution that will be "injected" into the fracture plane by the natural flowing water passing through the compartment. It is possible to measure the dilution of tracers in the injection compartment when using the dilution probe. This information can be used for an calculation of the mass flowrate as function of time.

The soluble compound method is similar to the dilution probe method. In this case, the compartment is filled with a soluble compound containing the tracers that should be injected. The natural flowing water passing through the compartment will then transport these tracers into the fracture plane. The remaining tracer mass can be measured once the experiment is terminated giving the average mass flowrate during the experimental time, but not the mass flowrate as function of time. Another severe drawback with this type of injection is that neither the concentration of tracers entering the fracture plane nor the flowrate will be known. The flowrate through the compartment can however be measured before and after the experiment, but the flowrate as function of time during the experiment will be unknown. Table 1 gives a subjective compilation of advantages using different injection methods. An X indicate an advantage for that method.

Table 1. Compilation of advantages with the different injection methods. An X indicate an advantage.

	Q=const	p=const	Dilution	Soluble compound
Injection conc known	X	X	(X)	
Q=f(t) known	X	X	X	
Q constant	X			
Possible to change Q	X	X		
Change tracers	X	X	X	
Pre-work, equip		X		X
Pre-work, tracers	X	X	X	
Simple equipment		X		X
Without daily checks		(X)		X
Mass balance for the tracers	X	X	X	(X)
Sorption on equip				X
Technique used earlier in this context	X	X	X	
No overpressure when injecting			X	X

The methods with constant pressure and constant flowrate are both well known from earlier experiments. Constant pressure has some practical advantages, but the constant flowrate is to be preferred when evaluating the experiments. The constant flowrate method is actually the best of the four methods when evaluating the experiments.

The dilution method and the method using a soluble compound are both very attractive since no overpressure has to be used for the tracer injection. This is an important factor since the experiment aims at demonstrating that diffusion and sorption exists under natural conditions. Especially the method with the soluble compound is attractive because of the simplicity.

The short discussion above indicate that the two most attractive types of injection is the constant flowrate method and the soluble compound method. We prefer the method with the constant flowrate because then we will know the injection concentration, flowrate as function of time, mass flowrate. This will not be the case if the tracers are injected using the soluble compound method. Furthermore, applying a small overpressure will increase the chance that the concentration in the vicinity of the source has been equal to the injection concentration, C_o, which is the basis when evaluating the numbers on $K_d\rho_p$ and D_e. The major drawback with the constant flowrate method is that the tracers have to be injected with a overpressure.

It could be argued that this overpressure will cause advection from the fracture plane and into the rock matrix and that this advective transport could be very hard to separate from the diffusive transport. However, if it is possible for tracers to migrate into the rock matrix by advection then there must exist a connected pore system that can be utilized for diffusion.

A rough estimate of the impact of an applied overpressure shows that the penetration depth into the rock matrix due to flow will be very small. Consider the case where

$K_p=10^{-13}$ m/s	"Typical" hydraulic conductivity in the rock matrix in Stripa.
i=1 m/m	The tracer injection is envisioned to be carried out using an overpressure of one or a few meters. This overpressure will prevail in the vicinity of the injection hole and rapidly decrease with distance from the hole. A pressure gradient through the rock matrix will be established if other fractures in the vicinity of "our" fracture has a lower pressure. It is hard to estimate this pressure gradient, but let us assume a value of 1 m/m which is not unreasonable.
$\varepsilon_p=2$ %	The porosity in the rock matrix has been found to be in the order of 0.5 % in Stripa. The porosity close to fracture surfaces has however been found to be significantly higher giving that a value of 2 % is reasonable.

This gives a velocity through the rock matrix of about 1.5 mm/year due to advection. The calculation has been based on somewhat conservative numbers for the Stripa granite. The hydraulic conductivity and the porosity is, today, unknown for the rock at the location where the experiment will take place in Äspö, but might be somewhat higher which would give a higher velocity than 1.5 mm/year through the rock matrix. However, these 1.5 mm/year is the velocity that the water, or a non-sorbing tracer, will have into the rock matrix due to advection. The penetration depth into the rock matrix for a non-sorbing tracer, see Equation 3 and Figure 11, will for the first year be about 60 mm for an effective diffusivity of $D_e=2*10^{-12}$ m^2/s (this is another "typical" value from Stripa) and a porosity of $\varepsilon_p=2$ %. This gives that the migration into the rock matrix will be determined by diffusion even though one would inject the tracer with a moderate overpressure.

The migration due to diffusion as well as advection will be slower for a sorbing tracer. The migration rate into the matrix due to advection will be indirectly proportional to the sorption coefficient ($U_{tracer} \propto U_{water} / (K_d\rho_p)_{tracer}$) while the penetration depth due to diffusion will depend on the square root of the sorption coefficient, see Equation 3. The sorption coefficient, $K_d\rho_p$, for a non-sorbing tracer equals the porosity, ε_p, which in our case has been assumed to be 2 %. Consider a tracer with 100 times larger sorption coefficient, $K_d\rho_p=2$, which in our context is a weakly sorbing tracer. The migration rate into the matrix due to advection will then be 100 times lower than for our non-sorbing tracer, but the penetration depth will just be 10 times lower. Applying the values that were obtained for the non-sorbing tracer then gives that this tracer with $K_d\rho_p=2$ will migrate 0.015 mm into the matrix due to advection and 6 mm due to diffusion. The impact of advection due to the applied pressure is therefore negligible. The difference between diffusive and advective transport will increase as the sorption coefficient increases.

The above discussion indicate that the effect of advective transport through the rock matrix due to an applied overpressure will be negligible especially for a sorbing tracer. The migration into the rock matrix will be dominated by diffusion even if the pressure gradient should be increased with a factor 100 giving a pressure gradient of 100 m/m. Advective migration into the rock matrix is therefore not likely to complicate the evaluation of the effective diffusivities.

6.2 SAMPLING HOLES IN THE VICINITY OF THE INJECTION HOLE

The experimental layout, see Figure 2, has been illustrated with an injection hole intersecting the fracture plane, but without sampling holes in the vicinity of the injection hole. It is of course possible to drill a number of sampling holes that can be used for pressure monitoring and/or tracer sampling. These sampling holes can be located within the area that will be excavated or, to reduce the disturbance on the flowfield, outside the area to be excavated. The alternatives that has been considered are illustrated in Figure 27.

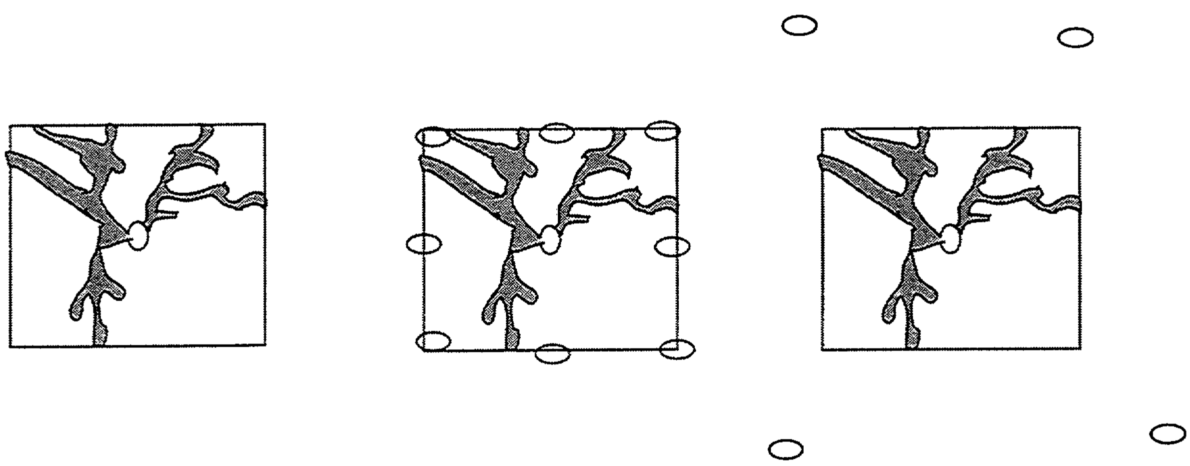

Figure 27. Sampling holes in the vicinity of the injection hole.

Table 2 gives a subjective compilation of advantages with the different layouts. An X indicate an advantage for that layout.

Table 2. Compilation of advantages with different experimental layouts. X indicates an advantage.

	None	Within	Outside
Simple equipment	X		
Without daily checks	X		
Information regarding mixing		(X)	
Evaluation of $K_d\rho_p D_e$		(X)	
Results while running the experiment		X	X
Undisturbed flowfield	X		X

Sampling holes outside the area that will be excavated do not seem to be of any major advantage. The tracer breakthrough curves one could obtain in the sampling hole would probably be of minor importance for the evaluation of the experiment. The alternative with sampling holes within the area that will be excavated is in some ways more attractive. The breakthrough curves in the sampling holes might aid in the understanding of mixing of waters in the fracture plane and in the evaluation of $K_d\rho_p D_e$. However, waters from different flowpaths will certainly mix in the sampling hole giving that the concentration in the flowpath(s) of interest will probably remain unknown. Furthermore, the evaluation of $K_d\rho_p D_e$ will probably anyway be uncertain because of flakes etc as discussed above. Finally, a number of sampling holes 0.5-1 m from the injection hole would certainly disturb the natural flowfield.

The short discussion above illustrates that we at present do not think that it is motivated to have sampling holes in the fracture plane. The extra information that could be obtained do not motivate the extra cost connected with drillings, equipment, sampling etc. Giving a fixed budget it seems better to avoid the sampling holes and instead investigate some extra fractures. There are however some advantages with sampling holes in the fracture plane so the discussion will certainly continue and we might also change our opinion.

7 CONCLUSIONS

A mixture of tracers having different sorption capacities should be injected simultaneously. These tracers should have sorption capacities, $K_d\rho_p$ values, ranging from, at least, 1 m^3/m^3 to 10000 m^3/m^3. Most of the injected tracers should however have sorption capacities in the range $K_d\rho_p$=1 to 100 m^3/m^3, since the concentration profile into the rock matrix can not be determined for stronger sorbing tracers. The tracers should have linear sorption isotherms.

The report also includes some practical considerations. Different injection methods and the need for sampling holes in the vicinity of the injection hole has been discussed. We prefer if the tracers were injected using a constant flowrate method. This has significant advantages compared to the other considered methods when the experiment is evaluated. Furthermore, we do not at present think that it is motivated to have sampling holes in the fracture plane. The cost associated with these holes should instead be used to investigate more fractures. There are however some advantages with sampling holes in the fracture plane so the discussion will certainly continue and we might also change our opinion.

8 REFERENCES

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Neretnieks, I., Diffusion in the Rock Matrix – An important Factor in Radionuclide Retardation?, SKBF Technical Report 79-19, Stockholm 1979.

APPENDIX

NUMERICAL MODELLING,
DOCUMENTATION OF HARDWARE AND SOFTWARE

Documentation of numerical simulation by Hans Widén (Name) 1993-11-17 (Date)

OBJECT
SKB purchase order no: 8-10-160
Title of SKB purchase order: Äspölaboratoriet-försök i detaljskala
Author of report: Birgersson et al. Company: Kemakta
Operator of software: Hans Widén Company: Kemakta

Computer
Unitron 486

Software
Operative system: DOS 5.0

Code Name: TRUMP, which is a computer program for transient and steady state temperature distribution in multi-dimensional systems. It is often used for calculation of flow and diffusion of dissolved species. The program was originally developed by A.L. Edwards in 1972. The version used have been modified by Kemakta at several occasions up to 1992 to facilitate easier data handling.

Program language:FORTRAN

Compiler: Microway NDP FORTRAN

Input files:
S2\prj:2011\TRUMP\CYLINDER\INPUT\CYLA1.INP
S2\prj:2011\TRUMP\CYLINDER\INPUT\CYLA2.INP
S2\prj:2011\TRUMP\CYLINDER\INPUT\CYLA3.INP
S2\prj:2011\TRUMP\CYLINDER\INPUT\CYLB1.INP
S2\prj:2011\TRUMP\CYLINDER\INPUT\CYLB2.INP
S2\prj:2011\TRUMP\CYLINDER\INPUT\CYLB3.INP

Output files:
S2\prj:2011\TRUMP\CYLINDER\INPUT\CYLA1.CNC
S2\prj:2011\TRUMP\CYLINDER\INPUT\CYLA2.CNC
S2\prj:2011\TRUMP\CYLINDER\INPUT\CYLA3.CNC
S2\prj:2011\TRUMP\CYLINDER\INPUT\CYLB1.CNC[1]
S2\prj:2011\TRUMP\CYLINDER\INPUT\CYLB2.CNC
S2\prj:2011\TRUMP\CYLINDER\INPUT\CYLB3.CNC

[1] No useful output from this simulation.

List of ICR Reports 1993

ICR 93-01
Flowmeter measurement in borehole KAS 16
P Rouhiainen
June 1993
Supported by TVO, Finland

ICR 93-02
Development of ROCK-CAD model for Äspö Hard Rock Laboratory site
Pauli Saksa, Juha Lindh,
Eero Heikkinen
Fintact Ky, Helsinki, Finland
December 1993
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CM Gruppen AB, Bromma 1994